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AMS and ICP-MS for determination of long-lived environmental radionuclides

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ABSTRACTS

Ninth International Conference on
Methods and Applications of
Radioanalytical Chemistry

MARC IX

March 25-30, 2012



Log: 105. **A BLUEPRINT FOR RADIOANALYTICAL REFERENCE MATERIALS.** Kenneth G.W. Inn, NIST.

Reference materials are fundamental tools used by low-level environmental measurements laboratories to establish and evaluate analytical methods, test measurement capabilities, compare analytical results over time and between programs, and establish legal confidence in measurement results. Over the past decades low-level environmental reference materials have been developed mostly for environmental monitoring and geosciences. However, more recently radionuclide reference materials have been developed for other communities such as occupational safety, ocean studies, food safety, nuclear power decommissioning, and environmental restoration. Currently, new national and international priorities has resulted in demands for improved, broader, and new environmental reference materials for urban remediation and recovery, radiobioassay, emergency consequence management, and nuclear forensics, attribution, non-proliferation and environmental safeguards. With the application of new generation mass spectrometry to complement traditional radioactivity counting the new reference materials will need to be certified on both the radioactivity and mass basis. The reference material producers will need to be able to keep up with the increasing diversity of needs, and the program directors and measurement community need to communicate their needs early to the producers and support the efforts. It is important to keep in mind that developing good reference materials is a complex process that requires careful metrology and may take a considerable amount of time with a commiserate expense. In this current financial climate, there just aren't enough resources for each reference material producer to serve all of their potential customers. There is need for communication, coordination and funding among all programs and countries to move the reference material effort into the future.

Log: 106. **THE URGENT REQUIREMENT FOR NEW RADIOANALYTICAL CERTIFIED REFERENCE MATERIALS FOR NUCLEAR SAFEGUARDS, FORENSICS, AND CONSEQUENCE MANAGEMENT.** Kenneth G.W. Inn, NIST.

A multi-agency workshop was held from 25–27 August 2009, at the National Institute of Standards and Technology (NIST), to identify and prioritize the development of radioanalytical Certified Reference Materials (CRMs, generally provided by National Metrology Institutes; Standard Reference Materials, a CRM issued by NIST) for field and laboratory nuclear measurement methods to be used to assess the consequences of a domestic or international nuclear event. Without these CRMs, policy makers concerned with detecting proliferation and trafficking of nuclear materials, attribution and retribution following a nuclear event, and public health consequences of a nuclear event would have difficulty making decisions based on analytical data that would stand up to scientific, public, and judicial scrutiny. The workshop concentrated on three areas: post-incident Improvised Nuclear Device (IND) nuclear forensics, safeguard materials characterization, and consequence management for an IND or a Radiological Dispersion Device detonation scenario. The workshop identified specific CRM requirements to fulfill the needs for these three measurement communities. Of highest priority are: (1) isotope dilution mass spectrometry standards, specifically ^{233}U , ^{236}gNp , ^{244}Pu , and ^{243}Am , used for quantitative analysis of the respective elements that are in critically short supply and in urgent need of replenishment and certification; (2) CRMs that are urgently needed for post-detonation debris analysis of actinides and fission fragments, and (3) CRMs used for destructive and nondestructive analyses for safeguards measurements, and radioisotopes of interest in environmental matrices.

Log: 111. **PLUTONIUM AND NEODYMIUM REACTIONS WITH BRINE AT ELEVATED TEMPERATURES.** Jeff Terry Department of Physics Illinois Institute of Technology.

The problem of the disposal of high level waste from commercial nuclear reactors must be solved. The recent incident at the Fukushima Dai-ichi nuclear reactor site clearly shows that spent nuclear fuel cannot be stored on-site indefinitely. One possible solution is to dispose of the

spent fuel in ancient salt beds. This requires an understanding of the actinide and fission product chemistry at elevated temperature and pressure. The chemistry of plutonium and neodymium with salt brines consistent with the Salado formation in southern New Mexico has been studied with x-ray absorption, UV/Vis spectroscopy, and GC/MS as a function of temperature in order to determine if this formation would be a suitable site for high level commercial waste disposal. To date the chemistry favors the long term reduction of plutonium. This typically leads to less migration in the environment. The results of the chemistry at temperatures up to 100 °C will be presented.

Log: 112. **IMPROVING NEUTRON ACTIVATION ANALYSIS ACCURACY FOR THE MEASUREMENT OF GOLD IN THE CHARACTERIZATION OF HETEROGENEOUS CATALYSTS USING A TRIGA REACTOR.** Siaka Yusuf; Melinda Krahenbuhl; Bryan Haskins. The Dow Chemical Company.

To measure the gold content of a catalyst accurately, Neutron Activation Analysis (NAA) is one of the methods of choice. NAA is generally a preferred method, for those who have access to a neutron source, for the measurement of the elemental contents of such heterogeneous catalysts because: 1. NAA requires minimal sample preparation. 2. NAA provides consistent and accurate results. 3. In most cases results are obtained much quicker than competing methods. NAA is also being used as a referee for the other elemental techniques when results do not fall within expected statistical uncertainties. However, at very high gold concentrations, applying NAA to determine the gold in a heterogeneous catalyst is more challenging than a routine NAA procedure. On the one hand, the neutron absorption cross section for gold is very high which easily results in significant self-shielding related errors. On the other hand, gold exhibits low energy resonance neutron absorptions. In this particular application the self-shielding minimization effort was handled more rigorously than the classic simple suppression of neutron flux within a specimen. A non-routine approach was used because: 1) for most applications, high accuracy, better than 3% relative, is desired, 2) the low energy resonances of gold make its neutron reaction rate much more complex and 3) the TRIGA reactor flux profile used in this study contains both thermal and significant epithermal neutron fluxes. Accuracy and precision for gold analysis in heterogeneous catalysts using this new approach is expected to improve from 15% to better than 3% relative uncertainty. This has been accomplished through a rigorous assessment of the observed effects of low energy resonance on the neutron flux spectral-shape within the sample and designing an experiment to minimize the effects.

Log: 113. **UV/VIS OBSERVATION OF NP REDOX REACTIONS IN IRRADIATED SOLUTION.** Bruce J. Mincher, INL; Martin Precek and Alena Paulenova, Oregon State University, Stephen P. Mezyk, Cal State Long Beach.

The redox chemistry of neptunium is problematic in fuel cycle separations because Np(V) is inextractable and Np(VI) is extractable in most solvent extraction systems. However, regardless of the initial distribution of neptunium oxidation states, an equilibrium distribution containing both valences is achieved upon irradiation in aqueous nitric acid. This is due to a complicated series of reactions involving both radiolytically-produced free radicals and nitrous acid. Here we have used UV/Vis spectroscopy to follow the oxidation state changes during irradiation with a gamma-ray irradiator, using starting solutions of predominantly Np(V) or Np(VI). In either case, oxidation of Np(V) to Np(VI) occurred initially, until adequate HNO₂ grew in to solution to favor Np(V). No Np(IV) was found at any time. A mechanism for these reactions is proposed that depends on the concentration of HNO₃ and dissolved oxygen in the irradiated solution.

Log: 114. **APPLICATION OF TWO EXECUTABLE COMPUTER CODES FOR CONFIDENCE INTERVALS, DECISION LEVELS AND DETECTION LIMITS WHEN THE SAMPLE IS COUNTED AN INTEGER TIMES EQUAL TO OR GREATER THAN THE BLANK.** Potter, WE(1); Strzelczyk, J(2). (1) Consultant, (2) University of Colorado Hospital.

Frequently the sample contribution to the gross count is well approximated by a discrete Poisson distribution; this paper makes this assumption. The ratio of the sample count time to the blank count time is taken to be an integer IRR, and the net count is denoted by OC. The expected blank count in the sample count time, B, is assumed either known or well known. In the well known case, the net count is taken to be the difference between the gross count, a random variable, and a constant equal to the well known blank count; otherwise the net count is the difference between two random variables, the gross count and the product of IRR with a Poisson distributed blank count. In the later case the probability density function for OC is given in the proceedings of the 2009 Midyear Meeting of the Health Physics Society. Confidence intervals are determined by mimicking a standard approach in discrete probability. A C++ computer code computes confidence intervals and detection limits for the expected net count. A separate code computes decision levels. Executable versions of both codes have been developed which are expected to run on most Windows based computers.

Log: 115. **MULTI-COLLECTOR ISOTOPE RATIO MASS SPECTROMETER FOR ULTRA-TRACE ACTINIDE DETECTION.** Watrous, MG ; Appelhans, AD; Olson, JE. Idaho National Laboratory.

This paper describes the operational testing of a thermal ionization isotope ratio magnetic sector mass spectrometer that utilizes seven full-sized discrete dynode electron multipliers operating simultaneously. The instrument includes an ion dispersion lens that enables the mass dispersed individual isotope beams to be separated sufficiently to allow a full-sized discrete dynode pulse counting multiplier to be used to measure each isotope beam. The performance of the instrument was measured using SRM 996 (^{244}Pu spike) at loadings of 2.4 and 12 fg on resin beads and with SRM 4350B Columbia River Sediment samples. The measured limit of detection (3σ) for ^{240}Pu was 3.4 attograms for SRM 996; The limit of quantitation was 11.2 attograms.

Log: 116. **INDOOR RADON SURVEY IN WORKPLACES USING NUCLEAR TRACKS METHODOLOGY.** Arturo Angeles (1), Guillermo Espinosa (2); (1) Instituto Nacional de Investigaciones Nucleares, and graduate student of the UAEMex. (2) Instituto de Física, Universidad Nacional Autónoma de México, Circuito de la Investigación Científica, Ciudad Universitaria. 04520, México, D.F.

In this paper, the preliminary results of the indoor radon concentration measurements in some Mexico Cities are shown. As it is known, the indoor radon concentrations can be represent a health problem. In several countries actually exist regulations which restrict or suggest "acceptable" radon air concentrations in workplaces. Because of the fact that, in Mexico there are no regulations in this issue to reach this objective, we've been evaluating radon air concentrations in some mexican workplaces as kindergartens, primary and high schools, research labs and financial offices; by the nuclear track method. With the obtained radon concentrations values the equivalent dose is calculated regard the places and stay work times in such cases. Also history radon concentration values in other mexican places published in some papers has been considered to enrich the values quantity. By the knowledge of equivalent dose values the health damage likelihood is regard to suggest a restrict value in that work conditions. It's important to stand out the fact of the first time this kind of measurements to place the base for the mexican regulation in this matter. The author wishes to thank J.I. Golzarri for his technical help. This work was partially supported by PAPIIT-DGAPA-UNAM Project 1N101910.

Log: 117. **STUDY OF INDOOR RADON IN GREENHOUSES IN MEXICO CITY, MEXICO.** Espinosa, G., Chavarria, A.C., Golzarri, J.I. Instituto de Fisica, Universidad Nacional Autonoma

de Mexico; Circuito de la Investigación Científica, Ciudad Universitaria, 04510, Mexico, D.F., Mexico.

As enclosed spaces where soil, the main source of radon, is exposed, greenhouses have potentially high radon (^{222}Rn) concentrations. Greenhouses are frequented by visitors and are also workplaces: the study of radon concentrations in greenhouses is thus a relevant concern for public health authorities. For this study the radon concentrations in 10 greenhouses at different locations within Mexico City were measured using Nuclear Track Methodology (NTM). The used detectors for the study consisted of the well-known closed-end cup device with CR-39 Lantrack® as detector material. The measurements were carried out over a period of one year, divided into four three-month sub-periods. The minimum and maximum radon concentrations found were 15 and 133 Bq/m³ respectively; the annual mean was 62 Bq/m³. Significant seasonal variation was also observed. Using an equilibrium factor of 0.4, the effective dose from ^{222}Rn and its progenies was calculated to be 467.3 nSv/h. This corresponds to an annual dose rate of 467.3 µSv/y (0.039 WLM/a) for a worker spending 4 h/day, 5 days a week, 50 weeks a year inside the greenhouse. For a visitor spending 12 hours a year inside the greenhouse the annual dose rate is 5.607 µSv/y. The study of indoor radon concentrations in buildings such as greenhouses, which are both workplaces and open to visitors, is an important public health consideration.

Log: 118. **INDOOR RADON CONCENTRATION LEVELS IN MEXICAN CAVES, USING NUCLEAR TRACK METHODOLOGY, AND THE RELATIONSHIP WITH LIVING HABITS OF THE BATS..** Morales-Malacara, J.(1), Vega-Orihuela, E.(1), Golzarri, J.I.(2), Espinosa, G.(2); (1) Facultad de Ciencias, UNAM; (2) Instituto de Física, Universidad Nacional Autónoma de México. Circuito de la Investigación Científica, Ciudad Universitaria, 04510, México, D.F.

This work presents the study of radon levels in 4 different caves in the regions of Querétaro: "Cave in the cliffs" and "Cave of the Jew"; and Puebla: "Coyozochico" and "Karmidas", in Mexico. The measurements were made using the passive close-end-cup system, developed at the Physics Institute of the UNAM, using CR-39 (Lantrack®) as sensible material, and following protocols established for the measurements of indoor radon by the dosimetry applications project. The distribution of radon in each of the caves turned out to be very interesting, taking values between 300 Bq m⁻³ to 1,284 Bq m⁻³. Found in Karmidas cave indoor radon concentration levels exceeding 60,000 Bq m⁻³, being this a relevant case for analysis. In addition from the radiological point of view, was found a relationship between the concentrations of radon and the cave habitat where take refuge populations of bats. As that based on the results obtained in particular on the crags, areas inhabited by bats are those who have low levels of radon between 60 and 350 Bq m⁻³ approximately, since this is a surprise, longer than it could infer the possible sensitivity of these small mammals to high concentrations of radon which makes them take refuge in areas of low concentration, suggesting a line of interesting study as a potential biomarker of risk areas to radon.

Log: 119. **AIRBORNE GAMMA-RAY EMITTERS FROM FUKUSHIMA DETECTED IN NEW YORK STATE.** Kitto, ME (1,2); Menia, TA (1); Haines, DK (1); Beach, SE (1); Bradt, CJ (1); Syed, U-F (1); Semkow, TM (1,2); Bari, A (1); Khan, AJ (1). (1) New York State Department of Health, (2) State University of New York at Albany.

A network of air-sampling stations that operates continuously as part of New York State's environmental surveillance program collected radionuclides emitted as a result of the Fukushima nuclear accident. Samples were collected, typically for 7 days each, by drawing ~600 m³ of air through a 5.1-cm diam particulate-collecting filter followed in series by a canister containing activated charcoal. Gamma-ray spectroscopy was used to confirm the detection of I-131, Cs-137, and Cs-134 in the particulate phase at all sites, with maximum concentrations near 5, 3, and 1 mBq/m³, respectively. Gas-phase I-131, collected on the activated charcoal, exhibited a maximum concentration of 3 mBq/m³ at the sites. Assessments suggest that there were minimal

health impacts from the airborne radionuclides as the activities contributed an insignificant amount to the annual dose.

Log: 120. **RADIONUCLIDE AND CHEMICAL HAZARDS OF A RADIUM ORE REVIGATOR.**
Kitto, ME (1,2); Judd, CD (1); Bradt, CJ (1); Torres, MA (1).

A study to characterize the radionuclide and chemical components in a radium-ore revigator has been completed. Measured activities of dissolved Rn-222, Ra-226, Ra-228, and U isotopes were determined in the water using radioanalytical techniques. Trace-metal concentrations, determined using inductively coupled plasma optical emission spectrometry (ICPOES) and mass spectrometry (ICPMS), increased in the water over time. The contribution, due to radon gas emanation from the revigator, to the airborne radon (Rn-222) level in a room were evaluated. The effective doses resulting from drinking the revigator water were estimated for the radionuclides.

Log: 121. **MODELISED TRITIUM FREE WATER VS EXPERIMENTATION: A KING OF ORGANICALLY BOUNDED CONSIDERATION.** Vichot, L (1); Patryl, L (2); Fromm, M (3); Badot, PM (4). (1) CEA, VALDUC, F-21120 Is-sur-Tille, France, (2) CEA, DIF, F9100 Bruyère le ch tel, France, (3) UFR Sciences et Techniques, F-25030 Besan on, France, (4) CNRS-Universit  de Franche Comt , F-25030 Besan on, France..

Tritium is a naturally, but also an industrially, radioactive isotope of hydrogen. Because Organically Bounded Tritium (OBT) is synthesized by vegetable from tritium free water, mainly through photosynthesis process, it's firstly needed to determine tritium concentration in plant's water. We have improved available models developed by CEA in regard to determined and controlled experimentations on suitable vegetables. In this article considerate equations for modelling are developed, and condition for experimentations are strictly defined and controlled. Experimental results show that tritium concentration in air is twice high than this of concentration measured in tissue free water of plant. This can be explained if tritium integration by root is the main process, even if tritium intake by leaves is far away to be negligible: as a matter of discussion equations will be proposed for publication. Considering OBT, since health effect and Relative Biological effect are always discussed, several experimentations have been done, as a function of available energy to the plant and vegetable growth stage. It is interesting to note that the same conclusion can be obtained for different conditions, and regardless of the season taken into account. Considering chronic releases, simple relations between air moisture and OBT can be done. As a conclusion, it appears that simple models can describe tritium transfer in different compartments of the biosphere. This is relevant for tritium migration and dose assessment regarding tritium release and production.

Log: 123. **OVERVIEW OF NEW URANIUM ISOTOPIC REFERENCE MATERIALS AT IRMM.**
S Richter, R Eykens, U Jacobsson, H Kuehn, Y Aregbe, A Verbruggen, R Wellum, Institute for Reference Materials and Measurements, JRC-EU, Retieseweg 111, 2440 Geel, Belgium.

For many geological and radioanalytical applications, isotope ratio measurements play a significant role. In order to validate mass spectrometric measurement procedures and to calibrate detector systems, suitable isotope reference materials are needed. For many years the European Commission's Institute for Reference Materials and Measurements - IRMM - is one of the main providers of certified uranium and plutonium isotopic reference materials. These materials are applied in nuclear fuel cycle measurements, in safeguards verification and in environmental studies. They are particularly also used for geological and radioanalytical applications. This poster gives an overview of isotope reference materials for uranium prepared and certified at IRMM. These materials are synthetic isotope reference materials prepared based on proven methods of purifying and mixing highly enriched oxides. Firstly, the double spike IRMM-3636 with a $^{233}\text{U}/^{236}\text{U}$ ratio of 1:1 was prepared which allows internal mass fractionation correction for high precision $^{235}\text{U}/^{238}\text{U}$ ratio measurements. The ^{234}U abundance of this double spike material is low enough to allow an accurate and precise correction of $^{234}\text{U}/^{238}\text{U}$ ratios, even for measurements of

close to equilibrium uranium samples. The double spike IRMM-3636 is offered in 3 concentrations: 1mg/g, 0.1mg/g and 0.005mg/g. Secondly, the ^{236}U single spike IRMM-3660 was prepared and is offered in 2 concentrations: 0.1mg/g and 0.01mg/g. Thirdly, a quad-mixture, IRMM-3100a, has been prepared which is characterized by $^{233}\text{U}/^{235}\text{U}/^{236}\text{U}/^{238}\text{U}=1/1/1/1$. This material is useful for checking Faraday cup efficiencies and inter-calibration of MIC (multiple ion counting) detectors. Furthermore, earlier prepared reference materials such as the IRMM-072, IRMM-073 and IRMM-074 series for detector linearity testing will be reviewed.

Log: 124. **A COMPTON-SUPPRESSED PHOSWICH DETECTOR FOR GAMMA SPECTROSCOPY.** Abdollah Farsoni, Oregon State University.

To identify and reject Compton events and eventually reduce the Compton continuum in gamma energy spectrum, a phoswich detector with two scintillation layers has been designed and assembled at the Oregon State University. In this detector, CsI(Tl) crystal is used to primarily detect photoelectric events. The CsI(Tl) crystal is partially surrounded by a BGO crystal layer to capture and identify Compton-scattered photons. Both crystals are optically coupled to a single photomultiplier tube. A real-time digital pulse shape analysis was employed to discriminate and reject Compton-induced pulses. All the digital pulse processing functions including pulse shape analysis and energy measurement were implemented in an on-board FPGA device. In this paper, the recent measurement results using radioactive lab sources will be presented and discussed.

Log: 125. **DIGITAL GAMMA-GAMMA COINCIDENCE AND ANTICOINCIDENCE SPECTROMETRY DEVELOPMENT AND ITS APPLICATIONS TO COMPTON SUPPRESSION WITHOUT ANY GAMMA-RAY SENSITIVITY LOSS, SPECIAL ENVIRONMENTAL RADIONUCLIDE MONITORING AND NUCLEAR MATERIAL CHARACTERIZATION USING LIST-MODE DATA ACQUISITION.** Weihua Zhang; Pawel Mekarski; Maxime Dion; Jing Yi; Kurt Ungar.

Digital pulse analysis as applied to gamma spectroscopy is no longer new although recent implementations by vendors claim improved throughput, stability and functionality. Potential sources of counting errors such as peak broadening at high count rates, detector pulse pileup and pileup rejection have previously been addressed by standard methods in analog spectroscopy and were routinely considered when setting up a gamma counting system. With the new adjustable parameters in digital signal processing such as trapezoidal peak rise time and flat top as well as live time trim and the pileup rejection multiplier, it is important to assess their effects on the accuracy and reproducibility of peak extraction software. In this work, in an effort to determine optimum setup procedures for incorporation of Canberra Lynx DSP instruments into our laboratory's automated gamma spectroscopy systems which are employed in routine neutron activation analysis, we have evaluated the effect of these parameters, both individually and in concert with the other settings. Optimum settings were determined for our instruments. Variation of peak area count rate, peak width, peak centroid, FWHM are shown as a function of input count rate will all be discussed as related to parameter setting. Procedural steps for consideration of all of these parameters in system setup are suggested.

Log: 126. **INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS OF RADIONUCLIDES AND TRACE METALS IN SOIL-PLANT SYSTEMS** . Ramakrishna Naidu, G; Kalyan, Y; Gangadhar, B; Tharakeswar, Y; Sujana Kumar, K. Department of Environmental Sciences, Sri Venkateswara University, Tirupati-517502, INDIA.

The environmental impact of elemental contaminants in soils and sediments is dependent both on the chemical speciation of the metal and the response of the matrix to biological and physiochemical conditions. Overall these factors are responsible for the mobilization of the elements from solid into the aquatic phase and hence for the transport within the immediate vicinity, influencing the rate of dispersal, dilution, uptake and transfer into the living systems. In our present study an effort was made to evaluate the extent of elements that are present in soils and finally compared with the values of the same elements present in some plants (bioavailable)

which are grown in respective areas of Tirupati (Agricultural and Industrial areas). Instrumental Neutron Activation analysis was employed for the analysis of various elements like Sodium, K, Sm, Cr, Zn, Th, Rb, Sr, Fe, La, Co, Ce, Cs, and Eu in plant-soil system and the samples were irradiated with neutron flux ($7 \times 10^{11} \text{ n.cm}^{-2}.\text{S}^{-1}$) at the 100 kW TRIGA - MAINZ research reactor and the induced activities were measured by gamma ray spectrometry using high resolution High Purity Germanium (HPGe) detector. By employing this technique the above 14 elements were determined and moreover the migratory mechanism of radionuclides and trace metals were investigated in chosen soil-plant systems.

Log: 127. **BIODISTRIBUTION OF ^{212}Pb -TRASTUZUMAB IN MICE.** Schneider, N (1); Lobaugh, M (2); Sandwall, P (3); Glover, S (4); Spitz, H (5). (1) University of Cincinnati.

The biodistribution in mice of ^{212}Pb -trastuzumab, a HER2/neu-targeting immunoglobulin (monoclonal antibody), was investigated for its potential as a therapeutic agent with immunocytotoxic applications. ^{212}Pb -trastuzumab is an α emitting radioimmunoconjugate that can deliver a short-range, high LET radiation dose to targeted tissue. ^{212}Pb is thought to be a good isotope for medical applications because of its short half-life (10.64 hr), the high LET from ^{212}Po (a decay product), and it's readily available using a commercially-produced ^{224}Ra generator. The aims of this study were to determine the trastuzumab biodistribution data - specifically tumor uptake, and to support novel antibody based targeted radionuclide tumor therapy for clinic use. Radiolabeled ^{212}Pb -trastuzumab was found to be pure, functional, and intact by both ELISA and SDS-PAGE evaluation. The distribution of ^{212}Pb -trastuzumab was determined as the percent mass concentration of trastuzumab, based on decay corrected-injected dose by analysis of nine different organs from male nude mice bearing PC-3MM prostatic carcinomas. An increase in uptake was observed in 5 organs and the highest tumor %ID/g (7.78 ± 9.03) was obtained at 12 h post i. v. injection. High-resolution gamma spectrometry was used to determine the content of ^{212}Pb -trastuzumab in each organ at ten fixed times post intravenous injection and demonstrates good correlation between biodistribution data and HER2/neu receptor-specific tumor uptake. This biodistribution study supports further investigation of radiolabeled ^{212}Pb -trastuzumab; specifically the radiobiological organ dosimetry and optimal dosing regimens of trastuzumab as a therapeutic agent.

Log: 128. **PAVING THE WAY TO PERSONALIZED MEDICINE: PRODUCTION OF SOME PROMISING THERAGNOSTIC RADIONUCLIDES AT BROOKHAVEN NATIONAL LABORATORY.** Srivastava, SC, Brookhaven National Laboratory, Upton, NY.

This presentation will introduce a relatively novel paradigm that involves specific individual radionuclides or radionuclide pairs that have emissions that allow pre-therapy low-dose imaging plus higher-dose therapy in the same patient. We have made an attempt to sort out and organize a number of such theragnostic radionuclides and radionuclide pairs that may potentially bring us closer to the age-long dream of personalized medicine for performing tailored low-dose molecular imaging (SPECT/CT or PET/CT) to provide the necessary pre-therapy information on biodistribution, dosimetry, the dose limiting critical organ or tissue, and the maximum tolerated dose (MTD), etc. If the imaging results then warrant it, it would be possible to perform dose ranging studies and higher-dose targeted molecular therapy in the same patient with the same radiopharmaceutical. A major problem that remains yet to be fully resolved is the lack of availability, in sufficient quantities, of a majority of the best candidate theragnostic radionuclides in a no-carrier-added (NCA) form initially for clinical trials and subsequently for widespread clinical use in patients for therapy of cancer and other inflammatory disorders. A brief description of the recently developed new or modified methods at BNL for the production and radiochemistry of a number of potential theragnostic radionuclides, whose nuclear, physical, and chemical characteristics seem to show great promise for personalized cancer therapy will be provided. Work supported by the Office of Nuclear Physics, U.S. Department of Energy, under Contract # DE-AC02-98CH10886 at Brookhaven National Laboratory.

Log: 129. **ANALYTICAL SUPPORT TO THE DOE FUKUSHIMA RESPONSE BY THE LAWRENCE LIVERMORE NATIONAL LABORATORY.** Kreek, SA; Wimer, NG; Bandong, BB (Lawrence Livermore National Laboratory).

The Lawrence Livermore National Laboratory (LLNL) was the principle consequence management lab analysis (CMLA) capability supporting the U.S. Department of Energy (DOE) response to the Fukushima Daiichi incident. Over the span of several months, LLNL was tasked to perform thousands of analysis of samples that were collected by U.S. field teams in the region over the same period. The scale and duration of the response strained even our significant capability and highlighted the need for additional surge capacity, robust processes and the benefit of applying corroborative techniques to support and affirm technical conclusions. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL-ABS-502551

Log: 130. **SHELLED MORINGA OLEIFERA SEEDS: A UNIQUE BIOSORBENT FOR THE REMOVAL OF HEAVY METALS FROM AQUEOUS PHASE USING NAI(TI) DETECTOR AND NEUTRON ACTIVATION ANALYSIS TECHNIQUES..** Parul Sharma (1), M.M. Srivastava (2) and Shalini Srivastava (3). (1) Department of Chemistry, Amity school of Engineering & Technology, Amity University, Manesar, Haryana. (2) Department of Chemistry, Faculty of Science, Dayalabgh Educational Institute, Dayalbagh, Agra, India..

Heavy metals are among the conservative pollutants whose presence in the ecosystem is of major concern because of their toxicity, bioaccumulating tendency, threat to human life and the environment. Among dirty dozen club of toxic pollutants, high level of Cd (II), Cr (III), Cr (VI) and Ni (II) is a burning issue round the globe. Due to economic barrier for remediation, utilization of agricultural byproducts or plant biomaterials for the removal of heavy metals from the aqueous system is on the cards. High uptake capacity and cost effective source of the raw material made biosorption a progression towards a perspective method. With this aim in view the present study highlights the efficacy of shelled Moringa oleifera seeds (SMOS) for the decontamination of Cd (II), Cr (III), Cr (VI) and Ni (II) at laboratory scale. The percent sorption in case of Cd (II), Cr (III), Cr (VI) and Ni (II) was computed on the basis of metal estimation using NaI(Tl) detector coupled to 4K MCA (Canberra Accuspec Card with PC-AT 386) while Ni (II) was estimated using Neutron Activation Analysis. The adsorption data fitted accurately in Langmuir and Freundlich isotherm. Sorption studies resulted in the standardization of optimum condition for the abatement of Cd (II) [85.10%], Cr (III) [81.02%], Cr (VI) [88%] and Ni (II) [75.64%] at pH 6.5 for Cd (II) and Cr (III), pH 2.5 for Cr (VI) and pH 7.5 for Ni (II). Fourier Transform Infrared Spectrometry (FTIR) highlighted amino acid-metal interactions responsible for sorption phenomenon.

Log: 131. **ELECTRODEPOSITION AND CHARACTERIZATION OF PLUTONIUM AND URANIUM DEPOSITED ONTO THIN CARBON AND TITANIUM SUBSTRATES.** Roger Henderson, Julie Gostic, Ching-Yen Wu, Jason Burke, and Scott Fisher
Lawrence Livermore National Laboratory

Physics experiments aimed at deducing key parameters for use in a variety of programs critical to the mission of the National Laboratories require actinide targets placed onto various substrates. The target material quantity and the substrate desired depend upon the type of experiment being designed. The physicist(s) responsible for the experimental campaign will consult with the radiochemistry staff as to the feasibility of producing a desired target / substrate combination. In this report we discuss the production of U and Pu targets on very thin C (100 g/cm²) and Ti (2 & 3 m) substrates. The Ti targets are plated on both sides simultaneously. The techniques used, plating cells designed for, tips, and limits will be discussed. A new apparatus for performing high-precision alpha spectrometry on the produced targets has been constructed using an alpha pinhole camera and a micropositioning X-Y stage. Results for the assay of large area targets (4-cm diameter) will be discussed. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 132. **FABRICATION AND CHARACTERIZATION OF AN IRRADIATION FACILITY FOR LARGE-SAMPLE GEOMETRY.** Talnagi, JW(1); Lei, LR(1); Spitz, HB(2); Glover, SE (3). (1) The Ohio State University, (2) University of Cincinnati, (3) NIOSH.

An irradiation facility consisting of a modified beam port shielding plug has been designed, fabricated built and characterized for use in irradiating non-standard sample geometries. The shielding plug features a graphite moderator at the core end with a hole, or "well" drilled of sufficient diameter and depth to accommodate an eight ounce (227 gram) sample bottle. Added shielding behind the graphite consists of castable neutron and -gamma-ray shielding. The modified shielding plug can be removed relatively quickly from its irradiation position to minimize personnel exposures. It is mounted in close proximity to the OSURR reactor core to allow performance of high-sensitivity neutron activation analysis (NAA) studies. Using the SAND-II unfolding code, the energy-dependent neutron flux has been measured in the sample irradiation position. When operating at 100% power, the total flux is 3.9×10^{12} n/cm²/second. Of this, 56% is thermal (< 0.5 eV), 23% is epithermal (>0.5 eV, <0.5 MeV), and 21% is "fast" (>0.5 MeV). This makes the facility suitable for neutron activation studies. Recently it has been used for irradiation of filter papers collected in a study of particulate air pollution in the form of atmospheric particulate matter (PM) in an urban environment.

Log: 133. **RADIOACTIVE TARGETS FOR NUCLEAR ASTROPHYSICS EXPERIMENTS.** Dorothea Schumann Paul Scherrer Institute Villigen Switzerland.

The description of the galactic evolution and the understanding of the history of the universe require experiments to study the production and destruction rates of the elements under galactic conditions. Isotopes with relatively long-lived half-lives play an essential role in the s-process as so-called "waiting points", where the usual neutron capture and beta decay cycle is competing with the possibility of additional neutron capture processes. "Hotspots" in this research field are for instance ⁶⁰Fe or ⁶³Ni, where nuclear data are scarce, uncertain or completely missing due to the poor availability of the material in the required quality and quantity. One possibility to gain such samples is "mining" accelerator waste from high power proton accelerators, what we did for producing a ⁶⁰Fe-target. ⁶³Ni can be obtained by neutron activation in high-flux reactors using specially enriched ⁶²Ni targets, but older samples suffer from a high content of the decay product ⁶³Cu disturbing the measurements due to high neutron resonances. Before using such samples, an additional chemical purification is necessary. The paper describes the separation and purification of ⁶⁰Fe and ⁶³Ni samples and the following preparation of the targets used in neutron capture cross section measurements. The nuclear astrophysics experiments were performed at KIT Karlsruhe in collaboration with the University of Notre Dame, the GSI Darmstadt, the University of Lodz and the Keele University as well as at CERN in the frame of the n-TOF collaboration.

Log: 134. **THE POTENTIAL FOR RADIONUCLIDE AND ISOTOPIC ANALYSES USING A RAPID SCANNING MAGNETIC SECTOR ICP-MS.** John Cantle & Sabine Pawlig, Nu Instruments UK.

Precise and accurate elemental and isotopic analysis remains a major demand of research and contracting laboratories. To increase sample throughput and achieve ultimate performance instruments have to combine speed, reliability, flexibility and ease-of-use. The Nu Instruments AttoM® is a new generation, high-resolution magnetic sector ICP mass spectrometer which incorporates a number of novel and unique design features. With the so-called "FastScan Ion Optics" extremely fast peak jumping acquisitions can be performed. These peak jumping acquisitions can be performed over a 40% relative mass range without changing the magnetic field. Jumping times between peaks are <20 microseconds and dwell times for peaks can be set individually with a minimum dwell time of 100 milliseconds. This offers the great flexibility to adapt application parameters for best analysis precision. This paper will present HR-ICP-MS data to illustrate the instrument's potential for rapid isotopic ratio acquisition for a variety of applications. Examples will include Uranium isotope ratios on different low concentrated isotopic

standards, Uranium samples with different isotopic compositions from a nuclear fuel processing source as well as Plutonium isotopes in seawater.

Log: 135. **A STUDY OF ENVIRONMENTAL RADIOACTIVITY ALONG THE NORTH WESTERN COAST OF EGYPT WAS PERFORMED BY CENTRAL LABORATORY FOR ENVIRONMENTAL RADIOACTIVITY MEASUREMENTS INTER-COMPARISON AND TRAINING (CLERMIT) WITHIN THE FRAMEWORK OF THE RESEARCH PROJECT: ENVIRONMENTAL RADIATION MEASUREMENTS AND HARMONIZATION.** Hanan M. Diab, Ph.D., A. A Ramadan, Ph.D.

A study of environmental radioactivity along the North western coast of Egypt was performed by Central Laboratory for Environmental Radioactivity Measurements Inter-comparison and Training (CLERMIT) within the framework of the research project: "Environmental Radiation Measurements and Harmonization". The paper contains the most important conclusions and some correlations and interpretations of the obtained results. The measurements and samples collection were planned according to the monitoring plan which included locations along the North western coast of Egypt and its main tributaries of interest. The specific activities of ^{226}Ra (^{238}U series), ^{228}Ra (^{232}Th series), ^{40}K and ^{137}Cs (Bq/kg dry weight) were measured using gamma ray spectrometers based on hyper-pure germanium detectors. The absorbed radiation dose rates in air (nGy/h) due to natural radionuclides in shore sediment and radium equivalent activity index (Bq/kg) were calculated. The specific activity ratios of $^{228}\text{Ra}/^{226}\text{Ra}$ were calculated for evaluation of the geo-chemical behavior of these radionuclides. Also samples texture, CaCO_3 and total organic matter contents were determined. pH, major cations and anions as Na^+ , K^+ , Ca^{++} , Mg^{++} and Cl^- , SO_4^{--} and HCO_3^- , respectively were also determined in water samples

Log: 136. **DEVELOPMENT OF A PROMPT GAMMA ACTIVATION ANALYSIS FACILITY AT DALAT REACTOR.** Nguyen Canh Hai(1), Vuong Huu Tan(2), Nguyen Nhi Dien(1), Pham Ngoc Son(1), Tran Tuan Anh(1), (1) Nuclear Research Institute, 1 Nguyen Tu Luc street, Dalat, Viet Nam, (2) Viet Nam Atomic Energy Institute, 59 Ly Thuong Kiet street, Hanoi, Viet Nam.

A prompt gamma activation analysis facility has been developed at the channel No. 4 of Dalat Nuclear Research Reactor. Thermal neutron beam was received by setting filter combination of silicon, titanium and sulphur in the channel. The parts of the facility consist of collimators, beam stopper, sample holder table, beam catcher, and biological shield. Digital signal processor includes amplifier, ADC and MCA was used to collect and sort the detector signal and the ORTEC Gamma-Vision was used for displaying and on-line analyses of the accumulating spectrum. GEM series HPGe-254 cc detector manufactured by ORTEC is used to count the prompt gamma rays (resolution of 1.9 keV at 1332 keV and relative efficiency of 58%). The characteristics of the facility such as neutron flux, cadmium ratio, gamma background, elemental sensitivities were examined and presented. The facility was used to demonstrate its application for determination of elemental concentrations of biological, geological materials. The analysis procedure for the facility is described in detail. The analysis results of Ti, K, Al, Si, Fe, Ca, Mn, Gd, Sm, B in geological sample and N, K, Cl, Fe, Ca, Mn, B, H in biological sample using synthetic standards are presented. The analytical capacity of the facility is discussed and a plan to upgrade the gamma spectrometer is also presented

Log: 138. **MONITORING OF THE ENVIRONMENTAL PATHWAYS OF RADIONUCLIDES RELEASED AFTER THE FUKUSHIMA REACTOR ACCIDENT IN AUSTRIA.** Merz, S (1); Steinhäuser, G (1); Hainz, D (1). (1) Vienna University of Technology, Atominstitut.

After the Tohoku earthquake on March 11, 2011, the nuclear power plants of Fukushima Daiichi, prefecture of Fukushima, suffered a major nuclear accident, which lead to the release of significant amounts of radioactive material. The monitoring of several radionuclides in various types of environmental samples will provide detailed information on the different contamination pathways, i.e. the transport of radionuclides via air, deposition caused by rain, accommodation into the food chain and bio-accumulation of radionuclides. Defined volumes of air, rainwater, soil,

different kinds of foodstuff and thyroid glands of wild animals are samples that are tested for radionuclides such as those of cesium and iodine. Using the very restrictive Austrian regulatory concerning radioactivity, the concentration of radionuclides in the environmental samples can be converted into useful dose rates in order to assess possible effects on human health. Marking the origin of the different samples, a map can be formed that might contain detailed information on the diffusion and accumulation of the released radionuclides. The gathered information should reveal which kind of environmental carriers are adequate to acquire an estimation of the dosage rate affecting the human body and its significance for human health. With respect to health effects, the accumulation of radionuclides in the human body and the food chain is of greatest importance. Our results indicate enormous bioaccumulation of radioiodine in animal thyroid glands, thus suggesting these organs for the use of radioiodine bio-monitors. This may be of importance for CTBT (Comprehensive Nuclear-Test-Ban-Treaty) verification.

Log: 139. **PURECOLD FAST ELECTRONICS FOR BETA-ALPHA- AND ALPHA-ALPHA PILE-UP SUPPRESSION.** Rugard Dressler and Peter Rasmussen, Paul Scherrer Institute, CH-5232 Villigen, Switzerland.

The unambiguous identification of isotopes of the heaviest elements is a very important prerequisite for the investigation of their nuclear properties. The time-resolved spectroscopic recording of consecutive alpha decays of the investigated isotope as well as its descendents gives the highest possible confidence of assigning an observed event to the isotope of interest. However, the formation of unwanted by-products in the used nuclear reactions may lead to large amounts of unwanted events in alpha-spectroscopic energy regions, where the decay of the investigated isotopes or their daughter nuclei is expected. In particular, the specific beta-decay of Bi-212 (half-life 60.6 min) and the following very short alpha-decay of Po-212 (half-life 0.3 us) as descendants of Rn-220, leading in conventional spectroscopic systems to so called pile-up pulse height amplification, disturbs the measurement. We present here the design and performance of the newly developed multichannel spectroscopic detection technology PURECOLD which is able to electronically resolve the consecutive hits of beta- and alpha-particles in time down to nanoseconds and register the data on an event-by-event base with overall event rates up to 12000 per sec and spectroscopic channel. Furthermore, this system has the capability to measure time-wise very short consecutive alpha decays, which appears to be increasingly important for the discovery of new elements and new isotopes.

Log: 140. **USING HIGH-RESOLUTION IN-SITU RADON MEASUREMENTS TO DETERMINE GROUNDWATER DISCHARGE INTO TONLE SAP LAKE, (CAMBODIA).** Burnett WC (1); Peterson R (2); Chanyotha S (3); Kritsanawanat R (3); Ryan B (4). (1) Florida State University, (2) Coastal Carolina University, (3) Chulalongkorn University (Thailand), (4) Department of Sustainability, Environment, Water, Population and Community (Australia).

Tonle Sap Lake (Cambodia) is the largest freshwater lake in SE Asia, and is reported to have one of the highest freshwater fish productions anywhere. The lake's biology is influenced by its very unique hydrologic cycle. During the dry season the lake drains through a tributary to the Mekong River. During the wet monsoon, the flow in the connecting tributary reverses, adding huge volumes of water and increasing the lake area about 4-fold. We hypothesize that nutrients are at least partially delivered via groundwater discharge, a direct consequence of the annual flood cycle. We surveyed over 100 km in the northern section of the lake using a customized system that measures Rn-222, temperature, conductivity, and GPS coordinates while underway. These surveys were performed at the end of the flood stage (Nov 2009) and at the low water point (Apr 2010). Results of these expeditions showed that there were portions of the lake that showed significant enrichments in radon, a groundwater indicator, and dissolved nitrogen. These same areas were generally characterized by very low electrical conductivities. Samples collected from nearby groundwater wells also showed this inverse relationship between radon and conductivity. Our results thus suggest that groundwater pathways are important, accounting for approximately 10-20% of the flow of the Tonle Sap tributary (connection to the Mekong River), the largest single

source of fresh water to the lake. Analysis of groundwater discharge in a lake setting is relatively straight-forward as the inputs would be balanced mostly by decay and atmospheric evasion.

Log: 141. **INVESTIGATION OF URANIUM TRIOXIDE POLYMORPHISM AND HYDROLYSIS.** Lucas E. Sweet (Pacific Northwest National Laboratory), Charles H. Henager Jr. (Pacific Northwest National Laboratory), Shenyang Hu (Pacific Northwest National Laboratory), Thomas A. Blake (Pacific Northwest National Laboratory), Timothy J. Johnson (Pacific Northwest National Laboratory), David E. Meier (Pacific Northwest National Laboratory), Shane M. Peper (Pacific Northwest National Laboratory), Jon M. Schwantes (Pacific Northwest National Laboratory).

This work focuses on progress in gaining a better understanding of the polymorphic nature of the UO_3 -water system; one of several important materials associated with the nuclear fuel cycle. The UO_3 -water system is complex and has not been fully characterized, even though these species are common throughout the fuel cycle. For example, most production schemes for UO_3 result in a mixture of up to six or more different polymorphic phases, and small differences in these conditions will affect phase genesis that ultimately results in measureable changes to the end product. Here we summarize our efforts to better characterize the UO_3 -water system with a variety of optical techniques for the purpose of developing some predictive capability of estimating process history and utility, e.g. for polymorphic phases of unknown origin. Specifically, we have investigated three industrially relevant production pathways of UO_3 and discovered a previously unknown low temperature route to beta- UO_3 . Powder x-ray diffraction and optical spectroscopies were utilized in our characterization of the UO_3 -water system. Pure phases of UO_3 , hydrolysis products and starting materials were used to establish optical spectroscopic signatures for these compounds. Preliminary aging studies were conducted on the alpha- and gamma- phases of UO_3 . In addition, development of a 3-D phase field model was used to predict phase genesis under a variety of industrially relevant conditions has been initiated.

Log: 142. **FORMATION OF A HETERONUCLEAR HYDROLYSIS COMPLEX IN THE TH(IV)/FE(III) SYSTEM.** Natallia Torapava (1); Artsiom Radkevich (2); Ingmar Persson (3); Dmitri Davydov (2); Lars Eriksson (4). (1) Chalmers University of Technology; (2) Joint Institute for Power and Nuclear Research-Sosny; (3) Swedish University of Agricultural Sciences; (4) Stockholm University..

Knowledge about actinoids speciation is important for assessment of their possible migration in the environmental systems, like either mine tailings, industrial wastes or nuclear final repositories. As in natural waters or waste waters usually more than one metal ions are present, it is important to consider two or more multicharged metal ions systems. A speciation of thorium(IV) and iron(III) present together has been studied in the pH range 2.3 –4.8 and at metal concentrations 0.02 – 0.05 mol·cm⁻³ in aqueous solution. Ferrihydrite precipitates after months of storage in solutions at pH values 2.0 (six-line) and 2.3 (two-line). Whereas solutions with pH ≥ 2.9 stayed clear over 24 months of experiments. Extended X-ray absorption fine structure, EXAFS, and large angle X-ray scattering, LAXS, techniques indicated Th-O and Fe-O bond distances of 2.40 and 2.45 Å, 1.96 and 2.00 Å, and Th···Th, Fe···Fe and Th···Fe distances of 3.94 and 3.96 Å, 3.04 and 3.02 Å, 3.41 and 3.43 Å, respectively. A heteronuclear complex with composition $[\text{Th}_2\text{Fe}_2(\mu_2\text{-OH})_8(\text{H}_2\text{O})_{12}]^{6+}$ is proposed to be formed in the acidic aqueous solution. The mechanism of heteronuclear complex formation is unknown, but probably hydrolyzed thorium(IV) and iron(III) diamers are the building blocks. The formation of such a complex may increase mobility of actinoids in the natural waters and lead to release of them into environment.

Log: 143. **RADIOCHEMICAL AND CHEMICAL ANALYSIS OF RADIOACTIVE SLUDGES FROM VVER NPP.** Pátzay, G(1); Vajda, N. (1); Feil, F. (2), Patek, G.. (2). (1) Budapest University of Technology and Economics, Hungary, (2) NPP Paks, Hungary.

There is about 6000 m³ concentrated evaporator bottom residues in the tanks of the VEER in Paks Hungary. There are some tanks at the power plant containing sludge type radioactive waste

containing more or less liquid phase too. The general physical and chemical characteristics and chemical and radiochemical composition are important information for volume reduction and solidification treatment of these wastes. We have investigated and constructed a complex analysis system for the radioactive sludge and supernatant analysis, including the physical, as well as the chemical and radiochemical analysis methods. Using well known analysis techniques as ion chromatography, ICP-MS, AAS, gamma- and alpha-spectrometry and chemical alkaline fusion digestion and acidic dissolution methods we could analyze the main inorganic, organic and radioactive components of the sludges and supernatants. The radiochemical composition of the sludge samples was determined using gamma- and alpha-spectrometry and liquid scintillation measurements. We analyzed the centrifuged supernatant liquid, the washing water of the separated solid phase and the resulting solution of the fusion digestion. For the gamma-spectrometry we used an ORTEC Model GMX25P4-76-C, Gamma-X HPGe Coaxial Detector with a CarbonFiber window, connected to a multichannel analyzer ORTEC DSPEC-jr-2.0, the spectra were measured with 100 cm³ polyethylene sample with a 3600 sec counting time. Spectra were evaluated with a Gamma Vision – 32 code. Finally we determined the alpha- and beta-emitting isotopes in the samples too using preconcentration and salt removal techniques. For the alpha-activity measurements we used TRU columns and alpha-spectrometry and for the determination of beta-activity liquid scintillation technique.

Log: 144. **PULSE TUBE REFRIGERATION FOR HPGE DETECTORS AND ASSOCIATED ELECTRONIC.** Michael E. Cournoyer (1), James Pecos (1), Howard N. Granzow (1), and Steve Chunglo (2). (1) Los Alamos National Laboratory, Los Alamos, NM 87545 (2) CANBERRA Industries, An AREVA company, 800 Research Parkway, Meriden CT 06450

High Purity Germanium (HPGe) gamma-ray detectors are used for Nondestructive Assay. Liquid nitrogen (LN2), a cryogen, is commonly used to cool these detectors. Cryogen use is associated with several health risks and operational problems. This has prompted the development of cryogen-free refrigeration. The features and benefits of a modified Joule-Thompson cooler have been previously reported at this conference. A new generation of commercial pulse tube refrigerator (PTR) has been developed during the last decade. A unique feature of the PTR is the absence of cold moving parts. This considerably reduces the generated noise and vibration and increases the reliability of the cold head, as expensive high-precision seals are no longer required and the cold head can be operated without service inspection. The refrigerant used in the PTR is pure helium and, therefore, both CFC-free and non-flammable. In the following report, we examined LN2, a modified Joule-Thompson cooler, and a PTR unit and compared their cooling effectiveness with HPGe gamma-ray detectors. Overall, PTR is an engineering equivalent to LN2 and modified Joule-Thompson cooler systems used in gamma spectroscopy and eliminate the health and physical hazards associated with LN2 systems without adding hazards.

Log: 145. **PROFICIENCY TEST PROGRAM FOR CTBT RADIONUCLIDE LABORATORIES: EXERCISE ON A FUKUSHIMA-SIMILAR SAMPLE.** Duran, E.B.; Khrustalev, K. and Auer, M. CTBTO.

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) Protocol provides for 16 radionuclide laboratories to support the network of radionuclide monitoring stations being established as part of its verification regime. These laboratories support radionuclide stations by: (i) corroborating the results of routine analysis of a sample from an IMS station, in particular to confirm the presence of fission products and/or activation products, (ii) providing more accurate and precise measurements and (iii) clarifying the presence or absence of fission products and/or activation products in the case of a suspect or irregular analytical result from a particular station (CTBT/WGB/TL-11/5/Rev. 10). The expert services provided by these laboratories are utilized in the QA/QC program of the IMS radionuclide network. In turn, the Proficiency Test Exercise (PTE) is used to assess the level of accuracy of nuclide identification and measurement by laboratories and trigger corrective actions when there are discrepant results. Since 2000, the Provisional Technical Secretariat (PTS) has organized annual PTEs. This paper presents the results of

PTE2010 which dealt with a sample spiked with nuclides produced from neutron irradiation of 99.9% enriched U-235 target with minimal chemical processing. Thus, the fission products present are similar to some samples which were detected subsequently at several stations following the Fukushima event, and which the certified laboratories were asked to re-analyze.

Log: 146. **RADIOACTIVITY ASSESSMENT IN SOIL FOR SELECTION OF REFERENCE AREA FOR IN SITU GAMMA SPECTROMETRY IN JEJU ISLAND, SOUTH KOREA.** Kim, CJ (1); Cho, YH (1); Byun, JI (2); Yun, JY (2) University of science and Technology (2) Korea Institute of Nuclear Safety (KINS).

In situ gamma spectrometry using the high purity germanium (HPGe) detector of soil activity provides sensitive and representative data, compared to data obtained by conventional analysis in a laboratory. A detector placed about one meter above the ground detects gamma rays from an area within about a 10 meter radius and comparable counting statistics can be obtained in only a small fraction of the time required for analysis in the laboratory. So, it is useful for investigation of radioactivity of a wide area and radiation accident. To verify in-situ spectrometry, reference field knowing the concentration of radioactivity in soil is required. The soil of Jeju Island in South Korea contains more anthropogenic radionuclides than any other areas in South Korea and has not been disturbed from external factors, so that it is suitable as a reference field. We selected an area of pasture in Jeju Island which was expected to be uniform concentration of soil radioactivity and divided the area into radius of about 10 meters. For divided section, the soil radioactivity was assessed in two methods which are gamma-ray analysis in situ and in a laboratory. These results will show the accuracy of in situ gamma spectrometry and validity of the area for reference site. This study can be used for international proficiency test and inter-comparison exercise about in situ measurements.

Log: 147. **THE INTERNATIONAL ATOMIC ENERGY AGENCY'S ECAS PROGRAM: DESIGN AND DELIVERY OF STATE OF THE ART NUCLEAR ANALYTICAL CHEMISTRY LABORATORIES FOR SAFEGUARDS SAMPLE ANALYSES.** David W. Swindle Jr; P.E. (1); Robert L. McGill, P.E. (2); Gabriele Voigt, PhD (2); David Donohue, PhD (2); Steven D. Balsley, PhD (2). (1) URS Federal Services Corporation; (2) International Atomic Energy Agency.

In October 2007, the International Atomic Energy Agency's (IAEA) Board of Governors was informed that the Agency's ability to provide independent and timely analysis of safeguards inspection samples was at risk because of aging technical infrastructure and analytical equipment at its existing Safeguards Analytical Laboratory (SAL) in Seibersdorf. Following this communication, the IAEA established the program "Enhancing Capabilities of Safeguards Analytical Services" – ECAS, with the aim to get Member States' support for modernization and replacement of the IAEA's aged nuclear analytical laboratory and infrastructure, and expand its capability for sensitive environmental sample analyses in order to more fully satisfy its safeguards and non-proliferation missions. This paper will describe the features and capabilities of the new laboratories being designed and constructed by the IAEA under the ECAS Program, and in particular, will describe the newly constructed state-of-the-art Clean Laboratory for highly sensitive environmental sample analyses, and the new Nuclear Material Laboratory under construction to provide expanded analytical capabilities for U, Pu and other minor actinides, including storage, training and standards preparation facilities in order to meet future safeguards needs. In addition, a summary will be provided on the engineering and project management techniques and processes used to effectively deliver the designs and construction on schedule and on budget.

Log: 148. **RADIOXENON SIGNATURES BY NATURAL ACTIVATION.** Biegalski, SR (1); Klingberg, FJ (1). (1) University of Texas at Austin.

For being able to distinguish between nuclear explosions and other man made releases, atmospheric monitoring of radioactive noble gas isotopes, xenon isotopes in particular, is of interest to the non-proliferation community. Radioxenon releases by nuclear weapons tests

(atmospheric and underground) and emissions caused by research, commercial reactors as well as medical isotope production facilities and their impact on atmospheric sample analysis have to be well understood to distinguish between those events. This work focuses on the contribution of xenon activation by thermal, epithermal/fast, and 14MeV neutrons to radioxenon signatures. Experimental measurements of neutron activation are compared to MCNP calculations. The theoretically determined source term of radioxenon isotopes created by activation will be compared to the isotopes originating from fission in a nuclear explosion are compared. Additionally, calculations to estimate radioxenon production by activation in research reactors around the world are performed. Thus the natural xenon activation signature can be compared in MIRC plots to the activation during a nuclear weapon test.

Log: 149. **RADIOANALYTICAL EVALUATION OF ENVIRONMENTAL SAMPLES FROM THE FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT CRISIS.** Bryan B. Bandong, Cynthia L. Conrado, Ross W. Williams, Nathan G. Wimer, Carolyn T. Wong, and Steven A. Kreek; Chemical Sciences Division, N Program - Global Security Directorate; Lawrence Livermore National Laboratory.

In support of the U.S. Department of Energy Consequence Management (CM) Program's response to assess the radioactivity releases to the environment from the failed Fukushima Dai-ichi Nuclear Power Plant (FDNPP) brought about by the magnitude 9.0 earthquake and tsunami of March 11, 2011, Lawrence Livermore National Laboratory performed radiochemical analyses on environmental samples taken from the perimeters of FDNPP between March – May, 2011. Laboratory-based gross alpha/beta, gamma spectrometric, alpha pulse height, and mass spectrometric analyses were performed on air, soil, and swipe samples. Sample management operations, laboratory processes, and analytical procedures were established for the response effort to meet CM's requirements on short turnaround time and minimum detection limits on a large number of samples received over a short period of time and, in many cases, limited sample sizes. Results will be presented to serve as examples on how initial and earlier data guided us through modifications of analytical procedures to meet the technical and operational requirements of CM and gain insight on measurement accuracy, chemical speciation and fractionation of detected radionuclides. Lessons learned concerning sample management and analytical operations in support of this real-world nuclear incident will be discussed and potential mechanisms for improvement for future consequence management response will be presented.

Log: 150. **SUBMARINE DISCHARGE OF NUTRIENT-ENRICHED FRESH GROUNDWATER AT BÚZIOS, BRAZIL SOUTHEASTERN COAST, USING NATURAL AND CONSERVATIVE TRACERS.** Souza, TA (1); Godoy, JM (1,2); Godoy, MLDP (2); Carvalho, ZL (2); Fernandes, FC (3); Moreira, I (1). (1) PUC-Rio, (2) Instituto de Radioproteção e Dosimetria (IRD/CNEN), (3) IEAPM.

Submarine groundwater discharge (SGD) is becoming acknowledged as an important source of materials to the coastal ocean, including nutrients, particularly when originating from contaminated continental aquifers. Four radium isotopes exist in the nature (^{226}Ra , ^{228}Ra , ^{223}Ra and ^{224}Ra). In fresh waters, the radium appears adsorbed to the particulate material while in the seawater radium presents a conservative behavior, being the concentration of different radium isotopes governed by the processes of dilution, advection and diffusion, as well as the radioactive decay. The objective of this work is to consolidate the application of radium isotopes and other tracers as salinity, ^{222}Rn , $\delta(18\text{O})$, $\delta(\text{D})$, Ba, Si, U and nutrients in the study of water mixing in coastal regions, mainly, enclosing the SGD. Seawater and groundwater samples were collected between December 2010 and December 2011 in the Búzios region. Seawater samples were collected along offshore transects and groundwater samples in wells located in the shoreline, and filtered through columns packed with manganese fiber. Short-lived radium isotopes activities (^{223}Ra and ^{224}Ra) were measured using a scintillation cell interfaced to a photomultiplier and a delayed coincidence counter. Long-lived radium isotopes activities were determined by the RaDeCC system after the development of methodology for the measurements of ^{226}Ra and ^{228}Ra . ^{222}Rn activities in the seawater were determined using the RAD7 Detector. The nutrients $\text{PO}_4^{=}$,

NO_3^- , NO_2^- , NH_4^+ were measured. Barium and silicon concentrations were measured using a ICP-OES and uranium using a ICP-MS. Salinity and temperature were measured in situ. $\delta(18\text{O})$ and $\delta(\text{D})$ were determined applying a PICARRO L1120i isotope water analyzer.

Log: 151. **USE OF A COTS X-RAY SCANNER FOR 2-D NEUTRON ACTIVATION ANALYSIS.** Bounds, JA; Goda, JM; Myers, W; Rose, E; Sanchez, RG. Los Alamos National Laboratory.

Results from the use of a COTS x-ray scanner using storage phosphors to measure neutron activation in one- and two- dimensions are presented. The technique consists of irradiating thin foils or wires of various elements, then placing the activated material on the storage phosphors to expose them. The amount of exposure is proportional to the activation obtained. Examples of wires, small foils, and large area foils with assymetric irradiation using critical assemblies are presented. Combined with isotope-specific gamma counting of the entire foil or wire, the technique offers a simple way to obtain both qualitative and quantitative 2-D activation information.

Log: 152. **UTEX MODELING OF SUBSURFACE RADIOXENON TRANSPORT.** Justin Lowrey, University of Texas at Austin; Steven Biegalski, University of Texas at Austin; Mark Deinert, University of Texas at Austin.

The Underground Transport of Environmental Xenon (UTEX) model is a finite-difference code that was developed at the University of Texas at Austin to simulate the transport of radioxenon from an underground nuclear detonation to the surface. UTEX handles a time dependent source term and includes the effects of radioactive decay on positive and negative contributions to the system inventories. Isotopic signatures of the various radioxenon species can be determined as a function of release time. The model shows that significant perturbations in the isotopic signatures are possible under some geologic conditions, and that these perturbations tend to level out once emissions reach a steady-state level at the surface. Transport of radioxenon gas in UTEX is driven in large part by atmospheric pumping. A study was undertaken to characterize the dependence of isotopic signatures on the driving barometric fluctuations, so as to gain a degree of understanding of what effects that various weather conditions can have on resulting signatures. Additionally, the model was used to simulate measurements at various depths below the system surface; the dependence of isotopic radioxenon signatures on sampling depth was examined.

Log: 153. **PULSE SHAPE EFFECTS IN ULTRAFAST LASER INDUCED BREAKDOWN SPECTROSCOPY.** K.C. Hartig, J.P. McNutt, P. Ko, I. Jovanovic Department of Mechanical and Nuclear Engineering The Pennsylvania State University.

Laser induced breakdown spectroscopy (LIBS) is a promising method for detection and characterization of materials at a range of standoff distances. LIBS involves the collection of light from a micro-plasma formed when a laser pulse ablates a material surface. The light emitted from the plasma can be spectrally analyzed to yield both elemental and isotopic composition. The non-destructive, expeditious, and in situ capabilities of LIBS have been identified by the IAEA as promising for the use in detection of nuclear materials and for the use in attribution process for interdicted materials. When compared to other sensitive methods for material detection, such as mass spectroscopy, LIBS signals exhibit a lower signal to noise ratio (SNR), resulting in higher detection limits. Increasing the SNR would improve the ability to obtain the sample composition with increased accuracy and speed and to reduce the amount of material needed to perform analysis. We are using the modern femtosecond laser technology and developing novel techniques to significantly improve the sensitivity of LIBS. In particular, we have been investigating the effect of laser pulse shaping to achieve coherent control of the breakdown and recombination processes that occur in the laser-produced plasma. The pulse shaping is performed using an acoustic-optic programmable dispersive filter. An adaptive learning algorithm is being developed to automate the pulse shape optimization process for maximization of LIBS SNR in nuclear security-relevant material characterization scenarios.

Log: 154. **ASSESSMENT OF THE ASSOCIATED PARTICLE PROMPT GAMMA NEUTRON ACTIVATION TECHNIQUE FOR TOTAL BODY NITROGEN MEASUREMENT IN VIVO.**

Shypailo RJ; Workeneh B; Ellis KJ. USDA/ARS Children's Nutrition Research Center, Department of Pediatrics, Baylor College of Medicine, Houston, TX 77030.

Total Body Nitrogen (TBN) can be used to estimate Total Body Protein (TBP), an important body composition component at the molecular level. A system using the associated particle technique in conjunction with prompt gamma neutron activation analysis has been developed for the measurement of TBN in vivo. The system uses a compact D,T neutron generator ($\sim 10^7$ n/s) coupled to an internal alpha-particle detector, and a counting system with six bismuth germinate (BGO) detectors. 14 Subjects were scanned from shoulders to hips (20 min scan time, <0.4 mSv dose) generating complex spectra dominated by signals from C, O, H, and N, with significant peak overlap. Fractional contributions from these elements to regions of interests (ROI) spanning a 4 to 8 MeV range were determined by algorithms comparing ratios of interrelated ROIs. In addition, multi-component least squares fitting was done to further resolve individual peak activities (MATLAB R2011b). Total body potassium (TBK) was also measured using a whole body gamma counter. Predicted TBN values, based on fat-free mass estimated from TBK, were compared to measured TBN results. Measured versus predicted results for all subjects were not statistically different. Separating subjects by gender also showed no difference between measured and predicted values. The associated particle system showed good agreement with predicted TBN values, but measurement precision was not better than that commonly seen in traditional prompt gamma thermal neutron activation analysis systems.

Log: 155. **CHROMATOGRAPHIC SEPARATION OF NO-CARRIER-ADDED RU-97 FROM C-12 IRRADIATED NATURAL YTTRIUM TARGET.** Susanta Lahiri(1); Moumita Maiti(1). (1)

Chemical Sciences Division, Saha Institute of Nuclear Physics, 1/AF Bidhannagar, Kolkata 700064, India.

No-carrier added (nca) ^{97}Ru (2.83 d) is one of the promising clinical radionuclides due to its favorable nuclear properties; absence of β^+ , low-energy γ -emissions: 215.70 (85.62%) and 324.49 (10.79%) keV, and rich chemistry of ruthenium. This paper reports a heavy ion induced production of ^{97}Ru on natural yttrium target and its subsequent separation from the bulk by ion exchange chromatography. To produce ^{97}Ru , a natural yttrium target was irradiated by 70 MeV ^{12}C projectiles for 3 h with an average beam current of 90 nA. Nca ^{97}Ru was produced via $\text{natY}(^{12}\text{C},4n)^{97}\text{Ru}$ and $\text{natY}(^{12}\text{C},p3n)^{97}\text{Ru}$ reactions. Assuring the production of ^{97}Ru by g-spectrometry, irradiated target was dissolved in 0.1 M HCl, spiked with ^{88}Y (106.6 d), evaporated to dryness, and residue was taken into 0.01 M HCl. About 0.5 mL of active solution was adsorbed on a column containing cation exchanger resin, DOWEX-50, preconditioned with dilute 10-2 M HCl. Nca ^{97}Ru and bulk yttrium were eluted from the column by HCl solutions adjusting elution rate to ~ 2 drops/min. About 70% ^{97}Ru was eluted from the column by 0.1 M HCl most likely as Ru(IV) species. Rest amount of ^{97}Ru ($\sim 30\%$) and bulk yttrium retained in the column were eluted by 6 M HCl in sequel probably as Ru(III) and Y(III) with a small overlap in between. A total of $\sim 85\%$ ^{97}Ru got separated without any contamination of bulk matrix leading to a high separation factor. It is one step, eco-friendly separation technique and is an evidence of ruthenium speciation.

Log: 156. **SPECIATION OF TECHNETIUM 99 IN SUPERACIDIC MEDIA UNDER ALPHA IRRADIATION (ARRONAX CYCLOTRON).** I,Denden (1); R, Essehli (1); F,V, Massoud (1) and (1) Subatech Laboratory France.

The speciation of pertechnetate ($^{99}\text{Tc(VII)O}_4^-$) in concentrated trifluoromethanesulphonic acid ($\text{CF}_3\text{SO}_3\text{H}$) was carried out. In the absence of alpha radiation, spectrophotometric results revealed for the first time the reduction of pertechnetate in triflic acid. Three species has been obtained as a function of the acid concentration [5M-11M]: oxo-polymer Tc (IV) for concentrations lower then 8 M, a new Tc (IV) species at 9 M and non reduced core at 11 M. However, the kinetics of formation of reduced technetium is very slow. Unexpectedly, the in-situ UV-visible

spectroscopy shows that the same reduced species have been obtained by irradiating few minutes with highly energetic alpha particles (70 MeV) in ARRONAX cyclotron (France). The local structure of Tc species have been recently studied using INE-beamline at the ANKA synchrotron light source of the KIT (Germany) dedicated to radionuclide research with X-Ray absorption spectroscopy (XANES/ EXAFS).

Log: 157. **THE POSSIBLE DISCOVERY OF NEUTRON ACTIVATION IN 1910.** Steinhauser, G (1); Merz, S (1); Loeffler, G (2); Adunka, R (3). (1) Vienna University of Technology, Atominstitut; (2) University of Klagenfurt; (3) Carl Auer von Welsbach Museum.

One-hundred years ago, on 21 April 1910, the Austrian chemist Carl Auer von Welsbach published a short comment on a fundamental discovery he had made in the field of nuclear sciences. He reported that “jonium” (Th-230) was able to induce radioactivity in other materials if stored next to the jonium sample. Auer von Welsbach proclaimed that “in this process, a concussion of the elementary inventory of the irradiated samples takes place as well as changes in their chemical properties”. He was well aware that this observation was “not quite in agreement with current theories”, because, as a basic principle, a radioactive substance cannot activate an inactive substance. Accordingly, his notes undoubtedly confirm how he tried to get rid of any “contamination” – but he could not. He found that the previously inactive materials have become radioactive themselves. Auer von Welsbach predicted that this observation “might be of importance for the mysterious field of radioactivity research”. In fact, we believe that in this experiment he incidentally discovered neutron activation and the production of artificial radionuclides (24 years before Irène Curie and Frédéric Joliot) or even induced nuclear fission. We will report on our ongoing efforts to repeat this historical experiment and our hypotheses behind the scenario, which Auer von Welsbach had observed and described in 1910.

Log: 158. **NEUTRON ABSORPTION ACTIVATION ANALYSIS.** Steinhauser, G (1); Merz, S (1); J.H. Sterba (1). (1) Vienna University of Technology, Atominstitut.

In INAA, some elements cannot be quantified despite their large neutron absorption cross section of one isotope, because the nuclear reaction yields a stable nuclide or a radionuclide which is difficult or inconvenient to measure. In particular this refers to lithium (Li-6), boron (B-10), cadmium (Cd-113), gadolinium (Gd-155 and Gd-157). We tested a novel nuclear analytical method with which these nuclides can be determined quantitatively. The principle of the method is that the neutron flux depression in a sample is correlated directly to the concentration of the neutron absorbing nuclides in the sample. The fundamental idea of the proposed method, which we term instrumental neutron absorption activation analysis (INAAA), is to add an activatable nuclide (indicator) to the sample prior to activation and to compare the resulting specific activities of the pure indicator and sample plus indicator via a calibration curve. INAAA follows some principles of both instrumental and prompt gamma neutron activation analysis. A preliminary experiment for boron evidences the validity of this approach. The detection for boron was found in the sub-mg range, which makes INAAA rather a bulk than a trace sensitive method. However, it may find some applications, e.g. in the quality control for classic neutron activation analysis, if the flux depression inside the sample caused by strong neutron absorbers needs to be considered.

Log: 159. **RADIOXENON EMISSIONS BY RESEARCH REACTORS.** Steinhauser, G (1); Saey, P (1); Lechermann (1); Ringbom, A (2); Boeck, H (1); Villa, M (1). (1) Vienna University of Technology, Atominstitut; (2) Swedish Defence Research Agency.

Radioxenon nuclides play a key role in the verification of the Comprehensive Test Ban Treaty (CTBT). As a chemically inert noble gas, the radioxenon isotopes are not retained after underground nuclear weapon tests and can be collected and measured even after long-ranging atmospheric transports. Civil applications of nuclear fission (power plants, isotope production) have been identified as sources of radioxenon which increase the global background. Profound knowledge on the background is important to assess whether a small increase of the activity concentration of a xenon nuclide is due to natural fluctuations or due to a nuclear explosion. In

this study we investigated the radioxenon emissions (Xe-131m, Xe-133, Xe-133m, and Xe-135) of the Atominsitut's TRIGA Mark II research reactor. In two measurement series we could establish a profile of radioxenon nuclides and ratios that are characteristic for our reactor. During these investigations we could identify a leaking fuel element that caused emissions.

Log: 160. **ANALYSIS OF FDA IN-HOUSE FOOD REFERENCE MATERIALS WITH ANTICOINCIDENCE INAA.** David L. Anderson and William C. Cunningham, US Food and Drug Administration, Office of Regulatory Science.

In-House reference material (IRM) Cocoa Powder (CCP) has been in use at US Food and Drug Administration laboratories for about 15 years. A single lot of commercial material was originally characterized for 32 elements by several laboratories and 5 techniques. A unique approach for basis weight determination based upon ambient relative humidity was developed for CCP, eliminating the need for dry weight determinations. The CCP Reference Sheet is updated by incorporating new results approximately every 5 years. The last update occurred in 2006. As part of an effort to revalidate and update values for CCP, anticoincidence instrumental neutron activation analysis (INAA) was used to determine mass fractions for 16 of the originally characterized elements, as well as to provide information on 18 other elements. Results were in very good agreement with 2006 Reference Sheet values. A new candidate IRM, fresh-frozen swordfish (FFSF) powder, was produced by adding inorganic As, Cd, Cr, Hg, Pb, Sb, and Se to liquid nitrogen-frozen commercial swordfish filets which were then homogenized. Portions of FFSF were analyzed by INAA to provide mass fraction and homogeneity information for As, Cd, Cr, Hg, Sb, and Se as well as for 8 other elements occurring naturally in the material. Non-homogeneities were $\leq 2.5\%$ for As, Br, Cs, Cd, Hg and $\leq 1.7\%$ for Cr, Rb, Sb, and Se. Certified reference materials DORM-3 Fish Protein powder and fresh-frozen SRM 1947 Lake Michigan Fish Tissue were analyzed as controls.

Log: 161. **INAA STUDY OF HG, SE, AS, AND BR IRRADIATION LOSSES FROM L-CYSTEINE TREATED AND UNTREATED REFERENCE MATERIALS.** David L. Anderson, US Food and Drug Administration, Office of Regulatory Science.

US Food and Drug Administration in-house reference material (RM) Cocoa Powder and National Institute of Standards and Technology Standard RMs (SRMs) 1515 Apple Leaves, 1547 Peach Leaves, 1571 Orchard Leaves, 1566a Oyster Tissue, and 1568a Rice Flour were co-irradiated together with polyethylene blanks and analyzed for Hg and Se by anticoincidence instrumental neutron activation analysis. The 3 botanical SRM portions showed a combined Hg recovery of 70% while the other portions showed a combined Hg recovery of 169%, indicating that volatile Hg was lost from botanical SRMs and absorbed by the other irradiated portions. Total Hg recovery for all portions was 82%. Se results showed no evidence of cross-contamination and all results agreed with certified and known values. National Research Council of Canada Certified RMs DOLT-3 Dogfish Liver, TORT-2 Lobster Hepatopaneas, and DORM-3 Fish Protein were separately analyzed either with no treatment or after treatment with L-cysteine solutions followed by drying over magnesium perchlorate. Each set of portions was co-irradiated with polyethylene and treated filter blanks. Analysis of all components of each treated portion irradiation package showed that essentially all Hg was retained within the package. Treated DOLT-3 portions (inorganic Hg content 53%) showed a 10-fold improvement with 99% Hg retention. Hg retention for DORM-3 (7% inorganic Hg) was 85% (a 2-fold improvement) while retention for TORT-2 (44% inorganic Hg), was 94%, similar to that for untreated portions (96%). Small irradiation losses ($\leq 0.5\%$) of volatile species of Se, As, and Br were observed.

Log: 162. **ANTICOINCIDENCE INAA CAPABILITIES FOR ANALYSIS OF FDA TOTAL DIET STUDY SEAFOODS.** David L. Anderson, US Food and Drug Administration, Office of Regulatory Science.

Anticoincidence instrumental neutron activation analysis was used to analyze 3 portions each of 5 fresh-weight FDA Total Diet Study seafoods from 3 Market Basket collections for fiscal years

2006-2008. Portions were treated with L-cysteine solutions to enhance retention of Hg during irradiation then dried at room temperature over magnesium perchlorate. Results or limits of detection were obtained for 33 elements. In general, results agreed with those available from FDA's Kansas City field laboratory (KAN-DO). Of 3 shrimp composites analyzed, one showed mass fractions of Ag, Fe, rare earths, U, Th, and Mo significantly higher (up to a factor of 10) than the other 2 shrimp composites. The same shrimp composite showed a lower Hg result (about 50% after accounting for any irradiation loss) compared to the KAN-DO value. This may represent a drying loss. There were other indications of Hg loss during the drying process. SRM 1947 Lake Michigan Fish Tissue, run as a control, yielded an INAA Hg mass fraction 20% lower (corrected for irradiation losses) than the certified value, similar to the difference between the INAA (0.171 mg/kg Hg) and KAN-DO (0.211 mg/kg Hg) results for a TDS canned tuna composite. Because previous studies showed that L-cysteine effectively sequesters inorganic Hg, these discrepancies likely represent methyl mercury loss. INAA results for As, Fe, Rb, and Se were in good agreement with SRM 1947 certified values.

Log: 163. **FEASIBILITY OF FUEL CYCLE CHARACTERIZATION USING MULTIPLE NUCLIDE SIGNATURES.** Kenneth Dayman, University of Texas at Austin; Dr. Steven Biegalski, University of Texas at Austin.

In this paper, the feasibility of identifying spent nuclear fuel arising from an unknown fuel cycle in terms of reactor type, time since discharge, and burnup using a database of the nuclide composition vectors generated for approximately 1,600 different combinations of these variables is examined. The database and test cases were generated using Origen-ARP, and in contrast to currently implemented methods that rely on a select number of isotopic ratios, the concentrations of 200 nuclides were analyzed for each sample. Different techniques were used to make predictions including finding the database entry most similar to the unknown by finding the minimum squared or absolute value difference between the unknown's composition vector and the database entries. To make burnup predictions regardless of the reactor type, enrichment, and cooling time, a regression model between the composition vector and fuel burnup was developed using the database as a training set. Nuclides that exhibited high variance as the reactor type, burnup, and cooling time were varied were identified by principle components analysis and the predictive power of the aforementioned approaches was evaluated using these truncated composition vectors. An initial estimation of the techniques' sensitivity to measurement error was made by perturbing different subsets of the unknown's composition vector and examining the effect on each of the technique's predictions. We demonstrate through the results of these experiments that investigation and development of methodologies using a large number of nuclide signatures for nuclear forensics is warranted.

Log: 164. **TRACER EXPERIMENTS DETERMINING COLUMN CHARACTERIZATION AND SEPARATION OF STRONTIUM AND ZIRCONIUM.** Roger Martinelli, Terry Hamilton, Steve Kehl, Scott Tumey, and Tom Brown; Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, PO Box 808, Livermore, CA 94550, USA.

Accelerator Mass Spectrometry (AMS) is emerging as a main stream technology for low-level detection and measurement of a wide range of different radionuclides. These technologies provide potential for improved detection sensitivity, ease of sample preparation, and reduction in measurement time compared with traditional or classical decay-counting techniques. We are attempting to expand on the measurement capabilities at Lawrence Livermore National Laboratory (LLNL) to include routine environmental and bioassay measurements of strontium-90 (^{90}Sr) based on Accelerator Mass Spectrometry. The Minimal Detectable Amount (MDA) for ^{90}Sr AMS is around 0.1 mBq. The most significant limitations on this measurement technology are (i) isobaric interferences caused by the presence of zirconium at mass 90; and (ii) the amount of stable strontium that can be loaded into the sample target. In this study, we designed a series of tracer experiments using strontium-85 (^{85}Sr) and zirconium-95 (^{95}Zr) to optimize conditions for the chemical separation and preferential column elution of strontium in the presence of added zirconium. We also report on preferred methods for ^{90}Sr AMS target preparation, and on the

stable strontium content and exogenous sources of zirconium in chemical reagents and different matrix types. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 165. **CALCULATING INGESTION DOSES FROM CESIUM-137 USING A WEB-BASED APPLICATION DEVELOPED UNDER THE MARSHALL ISLANDS PROGRAM**. Steve Kehl, Terry Hamilton, Roger Martinelli, Michael Tamblin, Sandra Peters, and Amy Simpson; Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, PO Box 808, Livermore, CA 94550, USA.

Assessment and Radioecology Program at the Lawrence Livermore National Laboratory (LLNL) has developed a series of interactive internet applications to provide the public with a free and open platform to learn more about radiological conditions in the Marshall Islands. The application outlined in this paper describes one feature whereby local atoll residents as well as potential visitors can calculate their hypothetical internal dose based on interactive user inputs matched to environmental data on the ^{137}Cs contents of subsistence food products. User inputs include the date, location, food type, numbers of servings, and dose type. The dose type function allows for assessments of "single intakes", "annualized intakes" or model dietary intakes based on "standard living patterns". The application computes dose in millisievert (mSv) and millirem (mrem) (for an adult) in table format, showing the island's name, ID, atoll name, date, and dose "intake" type selected. Environmental data is decay corrected to the date entered by the user using an effective half-life of ^{137}Cs of 8.5 years (<https://marshallislands.llnl.gov>). This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 166. **DISTRIBUTION OF CESIUM-137 IN COCONUT (COCOS NUCIFERA L.) ON RESIDENCE ISLANDS IMPACTED BY THE NUCLEAR TEST PROGRAM IN THE NORTHERN MARSHALL ISLANDS**. Sandra Peters, Steve Kehl, Roger Martinelli, Mike Tamblin, and Terry Hamilton; Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, PO Box 808, Livermore, CA 94550, USA.

Cesium-137 (^{137}Cs) is the highest contributor to nuclear test-related dose to people exposed to residual nuclear fallout contamination in the Marshall Islands. The primary exposure pathway is ^{137}Cs ingestion of locally grown products such as drinking coconut meat and juice, copra meat, Pandanus, and breadfruit. The Marshall Islands Program recently completed a series of radiological surveys. The data and information is used to improve predictive dose assessments accuracy and provide assurances to population groups that conditions comply with national and international radiological safety and health standards. Coconut samples are separated into meat and juice in the field. Sample material is shipped frozen to Lawrence Livermore National Laboratory. Coconut meat samples are dried, homogenized, packed into standardized geometry and then counted by gamma-spectroscopy. Coconut juice samples are processed by co-precipitation of cesium isotopes onto ammonium molybdophosphate (AMP) using cesium-134 (^{134}Cs) as an isotope dilution spike. The AMP is then separated by centrifugation and counted by gamma-spectrometry. The highest levels of ^{137}Cs are observed on Bikini Island and then decrease between Bikini, Rongelap, Utrök, and Enewetak (including Medren and Japan) Islands in significant incremental steps with median values in drinking coconut juice ranging between 400, 20, 5 and 1 Bq kg⁻¹, respectively. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 167. **OPTIMIZATION OF A PROMPT GAMMA ANALYSIS SETUP FOR ANALYSIS OF ENVIRONMENTAL SAMPLES**. Fares A. Al-Matouq(1), A. H. Isab(2), Khateeb-ur-Rehman (1), M. A. Gondal (1), M. Raashid (1) and M. Dastageer(1), (1)Department of Physics and (2)Department of Chemistry, King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia.

Thermal neutron capture based Prompt Gamma ray Activation Analysis (PGNAA) setup has been optimized to analyze elemental concentration of toxic elements in liquid and solid bulk samples using a D(d,n) reaction based portable neutron generator . Two different types of source-sample geometries namely source-in-moderator and sample-in-moderator were tested. For both cases moderator size was optimized through Monte Carlo simulations. Performance of both the setup were tested through concentration measurement of mercury in Hg-contaminated water samples using a cylindrical 4 inches x 4 inches (diameter x height) bismuth germinate (BGO) gamma ray detector. Yield of Hg prompt gamma ray were measured from water samples containing known concentration of mercury. Experimental yield of prompt gamma ray from Hg contaminated water samples have an excellent agreement with theoretical yield obtained through Monte Carlo simulations. Performance comparison of both the setup will be presented.

Log: 168. **CHLORINE DETECTION IN BFS CONCRETE SAMPLES USING A PORTABLE NEUTRON GENERATOR.** Zameer Kalakada(2), O.S. B. Al-Amoudi (2) , M. Maslehuddin (3) , M. Raashid (1) and Khateeb-ur-Rehman(1). (1)Department of Physics, (2) Department of Civil Engineering and (3)Center for Engineering Research, King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia.

A prompt gamma ray neutron activation (PGNAA) setup has been developed utilizing a D(d,n) reaction based portable neutron generator model MP320 (Thermo Fisher Company, USA) to analyze elemental concentration of concrete structure. The main features of the PGNAA setup is its source - detector geometry. In this setup the moderated neutron source and shielded gamma ray detector are placed side by side to detect prompt gamma rays emitted from bulk samples at backward angles. Inspite of lower yield of gamma rays at backward angles, this type of PGNAA setup geometry is highly desired for a PGNAA based portable concrete chloride analyzers. The design of the setup was optimized through Monte Carlo simulations. The setup has been tested through chlorine detection in chloride-contaminated blast furnace slag (BFS) cement concrete samples. Inspite of strong interference between chlorine prompt gamma rays and concrete constituents prompt gamma rays, chlorine concentration was successfully determined in BFS based concrete samples using 6116 and 3935 keV chlorine prompt gamma rays. An excellent agreement has been observed between the experimental and theoretical yield of 6116 and 3935 keV chlorine prompt gamma rays. Results of this study will be presented here.

Log: 169. **CARBON AND OXYGEN DETECTION IN BULK SAMPLES USING PROMPT GAMMA RAYS.** Fares A. Al-Matouq (1), A. H. Isab (2), M Raashid (1), Khateeb-ur-Rehman (1) and F. Z. Khiari(1). (1)Department of Physics and (2) Department of Chemistry, King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia.

A prompt gamma ray setup has been developed to determine carbon and oxygen concentration in organic and inorganic bulk materials samples using KFUPM 14 MeV neutron generator. The setup is based on detection of carbon and oxygen prompt gamma rays produced due to inelastic scattering of 14 MeV neutrons from bulk materials samples. Carbon and oxygen elemental concentrations in H₂O, H₂O₂, carbon, C₄H₄ bulk samples. were determined through 4.44 and 6.15 MeV prompt gamma rays yield measurements respectively using a cylindrical 5 inches x 5 inches (diameter x height) bismuth germinate (BGO) detector. Preliminary results of the performance tests of the setup will be presented here.

Log: 170. **RADIONUCLIDE ACTIVITY CALCULATIONS USING THE LARGE-VOLUME GAS IRRADIATION PROCEDURE DEVELOPED AT THE UNIVERSITY OF TEXAS.** Egnatuk,C. M.(1);Biegalski, S. R.(1) (1. The University of Texas at Austin).

The University of Texas Nuclear Engineering Teaching Laboratory produces a radioxenon and radioargon samples with a volume of approximately 0.5 cc. In order to produce a detectable amount of Ar-42, a new procedure that allows a large amount of gas to be irradiated. This new procedure allows for the placement of approximately 1.4 L of gas in the core, with 0.6 L next to

the 15" long fuel elements. Using a flux profile created in MCNPX, the possible activity production of several noble gases was calculated. The irradiation time, decay time, and irradiation power was also calculated. The model seeks to maximize the activity production of the desired nuclide while minimizing irradiation time.

Log: 172. **USE OF RARE-EARTH ELEMENTAL IMPURITY PATTERNS FOR ORIGIN ASSESSMENT OF URANIUM MATERIALS.** Perkin, MV; Zhao, Ke; Fischer, DM. International Atomic Energy Agency, Department of Safeguards..

Determination of the origin of uranium materials is not only essential for authentication studies in nuclear forensics, but can also support nuclear safeguards verification aimed at assessing a State declaration about source materials used for conversion into nuclear-grade products and address potential substitution of such materials. It has been recently suggested that patterns of the rare-earth element (REE) traces can be used, among other parameters, for the origin assessment of uranium ore concentrates (yellow cakes). As the chemical properties of the REE are, in general, very similar, their relative abundances can be expected to remain mainly unaltered during concentration of uranium and its further conversion, assuming that the REE impurities are not re-introduced during the processing. A study is presented that demonstrates how REE patterns were utilized to resolve a safeguards case where the provenance of yellowcake was in question. Moreover, as the verification task required also assessing the State declaration about source material used in the conversion experiments, it was shown that the REE patterns can represent a key signature of the material processed, while analysis of other elemental impurities in the feed and product samples provided inconclusive information.

Log: 173. **US IMS RADIONUCLIDE MONITORING NETWORK COMPLETION AND SUSTAINMENT.** Shipman, GR, Carty, RF, Comes, LR. GDAIS.

This presentation reviews the current status and performance of the US Operational Systems for Radionuclide Monitoring, including both particulate and noble gas, that support the International Monitoring System (IMS). Also described is the implementation of changes in approach to operations, maintenance and life cycle sustainment driven by anticipated Entry Into Force (EIF) requirements. The US operates eleven radionuclide monitoring stations under the IMS. All eleven are equipped with Radionuclide Automated Sampler/Analyzers (RASA) systems for particulate monitoring. Four are also equipped with the Swedish Automatic Noble Gas Analyzer (SAUNA-II) Systems. Presentations at MARC VII and VIII described the ongoing installation, certification and operation of the radionuclide monitoring systems and development of a Life Cycle Sustainment Plan for long term support. This presentation will address the completion of the network, installation and certification of the noble gas systems and implementation of various elements of the Sustainment Plan designed to mitigate the effects of obsolescence and keep the systems operating to CTBT EIF standards.

Log: 174. **COMPOSITION OF URANIUM OXIDE PARTICLES USING TOF-SIMS.** Hocking, HE (1); Burggraf, LW (1); Xiaofeng F. Duan (2); Gardella JA (3); Yatzor, BP (3); Schuler WA (4). (1) AFIT, (2) Air Force Research Laboratory, (3) University at Buffalo SUNY, (4) National Air and Space Intelligence Center.

Particles of uranium oxides, including natural and depleted UO_2 , U_3O_8 , and UO_3 were measured using an IONTOF TOF-SIMS V instrument. Samples of particles having diameters about tens of micrometers were mounted on gold substrates made using a novel technique that reduced hydrocarbon contamination and volatile outgassing. This mounting method also provided an internal mass standard of Aux ion peaks to calibrate high masses. For UO_2 surfaces the dominant U_3O_x and U_4O_y cations were U_3O_6^+ and U_4O_8^+ , whereas for both U_3O_8 and UO_3 surfaces they were U_3O_7^+ and U_4O_9^+ . For the observed cation masses, the structure and energy of the most stable isomers were determined by density functional theory (DFT) calculations with a stochastic searching algorithm and then refined by a high-level DFT calculation. Cation isomers having high oxygen content were doublet spin states and those having low oxygen content were

quartet spin states. Observed secondary ion abundance ratios were well correlated with calculated cluster relative stabilities. TOF-SIMS depth profile trends for “protonation” ratio and for Benninghoven’s “lattice valence” parameter were used to distinguish spectra of U_3O_8 from UO_3 . Cations containing a greater number of uranium atoms were found to have a lower protonation ratio. UO_2 and U_3O_8 surfaces show a steeper reduction in protonation ratio compared to UO_3 surfaces which exhibit a nearly constant near-surface protonation ratio followed by a more gradual smaller decline with depth. In future work we plan to interpret “lattice valence” concept for secondary ion distributions by analyzing DFT calculated electron affinities, ionization potentials and cluster relative stabilities for uranium oxide clusters.

Log: 175. **THE CASE FOR FRACTIONAL SOLUBILITY PROFILES.** R.L. Metzger¹ and K.F. Eckerman², 1) Radiation Safety Engineering, Inc, 2) Oak Ridge National Laboratory.

Default values for the solubility of various compounds in lung serum are provided in ICRP and NRC ALI tables. These estimated solubility profiles are conservative, and just list the dominant solubility class (Fast, Medium, or Slow) for the radionuclide. In practice, solubility profiles for many compounds, both natural and man-made, are complex, with a fraction of the compound in each Class. Only radiopharmaceuticals and soluble compounds of tritium and iodine can be accurately assumed to be solely in one solubility class (F). The assumption of a single solubility class for airborne distributions of solid particles can introduce order of magnitude errors in internal dosimetry calculations. The problem is particularly acute for isotopes with dual toxicity (e.g. Uranium which is both nephrotoxic and radiotoxic), and in dosimetry estimates where only one bioassay measurement is available. For inhalation exposures resulting from an accident, treatment decisions must frequently be made quickly to be effective. While much work has been done to develop rapid bioassay methods that will provide bioassay data in a clinically useable timeframe, little consideration has been given to the massive errors that can be introduced into the dosimetry estimates by use of the default solubility profiles where only one bioassay measurement is available.

Log: 176. **PREPARATION OF RADIO-SM BY NEUTRON ACTIVATION FOR ACCELERATOR MASS SPECTROMETRY.** Graham, J (1); Buchholtz, B (2); Biegalski, S (1). (1) Nuclear Engineering Teaching Laboratory, The University of Texas at Austin, Austin, TX 78758 (2) Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, Livermore, CA 94551.

Field measurement of isotopic ratios may be used to fingerprint an element's origin, be it from commercial power, industrial, medical or historical weapons fallout. Samples of Samarium radionuclides were prepared by neutron activation for subsequent analysis using accelerator mass spectrometry (AMS). High purity Samarium(III) oxide powder was irradiated in the University of Texas at Austin TRIGA reactor to a total neutron fluence of $3.3 \times 10^{14} \text{ cm}^{-2}$. An initial determination of the isotopic ratios was made using activation calculations with a BURN card in an MCNP model of the TRIGA core. Experimental validation of the MCNP results was achieved by analyzing gamma spectra of the irradiated oxide powers at various time intervals after irradiation. Subsequent measurement of ^{151}Sm was conducted at the CAMS facility at LLNL demonstrating the first measurement of this isotope at this facility. This work performed in part under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 177. **USE OF GAMMA SPECTROSCOPY IN THE MO-99 PRODUCTION IN LOW ENRICHED URANIUM SAMPLES.** Rawool-Sullivan M (1), Bounds J (1), Hutchinson J. (1), Garner S. (1), Myers S. (1), May I. (1) and Dale G. (1); (1) Los Alamos National Laboratory.

Results are presented of the gamma spectroscopy performed in Mo-99 production experiments being conducted at the Los Alamos National Laboratory as a part of the National Nuclear Security Administration's Molybdenum-99 Production Program. Nine-MeV bremsstrahlung X-rays from a Linatron were incident on heavy water to produce neutrons for irradiation of low-enrichment

uranium solutions. The thermal neutron flux was measured using bare and cadmium covered gold foils located near the solutions. The Mo-99 production in each sample was calculated using post-irradiation gamma measurements with a high purity germanium detector. The total number of fissions in each irradiated sample was also determined. Other prominent fission radionuclides were also observed and their quantities estimated.

Log: 178. **ARSENIC COMPOUNDS IN FIREWORKS? - AN ANALYSIS OF THE SUSPENDED PARTICLE EMISSION DURING VIENNA'S NEW YEAR FIREWORKS.** Johannes H. Sterba(1), Fritz Grass(1), Georg Steinhäuser(1), Johann Wernisch(2), Max Bichler(1); (1): Atominstitut, Vienna University of Technology, Stadionallee 2, 1020 Wien, Austria; (2): Institute of Solid State Physics, Vienna University of Technology.

To prove the applicability of a fine dust sampling method based on electrically charged plates, dust was collected and analysed during the New Year's fireworks in Vienna in the years 2007 and 2008. Apart from the expected increase in firework related elements (Ba, Sr, Al, K), a significant increase in the As concentration of the samples was measured. Indeed this increase can also be found in the literature but stands, up to date, uncommented. This is probably due to the instinctive dismissal of elevated As concentrations as a contamination of some other firework components. The ratios of those components versus the As concentrations measured clearly points to a different source. Considering the notorious difficulty to produce a brilliant blue light in pyrotechnics without the use of As compounds, we propose that arsenic compounds are intentionally added to cheaply produced fireworks. While the measured concentrations in the fine dust are too low to constitute health risks for the audience of fireworks, the working conditions in the major firework producing countries are not necessarily safe in this respect.

Log: 179. **DISSOLUTION AND DETERMINATION OF NEPTUNIUM IN RADIOACTIVE MATERIALS.** Xu, N.; Colletti, L.; Gallimore, D.; Garduno, K.; Hahn, T.; Lujan, E.; Martin, F.; Martinez, A.; Miller, J.; Porterfield, D.; Rodriguez, J.; Lisa Townsend, L.; Laurie Walker, L.; Tandon, L.; Kuhn, K.; Los Alamos National Laboratory .

Neptunium (Np) oxide dissolution and the subsequent Np determination have not been thoroughly studied due to its high radioactivity. Np is historically analyzed by radiochemical methods such as alpha and gamma spectroscopy. However, the sample preparation procedure is time consuming, tedious and highly dependent on the experience and skill level of the analyst. In this study, several novel methods to measure Np concentration, such as ICP-MS, ID-ICP-MS, and Controlled Potential Coulometry were evaluated and the results were compared with radiochemistry methods. A method to dissolve a neptunium oxide was also discussed. ICP-MS is a technique used for multi-elemental analysis at trace levels including ^{237}Np . The accuracy and precision of the ICP-MS method for trace levels of ^{237}Np analysis in Pu metal was studied. (LA-UR 11-06517)

Log: 180. **A NOVEL PATH IN PARTITIONING: WATER-SOLUBLE BTP LIGANDS FOR THE INNOVATIVE SANEX PROCESS.** C. M. Ruff^{1,2}, U. Müllich², A. Geist², P. J. Panak^{1,2}, 1-University of Heidelberg, 2-Institute of Nuclear Waste Disposal, KIT-Campus Nord.

With regard to an efficient partitioning process the separation of the trivalent minor actinides from the chemically similar fission lanthanides is one of the key issues. This separation process requires highly effective extracting agents. Alkylated bis(1,2,4-triazin-3-yl)pyridines (BTP) and alkylated bis([1,2,4]triazin-3-yl)-[2,2']bipyridines (BTBP) are among the most promising SANEX extracting agents, selectively extracting trivalent actinides from nitric acid solutions. Based on the conventional SANEX process an innovative SANEX (i-SANEX) process has been developed, combining the SANEX with the previous DIAMEX step. Hydrophilic aq-BTP is used as aqueous complexing agent for selective back extraction of actinides from organic phases loaded with trivalent actinides and lanthanides. Our studies focus on the complexation of trivalent actinides and lanthanides with aq-BTP (a) to gain fundamental knowledge on the unique properties of BTP, (b) to compare BTP with aq-BTP, and (c) to determine complex formation constants for trivalent

actinides and lanthanides required for future process development. In the present work complexation studies are performed with Cm(III)/Eu(III) in H₂O and HNO₃ using time-resolved laser fluorescence spectroscopy (TRLFS). With increasing ligand concentration the following complexes are identified and spectroscopically characterised: [M(aq-BTP)], [M(aq-BTP)₂], and [M(aq-BTP)₃] (M: Cm(III), Eu(III)). From our spectroscopic results the stability constants and thermodynamic data of the stepwise complex formation are derived. A comparison of the log K values of the [M(aq-BTP)₃]-complexes (M = Eu(III), Cm(III)) shows a significantly higher stability constant for trivalent actinides (difference: 2.7 orders of magnitude), which is in perfect agreement with results from liquid-liquid extraction tests.

Log: 181. TRACE ELEMENT DETERMINATION IN A MUSSEL REFERENCE MATERIAL USING SHORT IRRADIATION INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS.

Moreira, EG(1); Seo, D(1); Vasconcellos, MBA(1); Saiki, M(1). (1) IPEN - CNEN/SP.

The production of certified reference materials in Brazil, and the consequent availability to national end users, is an important task for the enhancement of Metrology in Chemistry status in the country, as these materials are used for method validation, equipment calibration and for establishing metrological traceability links. In this study, Instrumental Neutron Activation Analysis (INAA) was applied to the determination of bromine, chlorine, magnesium, manganese, potassium and vanadium in a mussel reference material produced at IPEN - CNEN/SP. For the determination of these elements via the comparative INAA method, the respective analytical radionuclides, ⁸⁰Br, ³⁸Cl, ²⁷Mg, ⁵⁶Mn, ⁴²K and ⁵²V, are short lived and then, short irradiations are used. The main advantage over longer irradiation methods is the faster output of analytical results. Six subsamples from two bottles of the Perna perna mussel reference material were analyzed. Each subsample was simultaneously irradiated with elemental standards for 10 s at the IEA - R1 research nuclear reactor through a pneumatic transfer system. After suitable decay periods, gamma radioactivity measurements were carried out, using a hyperpure germanium detector. The accuracy of the method was checked by using the NIST SRM 1566b - "Oyster Tissue" certified reference material. The comparison of the results obtained in this study to the robust mean of the interlaboratorial collaborative trial used for the characterization of the mussel reference material was performed via simple z-score tests. The comparison showed that the short irradiation INAA method is suitable for the characterization of new reference materials.

Log: 182. A CRYSTALLOGRAPHIC AND KINETIC INVESTIGATION OF METHANOL SUBSTITUTION KINETICS OF RE(I) TRICARBONYL COMPLEXES.. Schutte, M. (1), Visser, H.G. (1), Roodt, A. (1), (1) University of the Free State.

It has been shown by various studies that the aqua ligand in the iso-electronic [MX(H₂O)(CN)₄]_n complexes (M = Mo(IV), W(IV), Tc(V), Re(V); X = O²⁻, N³⁻) are all labile towards ligand substitution. To completely understand the reactivity and reaction mechanisms of the Manganese triad, the fac-[M(CO)₃]⁺ (M = ^{99m}Tc, ¹⁸⁸Re) core has been a continuing interest, mainly because of its potential biomedical properties. fac-[M(CO)₃(H₂O)₃]⁺ is a very versatile synthon with its low-valent, kinetic inert organometallic core and therefore high in vivo stability. The '2+1' approach is of big interest where the bidentate ligand, of which the lipophilicity can be controlled, acts as chelator and the monodentate bifunctional that acts as a potential linker to a biologically active molecule. Alberto et al focussed on the pH dependence of different polymeric species of [Re(CO)₃(H₂O)₃]⁺ but very few kinetic studies have been undertaken on these fascinating complexes. In vivo, the third coordination site in these compounds are also exposed to possible nucleophilic attack by ligands in the blood stream such as NCS⁻ and halides; thus it is imperative to investigate the reactivity and kinetics of these systems to potential entering ligands. The current available knowledge of substitution kinetics of Re(I) tricarbonyl complexes were expanded to a great extent and it contributed to the pool of available crystallographic data of these systems.

Log: 183. **HIGH-CAPACITY LAB-ON-VALVE BEAD INJECTION EXTRACTION CHROMATOGRAPHY FOR RAPID DETERMINATION OF PLUTONIUM IN HUMAN URINE.** Qiao, JX(1); Hou, XL(1); Miró, M(2); Cerdà, V(2); Roos, P(1). (1)Risø-DTU, (2)UIB.

A novel system exploiting high-capacity-column bead injection coupled with inductively coupled plasma mass spectrometry detection was developed for rapid and automated determination of plutonium in biological samples, namely, human urine. A renewable mini-column incorporated within the lab-on-valve bead injection framework was loaded with a defined amount of extraction chromatographic resin (TEVA) through programmable bead transport. Selective capture of plutonium was then performed within the mini-column after the sample pretreatment. Different sample pre-treatment methods were also investigated and compared, including evaporation, co-precipitation with calcium phosphate, iron hydroxide or manganese dioxide and their combination. The analytical results demonstrated high inner capacity of solid-phase extraction beads and good reproducibility of column renewal (0.305 ± 0.002 g) with the competence to process large-sized samples, e.g., 1 L of human urine. The chemical yields of plutonium could reach up to 95% under the optimal condition and the column separation could be accomplished within a short timeframe (< 1 h). Therefore the developed system is suitable for the rapid analysis of low-level plutonium in human urine samples as required in emergency situations.

Log: 184. **QUANTITATIVE ANALYSIS OF TRIVALENT URANIUM AND LANTHANIDES IN A MOLTEN CHLORIDE BY ABSORPTION SPECTROPHOTOMETRY.** Fujii, T(1); Uda, T(1); Fukasawa, K(1); Uehara, A(1); Sato, N(2); Nagai, T(3); Kinoshita, K(4); Koyama, T(4); Yamana H(1). (1) Kyoto University, (2) Tohoku University, (3) Japan Atomic Energy Agency, (4) Central Research Institute of Electric Power Industry.

As an analytical application for pyrochemical reprocessing using molten salts, quantitative analysis of uranium and lanthanides by UV/Vis/NIR absorption spectrophotometry was performed. Electronic absorption spectra of LiCl-KCl eutectic at 773 K including trivalent uranium and eight rare earth elements (Y, La, Ce, Pr, Nd, Sm, Eu, and Gd as fission product elements) were measured in the wavelength region of 300-2200 nm. The composition of the solutes was simulated for a reductive extraction condition in pyroreprocessing process of spent nuclear fuel, that is, about 2 wt% U and 0.1-2 wt% rare earth elements. Since U(III) possesses strong absorption bands due to d-d transitions, an optical quartz cell with short light pass length of 1 mm was adopted in the analysis. The quantitative analysis of U(III) was found to be possible with its f-f transition intensities at the NIR region. A few lanthanides (Pr, Nd, and Sm) showed absorption peaks in this region, and the analytical uncertainty for these elements was also investigated.

Log: 185. **NUCLEAR FIELD SHIFT EFFECT IN ISOTOPE FRACTIONATION OF THALLIUM.** Fujii, T(1); Moynier, F(2); Agranier, A(3); Ponzevera, E(4); Abe, M(5); Uehara, A(1); Yamana, H(1), (1) Kyoto University, (2) Washington University in Saint Louis, (3) Institut Universitaire Européen de la Mer (4) Institut Français de Recherche pour l'Exploitation de la Mer, (5) Tokyo Metropolitan University.

Thallium is a highly toxic heavy element. Pollution by Tl is found in mining districts and its migration introduces Tl into surface water, soils, plants, and so on. Environmental transport of Tl is affected by chemical exchange reactions of related Tl species, that is, redox reaction between Tl(I) and Tl(III) and ligand exchange reactions of them. In order to deepen the knowledge of Tl chemistry, we investigated fractionation of Tl stable isotopes (^{205}Tl and ^{203}Tl) in a chemical exchange system. Tl isotopes were fractionated in a liquid-liquid extraction system, in which aqueous and organic phases are hydrochloric acid solution and dichloroethane including a crown ether, respectively. This method is known to be effective for finding isotope fractionation. After purification by ion-exchange chemistry, the isotope ratio of $^{205}\text{Tl}/^{203}\text{Tl}$ in equilibrated aqueous phase was measured precisely by multi-collector inductively coupled plasma spectrometry (MC-ICP-MS) at the "Pole Spectrometry Ocean" (PSO) of Brest. A large isotope fractionation $>1\%$ was found. In order to check the validity, electronic structures of possible Tl species (hydrated Tl^+ , Tl_3^+ , and Tl chlorides) were calculated by ab initio methods, and the isotope fractionation factor

was theoretically obtained. The isotope fractionation via intramolecular vibrations was calculated to be much smaller than the experimental result. The isotope fractionation via isotopic change in nuclear volume, named the nuclear field shift effect, was calculated to be ~1‰ in Tl(I)-Tl(III) redox systems and/or ligand exchange systems of Tl(III).

Log: 186. **ASSESSMENT OF DIFFERENCES IN THE TRACE ELEMENT CONCENTRATIONS IN REGIONS OF HUMAN BRAIN BY INAA.** Saiki, M(1); Miranda, N(2); Leite, REP(2); Genezini, FA(1); Grinberg, LT(2); Ferretti, REL(2); Jacob-Filho, W(2). (1) Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP. (2) Faculdade de Medicina da Universidade de São Paulo.

Studies have shown that there is a potential relationship between the levels of trace elements in cerebral tissues and neurological disorders. However, there are few publications available to the elemental composition of this tissue as well as for different regions of the brain. The aim of this study was to investigate trace element differences in various human brain regions of an elderly population of normal individuals. This project was approved by the Ethic Committee and brain samples (n=18) from individuals of both genders aged 51-95 years were provided by the Brain Bank of the Brazilian Aging Study Group of the São Paulo University. The tissues from the regions of hippocampus, frontal, parietal, temporal, occipital lobes, cerebellum and amygdale were cut using a titanium knife, ground, freeze-dried and then analyzed by instrumental neutron activation analysis (INAA) technique. Samples and element standards were irradiated with a neutron flux at the IEA-R1 nuclear research reactor for Br, Fe, K, Na, Rb, Se and Zn determinations. Student's t test (p=0.05) used to compare the results showed, for several element concentrations, significant difference among the brain regions that were analyzed. Most of our results agreed with the published literature data. Biological standard reference materials SRM 1566b Oyster Tissue and SRM 1577c Bovine Liver analyzed for quality control indicated good accuracy of the results.

Log: 187. **CONTRIBUTIONS TO THE GLOBAL ³⁷Ar BACKGROUND BY RESEARCH REACTOR OPERATIONS.** Fay, A(1); Biegalski, S(1). (1) University of Texas at Austin.

The Comprehensive Nuclear-Test-Ban Treaty establishes a number of instruments for detecting nuclear explosions and verifying the compliance of member states. Among these instruments is On-Site Inspection (OSI), the deployment of an inspection team to the suspected test site. Recent work has shown that an underground nuclear explosion would produce ³⁷Ar, through the ⁴⁰Ca(n, α)³⁷Ar reaction, in sufficient concentrations to be used as a detectable signature of a weapon test. This ³⁷Ar signature would be quantified by subsurface sampling at a suspected test site by an OSI team.

In this paper, radiation transport simulation of the University of Texas at Austin 1.1 MW TRIGA reactor was conducted to determine the ³⁷Ar/⁴¹Ar production ratio. This ratio was used, along with published ⁴¹Ar release data for the US research reactor fleet, to approximate the total ³⁷Ar source term from research reactor operations. Using this source term, atmospheric transport modeling was then conducted to calculate the expected contribution to ³⁷Ar background at potential OSI sites.

Log: 188. **CHEMICAL SPECIATION FOR FORENSIC SCIENCE.** Marianne P. Wilkerson, Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545 – USA.

We present the application of chemical speciation for forensic science. For many years, environmental research from U.S. National Laboratories has focused on the chemical speciation of actinides in contaminated soils and groundwaters from former U.S. Nuclear weapon's production sites or disposal areas such as Rocky Flats, Hanford, or the Nevada Test Site. These findings add to a body of observations from around the world where transport of Pu can be traced to particles and colloids as in the case of groundwater transport of Pu from the Mayak Production Association in Russia. Extensive research has shown that particle matrix and radionuclide composition are dependent upon source, and structures and oxidation states are correlated to release conditions. Here, we will discuss our work on bulk and particle measurements on

environmental samples and bulk materials for the application of chemical speciation for forensic science. LA-UR 11-06789

Log: 189. **RADIOSTRONTIUM IN THE WESTERN NORTH PACIFIC: EFFECTS OF THE FUKUSHIMA DAIICHI NUCLEAR PLANT ACCIDENT.** Hirose, K(1); Aoyama, M(2); Povinec, PP(3). (1) Sophia University, (2) Meteorological Research Institute, (3) Comenius University.

Large amounts of radioactivity were emitted to the atmosphere after the Fukushima Daiichi NPP accident. Major radionuclides were radioiodine and radiocesium (^{134}Cs , ^{137}Cs), whereas radiostrontium (^{89}Sr , ^{90}Sr) was a minor component. On the other hand, significant amounts of radioactive wastewaters produced as a result of cooling of reactors were directly released into the ocean. Since the wastewater directly interacted with ruptured fuel rods, it is likely that large amounts of radiostrontium were released into the ocean. In order to assess oceanic effects of the Fukushima-derived radiostrontium, it is important to better understand long-term behavior of ^{90}Sr in the western North Pacific waters. We examine the long-term changes of surface ^{90}Sr in the subtropical waters (25° – 36° N) including Kuroshio Current water and mixed region (36° – 45° N), which corresponds to the east area of the Fukushima NPP. The ^{90}Sr concentrations in surface waters of both regions decreased gradually, although a peak in the surface ^{90}Sr concentration appeared in the early 1990s. The present level of the surface ^{90}Sr concentration, including coastal waters near Fukushima, is around 1 Bq m^{-3} . After the direct release of the radioactive wastewater to the ocean, high ^{90}Sr concentrations in waters near the Fukushima NPP site, from 0.18 to 7.7 kBq m^{-3} , were observed (as was the detection of ^{89}Sr), although measurements of radiostrontium were very limited comparing with ^{137}Cs . It is necessary to do more precious measurements of radiostrontium in the ocean, in order to assess the radiological effects of the Fukushima-derived radiostrontium.

Log: 190. **A NEW METHOD FOR THE DETERMINATION OF LOW-LEVEL ACTINIUM IN GEOLOGICAL SAMPLES.** Henrieta Dulaiova, University of Hawaii, Department of Geology and Geophysics.

We developed a new, simple method for the determination of Ac-227 in geological samples. The method uses extraction chromatographic techniques and alpha-spectrometry and is applicable for a whole range of natural matrices. Here we report on the procedure and results of the analysis of water and basalt samples. Water samples were acidified and rock samples underwent total dissolution via fusion and acid leaching. All samples were spiked using a NIST certified Th-229/Ac-225 tracer. Actinides were pre-concentrated via lead sulfate co-precipitation. The precipitate was dissolved in 4M HCl and run through a DGA (N,N,N',N'-tetra-n-octyldiglycolamide) extraction chromatographic column (Eichrom; Horwitz et al., 2005). Actinides are well retained on the column while radium and other alkaline earth elements have no affinity and pass through the column without retention. Actinium is eluted with 2 M HCl which is efficient for Ac elution but leaves Th and other actinides retained on the DGA column. The actinium fraction was prepared for alpha spectrometric measurement via cerium fluoride micro-precipitation (Dulaiova et al., 2001). Recoveries of water samples were $80 \pm 8 \%$ ($n=14$) and $70 \pm 12 \%$ for basalts ($n=30$). For rock samples the minimum detectable activities were 0.003 dpm/g with sample activities ranging from 0.01 to 0.5 dpm/g . For water samples the limits of detection were similar. From the analysis of several standard rock samples with the method we found very good agreement between our results and certified values.

Log: 191. **WHOLE BODY COUNTING FOR TIME-DEPENDENT RADIOACTIVITY ESTIMATION IN PATIENTS UNDERGOING ^{177}Lu PRRT.** Bode, P(1); Liu, B(1); Breeman, WAP(2). (1) Delft University of Technology, (2) Erasmus Medical Center Rotterdam.

Neuroendocrinal tumors can be treated using ^{177}Lu -DOTA-tate, since this radiopharmakon enters the tumorcell via the somatostatine receptor. The amount of ^{177}Lu radioactivity in the body has to be known accurately for assessment of the interne radiation dose and for evaluation of the effectiveness of the therapy. For long, the excreted urine was collected in a flask and the amount

of radioactivity was determined using a dose calibrator. There are several disadvantages of this method, such as spillover resulting in incomplete collection, feminine unfriendliness and unacceptability amongst certain ethnic populations. Moreover, this method has to be left for radiological safety considerations. A whole body counting technique (WBC) was developed. Two opposite WBC measurements from anterior and posterior views were carried out with a NaI(Tl) spectroscopic probe positioned 4 m from the patient. The individual reference response by the baseline measurement before the first voiding was used to calculate the WB activity retention at the subsequent times. The relative standard uncertainty of the WB fraction of the administered activity is up to >20% by the urine collection method, but about 4% by the WBC method. The whole body dose was calculated by the OLINDA/EXM code. The relative standard uncertainty of the whole body dose is <10%, more or less same for two methods. Further reduction of the patients' burden will be achieved in the next phase, by development of methodology for a non-invasive determination of the radioactivity in the patient's blood, serving as an estimate of the bone-marrow dose.

Log: 192. **NEW SCIENTIFIC INVESTIGATIONS REQUIRE NEW RADIOANALYTICAL TECHNOLOGIES.** Pavel P. Povinec, Comenius University, Faculty of Mathematics, Physics and Informatics, Bratislava, Slovakia.

Radioanalytical technologies have always been crucial for realization of new scientific ideas, especially in application of radionuclide tracers for studying processes in the terrestrial and marine environments. Many new ideas could not be realized in the past either because of lack of suitable samples (requiring too big samples), or because of limited radioanalytical sensitivities. Recent achievements in radiometrics and mass spectrometry technologies have opened windows for investigations requiring only e.g. a few litres of seawater. In the radiometrics sector the utilization of antic cosmic shielding in shallow underground laboratories equipped with large volume HPGe detectors helped to decrease detection limits for analysis of gamma-emitters below 0.1 mBq/kg of sample. In deep underground laboratories, such as in Gran Sasso (4000 m water equivalent, w.e.) and Modane (4800 m w.e.) the detection limits close to $\sim 1 \mu\text{Bq/kg}$ have been achieved. In the mass spectrometry sector the new developments have been accompanied by development of smaller and even smaller machines for accelerator mass spectrometry (AMS), which really achieved table top dimensions. Another example is the development of a new ultra-sensitive laser based analytical technique for ^{14}C analysis, which represents an alternative technique to AMS, combining the optogalvanic effect with laser assisted isotope ratio analysis. This technique will probably make a break specifically in routine laboratory analyses of large number of samples, required e.g. in biomedical research. We shall illustrate the recent radioanalytical developments in the investigation of high density radionuclide profiles in the water column of the Indian Ocean.

Log: 193. **PLANT UPTAKE OF RADIOACTIVE CORROSION PRODUCTS AS INFLUENCED BY DIFFERENT NUTRIENT TREATMENTS.** Daniele Mascanzoni, DAPIT, University of Basilicata.

The root uptake by wheat of three activation products (Mn-54, Co-57 and Zn-65) was studied in a field experiment. The aim of the investigation was to study the factors that mainly influence the root uptake and to verify the role played by different soil treatments (Ca and K). Data on the uptake recorded during a three-year period were used to calculate the soil-plant transfer factors of these nuclides. The temporal distribution of the transfer factors and the variations exhibited by the different nuclides were examined, as well as the differences between the treatments.

Log: 194. **ADSORPTION BEHAVIORS OF TRIVALENT ACTINIDES AND LANTHANIDES ON PYRIDINE RESIN IN LITHIUM CHLORIDE AQUEOUS SOLUTION.** Suzuki, T(1); Tanaka, M(1); Ikeda, Y(1); Koyama, S(2). (1)Tokyo Institute of Technology,(2)Japan Atomic Energy Agency.

The adsorption behaviors of trivalent actinides and lanthanides on the pyridine resin in hydrochloric acid and nitric acid solutions have been studied. The mutual separation of actinides

or lanthanides was achieved by the alcoholic nitric acid solution. The adsorption depends on the charge density of ions. The group separation between the actinides and lanthanides was achieved by the hydrochloric acid solution. The group separation in hydrochloric acid solution is thought to be due to the difference of softness of their groups. However the adsorption mechanism of lanthanides on pyridine resin in hydrochloric acid solution is not clear yet. In the present work, we investigated the adsorption behaviors of trivalent actinides and lanthanides on pyridine resin in the lithium chloride aqueous solution. The separation phenomenon between actinides and lanthanides was confirmed to be similar to hydrochloric acid solution. While, the adsorption behavior of lanthanides was not similar to hydrochloric acid solution; the ions eluted from heavy elements in hydrochloric acid solution, but in the lithium chloride aqueous solution is opposite. Since hydration of pyridine resin in the lithium chloride aqueous solution is much less than that in the hydrochloric acid solution, the pyridine resin is dominantly worked as soft donor ligand in lithium chloride aqueous solutions. The tendency of adsorption in hydrochloric acid solution is rather close to the nitric acid solution. We are concluded that the adsorption of lanthanide ions in hydrochloric acid on pyridine resin is inferred to be attributed to the ion exchange.

Log: 195. **URANIUM ISOTOPIC ANALYSIS OF NATURAL ENVIRONMENTAL SAMPLES PRE URANIUM MINING: APPLICATIONS TO PROPOSED WESTERN AUSTRALIAN OPERATIONS.** Nunes, LJ (1); Loss, RD (1); McCulloch, M (2); Edwards, R (1); Trotter, J (2). (1) Curtin University, Perth, Western Australia, (2) University of Western Australia, Perth, Western Australia.

The expansion of uranium mining in Australia has, expectedly, been accompanied by an increase in community concerns regarding the safety of mining, transport of ore and eventual remediation of mine sites. To assure the community that suitable precautions are being taken, various environmental monitoring measures have been proposed and new techniques are being developed. These measures include the extensive assessment of the natural environment, prior to any disturbance associated with mine site construction/operations, thereby providing a detailed natural baseline against which differences can be assessed. One potentially powerful monitor is the natural variation in the low abundance terrestrial $^{234}\text{U}/^{238}\text{U}$ ratio. Analysis of uranium concentrations and isotopic compositions thus provides a unique method for distinctly characterising this baseline. As yet, no such uranium baseline studies have been conducted in Australia. Consequently, we have conducted a preliminary investigation of uranium isotopic signatures and levels present in a suite of samples from five river systems, and aerosols from two locations, in the South-West of Western Australia. The ultra-low levels of uranium present in these samples have required the development of low-contamination and sensitive analytical and instrumental techniques that have enabled the $^{234}\text{U}/^{238}\text{U}$ to be measured with precisions better than 0.2%. This initial testing demonstrates the technique for low-level environmental uranium analysis thereby providing a sound basis for further, more extensive studies, particularly into groundwater analysis.

Log: 196. **COHERENT NEUTRINO SCATTERING AND ANTINEUTRINO NUCLEAR REACTOR MONITORING.** Foxe, M (1,2); Bernstein, A (2); Coleman, J (3); Hagmann, C (2); Joshi, TH (4,2); Jovanovic, I (1); Kazkaz, K (2); Mavrokoridis, K (3); Mozin, V (2); Pereverzev, S (2); Sangiorgio, S (2). (1) The Pennsylvania State University, (2) Lawrence Livermore National Laboratory, (3) University of Liverpool, (4) University of California: Berkeley.

Having the knowledge of the amount of uranium and plutonium within spent nuclear fuel is vital in non-proliferation. To this end, even the relatively compact antineutrino detectors have shown the ability to track both the power and isotopic content of a nuclear reactor. While current antineutrino detectors operate on a basis of inverse beta decay, coherent neutrino scattering (CNS) also has a potential for use in these applications. While CNS is predicted but has never been experimentally confirmed, its cross section would be $\sim 100\times$ over that of inverse beta decay and could reduce the size of detectors required for nuclear reactor monitoring. The average CNS nuclear recoil energies are just a few hundred eV, which is the most important challenge in

detecting CNS. In addition to the low recoil energies, nuclear recoils produce less observable ionization than electronic recoils and the ionization yield (e-/keV) should be measured prior to the search for CNS. We have built a small dual-phase argon detector to measure the ionization yield in argon at energies of a few keV to support the search for CNS. We will discuss the commissioning of the small dual-phase argon detector and the measurement of the ionization yield using elastic neutron scatters off argon to produce recoil energies of ~7 keV. Additionally, we will discuss the experimental plans for measuring CNS using a nuclear reactor and the potential non-proliferation uses.

Log: 197. **IN-SITU GEOSPATIAL MEASUREMENTS OF CESIUM-137 ON RONGELAP ISLAND IN THE MARSHALL ISLANDS USING A MOBILE CARBORNE GAMMA-SPECTROMETRY SYSTEM.** Terry Hamilton, Steve Kehl, Mike Tamblin, and Deborah Dennison#, Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, PO Box 808, Livermore, CA 94550, USA; #Jacobs Engineering Group Inc., Lawrence Livermore National Laboratory, PO Box 808, Livermore, CA 94550, USA;.

Nuclear accidents and events such as the Fukushima Daiichi nuclear power station accident in Japan emphasize the need for rapid reporting of geospatial information on the deposition of radioactive contaminants in the environment. The Marshall Islands Program in cooperation with Radiations Solutions Inc. (RSI) has successfully deployed an advanced mobile gamma-ray spectrometry system for use in the Marshall Islands. The RSI measurement system consists of two 4 L sodium-iodide (NaI) detectors coupled to a PMT system mounted on the roof of a Rough Terrain Vehicle (RTV). The system offers fully integrated digital spectral analysis with data processing, and a real-time geospatial surveying and mapping capability. Applications of this carborne gamma-ray spectrometry (CGS) system are discussed in terms of showing the non-homogenous distribution of cesium-137 (¹³⁷Cs) in soil on Rongelap Island, and demonstrating effectiveness of the soil remediation program to help reduce external gamma exposure rates in and around the proposed village and housing area. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 198. **A PROVENANCE STUDY OF COFFEE BY PHOTON ACTIVATION ANALYSIS.** Z. J. Sun(1),(2); D. Wells(1),(2); C. Segebede(1); H. Maschner(1),(3); B. Benson(3). (1)Idaho Accelerator Center; (2)Department of Physics, Idaho State University; (3)Department of Anthropology, Idaho State University.

Photon activation analysis (PAA) is a multi-elemental radioanalytical technique in trace elements analysis with high accuracy and precision. Researchers at the Idaho Accelerator Center performed PAA analysis on coffee samples from several locations around the world as an initial step in assessing the relationship between trace elements in illicit drugs and the soils in which they were grown. The preliminary results show coffees from different locations have different concentrations of trace elements. In the three cases where we have soil samples, the matrices of elements in the coffee samples are close to the matrices of the elements of the local soil samples. The majority of trace elemental content is similar to that of the local soil sample in which the coffee is planted. It may be that coffee assimilates numerous elements from the soil where it is grown in similar ratios as is found in the soil. Thus, it is conceivable that the elemental content could serve as "fingerprint" to trace the origins of the coffee. To verify our analytical results we applied X-ray fluorescence (XRF) methods as well. Our PAA results are consistent with XRF experimental data. The future of tracing the origin of illicit drugs with the PAA technique is promising.

Log: 199. **CHARACTERIZATION OF THE NIST SHELLFISH STANDARD REFERENCE MATERIAL 4358.** S. Nour(1*), K.G.W Inn(1), J. Filliben(1), Henk van der Gaast(2), Lee Chung Men(3), Dr. M.D. Calmet(4), T. Altitzoglou(5), P. Povinec(6), Y. Takata(7), M. Wisdom(8), K. Nakamura(9), Pia Vesterbacka(10), Ching-Chung Huang(11), S.M. Vakulovsky(12). (1)National

Institute of Standards and Technology, Gaithersburg, MD 20899, USA, (2) Australian Nuclear Science and Technology Organization, Menai, Australia, (3) Institute of Nuclear Energy Research, Taiwan, (4) Institut de Radioprotection et de Sûreté Nucléaire, Orsay, France, (5) Institute for Reference Material and Measurements, Belgium, (6) IAEA Marine Environment Laboratory, Monaco, (7) Japan Chemical Analysis Center, Japan, (8) National Air and Radiation Environmental Laboratory, USA, (9) National Institute Radiological Sciences, Japan, (10) Radiation and Nuclear Safety Authority, Helsinki, Finland, (11) Taiwan Radiation Monitoring Center, Taiwan, (12) Scientific Production Association "Typhoon", Obninsk Russia.

The radioactive contamination of the ocean is a major environmental problem. There are different monitoring systems established over the world that are doing periodical measurements of the ocean ecological system. It is of a critical importance the cost effectiveness and trustworthiness of these systems and the basis for measurements comparison must be ocean-matrix radioactivity Standard Reference Materials that are representative of the ocean system. A new Standard Reference Material (SRM 4358) of Shellfish was developed at the National Institute of Standards and Technology (NIST) via an international intercomparison project that involved 12 laboratories-participants from 9 countries. The results from the participants were statistically evaluated, and the most robust certified values were the median of laboratories reported mean values; the uncertainties of the certified values were derived using bootstrap method. Certified values were established for 14 radionuclides, 5 activity ratios, and informational values for 8 more radionuclides and 2 ratios. The certified radionuclide massic activities were derived from results of at least 3 laboratories. No suspected outlier was rejected, but each laboratory was notified and asked to confirm the data and to decide that they had complete confidence in their value. In a few cases some participants revised their submission, and alternatively, few voluntarily withdrew of some reported data. The massic activities for ^{241}Am , ^{239}Pu & ^{240}Pu , ^{238}Pu , ^{238}U , ^{235}U , ^{234}U , ^{232}Th , ^{230}Th , ^{228}Th , ^{228}Ra , ^{137}Cs , ^{210}Pb , ^{90}Sr , ^{40}K and five activities ratios for $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$, $^{228}\text{Th}/^{232}\text{Th}$, $^{230}\text{Th}/^{232}\text{Th}$, and $^{238}\text{Pu}/^{239}\text{Pu}$ & ^{240}Pu were certified, and 10 more values (^{244}Cm , ^{242}Cm , ^{241}Pu , ^{226}Ra , ^{214}Pb , ^{214}Bi , ^{212}Pb , ^{208}Tl , and the ratios $^{241}\text{Pu}/^{238}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$ & ^{240}Pu) were reported as informational values for users. To accommodate material heterogeneity a 30 g sample size for analysis is recommended.

Log: 201. THE CORRECT AND INCORRECT WAY TO CALIBRATE A COMPTON SUPPRESSION COUNTING SYSTEM FOR GAMMA RAY EFFICIENCY. Landsberger, S; Biegalski, S; University of Texas.

Gamma-ray efficiency calculations for a germanium detector have been made for a Compton suppression system. Results have shown that for radionuclides that have gamma rays in coincidence the photopeaks can be severely depressed leading to erroneous results. While this can be overcome in routine neutron activation analysis using a comparator method, special consideration must be given to determine the suppression factor for coincident gamma rays when calculating the efficiency curve and for radionuclide activities. This is especially important for users of the k0 method and for fission product identification using Compton suppression methods.

Log: 202. DETERMINING TRACE AMOUNTS OF NICKEL IN PLANT SAMPLES BY NEUTRON ACTIVATION ANALYSIS. Landsberger, S; Canion, B; University of Texas.

Neutron activation analysis of plant samples for trace amounts of nickel can be utilized to determine the effectiveness of phytoextraction methods when investigating contaminated abandoned mining sites. During a routine quality assurance evaluation, the concentrations of nickel for different NIST reference materials were calculated to be significantly greater than the certified values. A perusal of the decay scheme literature revealed a substantial interference on the determination of nickel by the presence of europium. The overestimation varies to the ratio of concentrations of europium to nickel that are present in the reference materials. The goal of this work is to present a solution for this discrepancy. Typically, in some biological samples such as fish, the rare earth elements such as europium are at very low levels. However, in other biological matrices such as plants the rare-earth concentrations can be significantly elevated.

Log: 203. **DEMONSTRATION OF SELF-SHIELDING FOR THE ANALYSIS OF GOLD WITH NAA.** Landsberger, S; Dayman, K; University of Texas.

Neutron activation analysis (NAA) has been widely used as an analytical method for the determination of trace elements in varying concentrations for a variety of samples. The ability to do so is dependent on the linearity of the correlation between the concentration of the element of interest and the measurement response, specifically the count rate of characteristic gamma rays; however, elements with sufficiently high cross sections can exhibit self-shielding, resulting in measurement response that is not linearly related to the elemental concentration. NAA has also been used for the assay of various gold ores through the usual $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction, although many researchers overlook the effect of self-shielding especially when investigating low gold concentrations. Since ^{197}Au has very high thermal and epithermal cross sections of 98.8 and 1,550 barns respectively, it is the goal of this work to show the effect of self-shielding as it pertains to the analysis of gold using NAA.

Log: 204. **CHARACTERIZATION OF AN INDUSTRIAL CONTAMINATED SOIL SITE.** Landsberger, S (1); Tamalis, D (2); Meadows, T (2); Hearnberger, D;(3). (1)University of Texas;(2) Florida Memorial University;(3) Kaizeninnovations.

Uranium has been used in many industrial processes during the last century leaving a legacy of possible contamination of soil and possible aquifers. We have determined the uranium concentrations of a contaminated soil site and background samples using epithermal neutron activation analysis (ENAA) and the TRIGA research reactor. As well, we have investigated the leaching dynamics of uranium using the US EPA Toxicity Characterization Leaching Procedure (TCLP) and sequential extraction and simulated rainwater procedures. All aqueous samples were also analyzed for uranium content using ENNA. A complete description of the methodologies used including homogenization of the samples, irradiation and counting procedures and detection limits will be given.

Log: 205. **MEASURING ACTIVITY OF ^{234}U , ^{235}U , ^{238}U AND ^{232}Th DECAY CHAINS AND K-40 IN GEOLOGICAL SAMPLES USING NEUTRON ACTIVATION ANALYSIS.** Landsberger, S; Dolloff, J; Kapsimalis, R; University of Texas.

The determination of activity levels in the naturally occurring radioactive material usually entails the determination of the U-234, U-235, U-238 and Th-232 decay chain series as well as K-40. Activity levels of the decay chain series usually involves alpha spectrometry with its inherent problematic dissolution methods and very long counting times to achieve good precision. K-40 always includes using very large sample sizes for passive gamma ray detection with 12-24 hours of counting. Although it cannot be assumed that the decay series is in secular equilibrium, we have used a combination of epithermal NAA to determine U-238 and then extracted out the activity concentrations of U-234 and U-235. By using thermal NAA we have determined Th-232. By evaluating the K-41 by epithermal NAA content we then extracted out the K-40 values. All these measurements had excellent detection limits with precision of just a few percent.

Log: 206. **MONITORING OF NEUTRON FLUX CHANGES IN SHORT-LIVED NEUTRON ACTIVATION ANALYSIS.** Landsberger, S; Dayman, K; University of Texas.

It is well known variations in neutron fluxes can adversely affect the final result in neutron activation analysis. The monitoring of neutron flux changes are usually described for medium and long-lived NAA using foils of cobalt, gold, zirconium, etc. However, for short-lived neutron activation analysis there appears to be no systematic study of the variations of the neutron flux. With our new automatic pneumatic system, where irradiation timing, decay and counting and

position are very reproducible, we have performed a series of experiments using thermal and epithermal neutrons using sulfur powder as a monitor to monitor the neutron fluxes. We used two different monitors. The $^{36}\text{S}(n,\gamma)^{37}\text{S}$ reaction is ideal to use, since it has a short half-life and due to its low cross-section and isotopic abundance does not become too radioactive to be counted immediately after irradiation. We also used the $^{27}\text{Al}(n,\gamma)^{27}\text{Al}$. Our experiments confirm that neutron fluctuations up to $\pm 8\%$ can be seen with a standard deviation of 2-3%. This effect can be seen regardless of the irradiation time and must be taken into consideration to achieve the best result.

Log: 207. **MONITORING OF NEUTRON FLUX CHANGES IN SHORT-LIVED NEUTRON ACTIVATION ANALYSIS.** Landsberger, S; Biegalksi; Schneider, E; University of Texas.

At the University of Texas have concluded the This is the end of the third year of the three year grant of the Nuclear Forensics Education Award Program. We have continued to leverage the current support to have a very comprehensive program in nuclear forensics by attracting funding from other national labs and federal agencies. This has allowed the University of Texas to continue on its success of two previous Department of Energy Radiochemistry Education Award Program (REAP). The current philosophy as with REAP was to seed several preliminary projects, maintain existing focus areas and expand its collaborations with national laboratories, Department of Defense and industry. Interactions with Florida Memorial University and Huston-Tillotson, a HBCU, were also continued by offering two undergraduate students internships at the Nuclear Engineering Teaching Lab in summer 2011. We have had excellent success in placing our graduate students into internships and permanent positions at the national laborarories.

Log: 208. **IDENTIFICATION OF THE RADIONUCLIDES IN SPENT NUCLEAR FUEL THAT MAY BE DETECTED BY COMPTON SUPPRESSION AND GAMMA-GAMMA COINCIDENCE METHODS.** Schreiber, S;(1) Landsberger, S;(1) Dayman, K;(1) Orton, C; (2); Schwantes J. (2;)(1) University of Texas; (2) Pacific Northwest National Laboratory.

In an attempt to further expand the tools available to monitor the processing of special nuclear material (SNM), Pacific Northwest National Laboratory (PNNL) is developing the Multi-Isotope Process (MIP) Monitor. The MIP Monitor is designed to collect the gamma ray signatures emitted from radioactive solutions in-process at recycling facilities, while simultaneously analyzing the signatures through pattern recognition and other multivariate analysis methods to detect off-normal conditions. Any off-normal conditions could indicate the possible diversion of nuclear material from the process streams. In order to accomplish the task of detecting the diversion of nuclear materials, the MIP Monitor relies on advanced gamma radiation detection and analysis techniques. To further improve the detection capabilities of the MIP monitor, future versions of the system may incorporate both Compton suppression (anti-coincidence) and gamma-gamma (coincidence) detection methods. The fifty nuclides with the highest activities in spent fuel were identified, their decay schemes were analyzed, and the best detection scheme for each was recommended.

Log: 209. **METEOROLOGICAL ANALYSIS OF THE MEASUREMENTS OF XENON AND BARIUM/LANTHANUM IN EASTERN ASIA IN MAY 2010.** Wotawa, G..

Between May 14 and May 23, 2010, multiple detections of radionuclides were recorded at monitoring stations in Asia. The stations are operated as part of the CTBTO International Monitoring System to record signals from nuclear tests. The CTBTO monitoring system is being built up since 1987. While radionuclide detections do occur from time to time, these detections are unprecedented in terms of temporal and spatial coincidence and duration. Also the concentration levels recorded were significantly above the detection threshold. An additional measurement taken by the radiation protection authorities of the Republic of Korea was published in a local Newspaper. In an effort to better understand the scenario, the measurements were looked at from the meteorological and atmospheric transport perspective. The major question

was, whether these detections could potentially come from the same source, and whether the location of this source could be on the territory of the Democratic People's Republic of Korea.

Log: 210. **COMPARISON OF BACKGROUND GAMMA-RAY SPECTRA BETWEEN LOS ALAMOS, NEW MEXICO AND AUSTIN, TEXAS.** S Horne, K Jackman, S Landsberger.

Background counts in gamma-ray spectrometry are caused by a variety of sources. Among these are naturally occurring radioactive materials (NORM) in the environment, interactions from cosmic radiation, and contamination within the laboratory. High-purity germanium (HPGe) detectors were used to acquire long background spectra in Los Alamos, NM (elevation ~7,300 feet) and Austin, TX (elevation ~500 feet). This difference in elevation has a sizeable effect on background spectra due to cosmic interactions, such as (n,n'). Los Alamos also has a fairly high NORM concentration in the soil, relative to Austin, and this gives way to various spectral interferences. When analyzing nuclear forensics samples, these background sources can have non-trivial effects on detection limits of low-level fission products. By accurately determining the influence that elevation and environment have on background spectra, interferences within various laboratory environments can be more accurately characterized.

Log: 211. **METHODOLOGICAL ADVANCES FOR MEASURING LOW LEVEL RADIUM ISOTOPES IN SEAWATER.** Henderson P.B(1), Morris P.J(1), Moore W.S(2), and Charette M.A(1) (1) Woods Hole Oceanographic Institution (2) University of South Carolina.

A new approach for quantifying radium isotopes in seawater was developed in advance of the international GEOTRACES program, which has the goal of identifying processes and quantifying fluxes that control the distribution of trace elements and isotopes (TEIs) in the ocean. High-resolution water column samples were collected via a commercially available in-situ pump modified to accept multiple filter media including a MnO₂ impregnated acrylic cartridge for extracting radioisotopes from seawater. The modifications mitigated prefilter clogging and allowed for up to 1800 L filtrations in four hours of pumping. Different MnO₂ cartridge preparation methods were investigated to achieve maximum Ra extraction efficiency under high sample flow rates. Full ocean depth profiles were measured for short-lived radium isotopes ²²³Ra and ²²⁴Ra in shipboard laboratories using a delayed coincidence alpha scintillation counter (RaDeCC). Samples were reanalyzed four weeks and two months after collection for ²²⁸Th and ²²⁷Ac to correct for supported ²²⁴Ra and ²²³Ra, respectively. Finally, the cartridges were measured on a gamma-ray spectrometer for the long lived radium isotopes (²²⁶Ra and ²²⁸Ra). Parallel 20 L samples at each pumping depth were collected from Niskin bottles and analyzed via alpha scintillation for ²²⁶Ra to determine radium extraction efficiencies for the cartridges. These modified methods will allow for increased sample throughput and, hence, spatial resolution for radium isotopes in the ocean. Such resolution will greatly improve the determination of oceanic vertical and horizontal-mixing rates over small and large scales, which in turn can be used to calculate fluxes of TEIs into the ocean.

Log: 212. **A RAPID METHOD FOR DETERMINING STRONTIUM-89/90 IN URINE SAMPLES.** Dai X., Cui, Y.; Kramer-Tremblay, S., AECL Chalk River Nuclear Laboratories.

Following a radiological/nuclear (RN) emergency (for example: a release of radioactive substances during a nuclear power plant accident; local contamination incidents involving misuse of radioactive sources; illicit dispersion of radioactive material in a radiological dispersal device (RDD) by terrorists), the first responders and public may be internally exposed to radionuclides. Rapid bioassay is then needed to provide a quick dose assessment so that the physicians can make decisions about potential medical treatments. Strontium-90 and ⁸⁹Sr are among the most hazardous radioisotopes generated in nuclear operations and nuclear weapon tests, since they are readily incorporated into bone tissues - due to their chemical similarity to calcium. As a pure beta emitter with notable commercial uses, ⁹⁰Sr is easier to get, shield and carry but more difficult to detect during delivery to the target location compared to a gamma source; thus, it is more attractive to terrorists for a nuclear attack. In response to an RN emergency involving internal

contamination of $^{89/90}\text{Sr}$, an emergency bioassay method for radiostrontium with a short analysis turn-around time and a high sample throughput is required. A rapid bioassay method for $^{89/90}\text{Sr}$ in urine has recently been developed. For this method, 20 mL of urine sample was used. After acidification to 8M HNO_3 , strontium isotopes were separated from the urine matrix using stacked Eichrom TRU and Sr resin columns. The Sr activity was then measured by liquid scintillation counting for 10 minutes. A minimal detectable concentration (MDC) of $<15 \text{ Bq/L}$ for $^{89/90}\text{Sr}$ has been achieved, with an 8-hour analysis turn-around time for a set of 24 samples using a vacuum box for chromatographic column separation. Urine samples spiked with known amounts of ^{90}Sr were tested to evaluate the performance of the method. Details of the method and the results will be presented.

Log: 213. **QUANTIFICATION OF CESIUM IN SEAWATER OFF JAPAN USING EXTRACTION WITH AMP-PAN RESIN AND QUANTIFICATION VIA GAMMA SPECTROSCOPY AND INDUCTIVELY COUPLED MASS SPECTROMETRY**. S.M. Pike(1), K.O. Buesseler(1), C.Breier(1), H. Duliaova(2), K. Stastna(2&3), F. Sebesta(3). (1) Woods Hole Oceanographic Institution, (2) University of Hawaii, (3) Czech Technical University.

The March 2011 earthquake off the Japanese coast and subsequent tsunami that devastated the Fukushima Dai-ichi nuclear power plant resulted in the largest accidental release of Cesium 137 and 134 to the oceans. Seawater samples were collected in June 2011 from 30 to 600 kilometers off the coast of Japan as part of initial mapping of the spread of contamination in the ocean. Cesium was extracted from unfiltered and filtered ($<1.0 \mu\text{m}$) seawater using an exchange resin based upon an organic polymer (polyacrylonitrile, PAN) containing ammonium molybdophosphate (AMP) (F. Sebesta and V. Stefula, 1990). The AMP-PAN resin can be counted directly using gamma spectroscopy for ^{134}Cs and ^{137}Cs . Stable ^{133}Cs was added to evaluate extraction efficiency and quantified by ICPMS. Our 5 ml AMP-PAN resin column was on average 95% efficient in the removal of Cesium from 20 Liter samples at an average flow rate of 35 ml/min. Measured activities of ^{134}Cs and ^{137}Cs ranged from a few Bq/m³ to $>1000 \text{ Bq/m}^3$. The extraction column can be adapted to different sample volumes and easily used in the field.

Log: 214. **HAWAII FRESH FRUIT EXPORTS EMPLOYING IRRADIATION QUARANTINE TREATMENT - PRESENT AND FUTURE**. Michael Kohn and Lyle Wong, Pa'ina Hawaii.

Hawaii is the only state under a full federal fruit fly quarantine with four established Tephritid fruit fly species, the Mediterranean, Melon, Oriental and Solanaceous flies. Other pests of quarantine concern to U.S. mainland agriculture are present in Hawaii and the movement of fresh agricultural products hosts of these pests is prohibited under 10 C.F.R. Section 318.13 unless subject to a USDA, APHIS, PPQ approved post-harvest quarantine treatment. For the broad range of tropical fruit and fresh produce in production in Hawaii, irradiation quarantine treatment offers the greatest processing efficiency and flexibility for the volumes and mixture of agricultural products currently in production in Hawaii. Test shipments of Hawaii tropical fruit to commercial irradiation facilities on the U.S. mainland in Chicago and in Whippany, N.J. from 1995 to 2000 established the feasibility of use of irradiation as a quarantine treatment for Hawaii agricultural products. Hawaii's first commercial irradiator (a SureBeam X-ray irradiator) for treatment of tropical fruit for export to U.S. mainland markets, located on the Island of Hawaii, was approved for operation by USDA, APHIS, PPQ in August 2000. The rapid growth of Hawaii's tropical fruit industry is directly attributable to the opening of export markets through the use of irradiation quarantine treatment. A second commercial irradiator (Paina Hawaii, GrayStar Cobalt-60 Genesis II) is planned for Hawaii to be located on the Island of Oahu. The need, planning, and use of this facility by growers to access export markets without restriction are discussed.

Log: 215. **TRACE ANALYSIS AND LEACHING DYNAMICS OF VOLCANIC ASH USING NAA AND ICP-MS**. B. Canion (1), S. Landsberger (1), (1) University of Texas at Austin, Nuclear Engineering Teaching Lab, R-9000, Austin, Texas, USA 78712.

In 2010, Mt Merapi in Indonesia erupted releasing approximately 160 million tonnes of ash, roughly 75% of which fell on or near Indonesia. With this extreme amount of a substance, even tract elements can become relevant. We have identified a variety of trace and major elements in this ash using instrumental neutron activation analysis. We have used a variety of techniques to improve our measurements, such as the use of epithermal neutrons and a Compton suppression system. This has led to us getting very good results for a wide variety of elements, including the rare earth elements, which have been in increasing demand in modern technology. We have also used US Environmental Protection Agency Toxicity Characterization Leaching Procedure (TCLP) in order to better understand the leaching dynamics at which various elements can enter the local waters and have either enriching or toxic effects on the local ecosystem.

Log: 216. DETERMINATION OF ZINC IN GEOLOGICAL SAMPLES USING COMPTON SUPPRESSION AND EPITHERMAL INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS.

B. Canion (1), S. Landsberger (1), (1) University of Texas at Austin, Nuclear Engineering Teaching Lab, R-9000, Austin, Texas, USA 78712.

The determination of Zn in geological samples using instrumental neutron activation analysis is usually done using the $Zn-64(n,\gamma)Zn-65$ reaction and its 115 day half-life. However this analysis has proven to be often difficult. This is due to its relatively low neutron absorption cross section and gamma ray intensity, and the relatively high neutron absorption cross section and gamma intensity of Sc-46, which has an energy peak that is only 5 keV greater than Zn-65. The use of a high resolution detector makes it possible to differentiate between the Zn-65 and Sc-46 photopeaks peaks. However, the dominating Sc-46 gamma-ray can even make peak fitting routines unsuccessful in the proper determination of Zn-65. The use of a Compton suppression system suppresses the Sc-46 peak, which has two coincident gamma-rays, and this greatly improves the ratio of the height of the Zn peak to the Sc peak. Irradiating the sample with epithermal neutrons also improves the measurement since Zn has a higher cross section for epithermal neutrons rather than thermal neutrons, where as Sc-46 has a higher thermal cross section. Another technique to determine zinc is the use of $Zn-68(n,\gamma)Zn-69m$ reaction with its 13 hour half-life. This also needs to be determined using epithermal neutrons and Compton suppression INAA. However, the 438 keV gamma ray of Zn-69m has no interference with any adjoining photopeak. A critical comparison of these two methods will be given.

Log: 217. ADVANCEMENT OF ISOTOPE SEPARATION FOR THE PRODUCTION OF REFERENCE STANDARDS . Horkley, JJ (1) Carney, KP (1) McGrath, CA (1), Edwards, AJ (1), Davies, JE (1)(2), Knighton, GC (1), Sommers, JD (1), Giglio, JJ (1), (1) Idaho National Laboratory, (2) Idaho State University.

Idaho National Laboratory (INL) operates a mass separator that is currently producing high purity isotopes for use as internal standards for high precision isotope dilution mass spectrometry. In 2008, INL began the revival of the vintage 1970's era mass separator. Advancements thus far include the successful upgrading and development of system components such as the vacuum system, power supplies, ion-producing components, and beam detection equipment. Progress has been made in the production, separation, and collection of species including Ar, Kr, Xe, Sr, and Ba. Particular focuses on ion source improvements and developments have proven successful with demonstrated output beam currents of up to 10 micro-amps ^{138}Ba and 350nA ^{134}Ba from a natural abundance source charge ($\sim 2.4\%$ ^{134}Ba). This is equivalent to deposition rates on the order of 60 micrograms per hour for ^{138}Ba and 2 micrograms per hour for ^{134}Ba . In order to increase production and collection of relatively high quantities (mg levels) of pure isotopes, basic beam profile techniques and beam production techniques have been applied. Several advancements have been made in ion source designs, source material introduction, and beam detection and collection. These advancements and future developments will be presented.

Log: 218. **SEPARATION OF LANTHANIDES FROM ACTINIDES BY METAL(IV) PHOSPHATE PHOSPHONATE ION EXCHANGE MATERIALS.** Burns, JD(1); Clearfield, A(1); Borkowski, M(2); Reed, DT(2). (1) Texas A&M University, (2) Los Alamos National Laboratory-Carlsbad Operations.

The waste streams in nuclear fuel recycling contain a wide variety of cationic species. One of the important steps in the nuclear fuel cycle is for recovery of usable fission products from spent rods is to separate the lanthanides from the minor actinides, especially Am and Cm. We have synthesized a group of M(IV) phosphonate phosphates of composition $M(IV)[O_3P(C_6H_4)_nPO_3](1-x/2)(HPO_4)_x$, where $n = 1, 2, 3$ and M(IV) is Zr, Sn. These compounds are not selective for ions of charge +1, +2 but are highly selective for +3 and +4 species. By keeping actinides in high oxidation states with preferably a plus one charge, effective separation of the two groups is possible. We will present data to that effect and also a path to separation of Am and Cm.

Log: 219. **TRITIUM CONTENTS VARIATION IN PRECIPITATION WITH REGIONAL DIFFERENCE.** Yoon Yeol Yoon, Kyoochul Ha, Kil Yong Lee, Soo Young Cho, Kyung Seok Ko.

Tritium is a constituent of water molecules and it is a ideal tracer in hydrology. Therefore, the tritium and stable isotopes in precipitation were analyzed monthly at Jeju island and Daejeon in Korea. The contents variation was compared with island and continent. The tritium concentration of Daejeon was ranged from 2.3 TU to 20.9 TU and Jeju was ranged from 1.4 to 5.4 TU. The tritium content was maximum value at March and minimum value at July due to dilution effect of heavy raining. Tritium contents in precipitation of Jeju island was lower than Daejeon and this results showed general tritium content value in northern hemisphere. The stable isotope analysis results showed $\delta^{18}O(‰)$ ranged from -11.70 to -1.67 and $\delta^2H(‰)$ ranged from -85.6 to 4.3. This values were well correlated with tritium results. These data can be usefully applied to hydrological studies and dating of groundwater.

Log: 220. **RADIOCHEMICAL DETERMINATION OF URANIUM AND RADIUM ISOTOPE IN NATURAL WATER USING LIQUID SCINTILLATION COUNTER.** Yoon Yeol Yoon, Soo Young Cho, Kil Yong Lee, Kyung Seok Ko.

Uranium and radium are toxic element. These element digestion through water consumption can cause damage to kidney and bones. Therefore, the determination of U and Ra isotopes are important in environmental studies and for the protection of public health. Hot spring water was used so many people so 14 hot springwater sample were selected and U-234, U-238, Ra-226, and Ra-228 were analyzed. For obtaining more simple and better detection limit of the uranium and radium isotope analysis, simple solvent extraction methods developed.

Uranium isotope was extracted with HDEHP and counted with LSC. Radium isotopes were separated using Ba coprecipitation and Ra-226 was counted with LSC and Ra-228 was analyzed with HPGe γ -detector. Among them Ra-226 was ranged <0.003-0.145 Bq/L and Ra-228 is below detection limit. And also, uranium content was ranged <0.01-49.7 ug/L and U-234/ U-238 ratio was ranged 0.69-1.17.

Log: 221. **DETERMINATION OF NICKEL-63 AND TECHNETIUM-99 IN ENVIRONMENTAL SAMPLES.** Krzysztof Kleszcz, Jerzy W. Mietelski; The Henryk Niewodniczański Institute of Nuclear Physics Polish Academy of Sciences.

Presentation shows methods and results of determination of Ni-63 and Tc-99 in environmental samples. The goal of determination of technetium is to recognize levels of concentration of this isotope in different elements of environment thus the samples were collected in a few different places. Peat samples were collected in east-northern Poland, on the area relatively higher contaminated by Chernobyl fallout than the rest of Poland. From Wisła river in Poland, near a sewage-farm a few organic sediment samples were collected, to study possible medical-origin

technetium presence. Also samples collected in Chernobyl zone were studied. These were insects (beetle) samples and soil samples. In Chernobyl samples both Ni-63 and Tc-99 were determinate. Since both studied isotopes are pure-beta emitters, chemical separation was necessary and some improvements and tests of chemical methods were performed. Also optimization of LSC measurement was done, including quenching compensation and study of counting efficiency depending on contains of water phase in cocktail. Preliminary results of technetium measurements show very low levels of concentrations and ^{63}Ni measurements are still in progress. Chemical yield for ^{63}Ni is monitored by stable Ni measurement (using AAS method) and for ^{99}Tc the tracer is ^{95}mTc (measured by HPGe spectrometry). Because of difficulties with obtaining of ^{95}mTc , use of stable Re as a tracer for Tc is considering and also some preliminary tests were done.

Log: 222. **APPLICATION OF NUCLEAR FORENSICS METHODOLOGIES IN NUCLEAR SAFEGUARDS AND NUCLEAR SECURITY**. Klaus Mayer, Maria Wallenius, Zsolt Varga. European Commission - Joint Research Centre - Institute for Transuranium Elements.

The challenges in nuclear safeguards and in nuclear security have significantly changed over the last decades. Safeguards evolved from the pure verification of declared amounts of material to a much more information driven approach based on the verification of absence of undeclared nuclear material and undeclared nuclear activities. Consequently, new sample types such as particle samples, environmental type materials, process materials or nuclear material intercepted from illicit trafficking need to be analysed. The parameters to be investigated range from isotopic composition, microstructure, chemical impurities to decay products. The development and application of more investigative (radio-) analytical methodologies is required and more thorough, interpretative and comparative evaluation of results needs to be performed for providing the information requested. The discipline which makes use of these analytical techniques is referred to as nuclear forensic science. It aims at providing investigative leads to law enforcement and at providing clues on the origin and intended use of nuclear or other radioactive material involved in illicit incidents. It benefits from the wealth of information inherent to the material. Specific applications, possibly in combination with only minute amounts of sample call for methods of high sensitivity, low detection limits, high selectivity and high accuracy. The selection of the method or combination of methods is done according to the sample and according to the information required. These new analytical challenges and the response will be illustrated using examples from recent work at ITU.

Log: 223. **IMPROVING THE SENSITIVITY OF RESONANCE IONIZATION MASS SPECTROMETRY: CORRELATION BETWEEN SPUTTERING DYNAMICS AND USEFUL YIELD**. Willingham, D (1); Savina, MR (1); Knight, KB (2); Pellin, MJ (1); Hutcheon, ID (2). (1) Argonne National Laboratory, Argonne, (2) Lawrence Livermore National Laboratory.

RIMS (resonance ionization mass spectrometry) has been shown to be a useful tool for analyzing the relative isotope abundance of uranium in nuclear materials. Because RIMS achieves elemental selectively spectroscopically, typical analysis times are significantly less than mass spectrometry techniques that rely heavily on complex chemical separations. The application of RIMS for the analysis of uranium oxides, however, is limited by a significant decrease in the useful yield of atomic species. For example, the useful yield of U atoms from U_3O_8 has been shown to be significantly less than from reduced U metal. Using an analytical model based on the saturated ionization technique, we successfully measured the absolute photoionization cross sections of U atoms and UO and UO_2 molecules sputtered from U_3O_8 . By comparing the RIMS ion signals to the photoionization cross sections, we determined that the majority of material desorbed from U_3O_8 consists of UOx molecules rather than U atoms. The data presented herein indicates that the preferential sputtering of UOx molecules over U atoms results in a lower useful yield of U atoms. We propose to increase the useful yield of U atoms in oxide systems by using cluster ion sputtering to promote U-O bond breaks. High useful yields are critically important to the application of RIMS for nuclear safeguards and forensics.

Log: 224. **SUCROSE TRANSPORTATION IN AN ARABIDOPSIS THALIANA USING AN IMAGING PLATE AND REAL TIME IMAGING SYSTEM.** Ohmae, Y (1); Hirose, A (1); Sugita, R (1); Tanoi, K (1); Nakanishi, TM (1). (1) University of Tokyo.

As an approach to increase production of rape seed, *Arabidopsis thaliana*, which is in the same brassica family as a rape seed plant, was used to investigate transport behavior and distribution of photosynthetic metabolite in plant tissue. In this study, sucrose, initial product of photosynthesis, labeled with carbon-14 was used. Sucrose labeled with carbon-14 was applied from the surface of a plant leaf. Then, the transport behavior of sucrose in plant tissue was investigated using an imaging plate and real time imaging system. *Arabidopsis thaliana* was cultured in normal MGRL water culture medium. During the growth, number of pods, height of each stem and branch as well as number of sprout of each rosette leaves were recorded. After application of labeled sucrose, transport behavior of sucrose was studied for the plants during the developmental stage of rosette, main stem and flower-bud formation. The distributed amount of labeled sucrose in the main stem and branches was measured and the relationship between each branch and pods was discussed. In the case of rosette leaves, the distributed amount of labeled sucrose from applied leaf was measured in all leaves, from fresh to old leaves, and the transportation behavior was discussed. At the same time, photosynthetic performance of leaves was measured by a portable photosynthesis and transpiration system, and the relationship between the amount of sucrose and photosynthetic performance was also considered.

Log: 225. **THE DEVELOPMENT OF C-14 DETECTABLE REAL-TIME RI IMAGING SYSTEM FOR PLANT UNDER INTERMITTENT LIGHT ENVIRONMENT.** Hirose, A(1); Yamawaki(2), M; Kanno, S(1); Igarashi, S(1); Sugita, R(1); Ohmae, Y(1); Tanoi, K(1); Nakanishi, TM(1). (1) The University of Tokyo, (2) AIST.

We present the improved real-time RI imaging system to study the kinetics of nutrient uptake manner and the transfer of photosynthetic products in a living plant. In order to study the transfer of photosynthetic products, detection of C-14 (middle-energy pure beta-emitter) under light irradiation is required. The 1st system which we developed was applicable only to dark condition experiments to protect highly sensitive CCD camera (Kanno et al. 2007), so that it was not able to keep the plant in condition suitable for photosynthesis. The 2nd system was applicable to light/dark condition (Yamawaki et al. 2009). However, it could detect only photon emitter or high energy beta-ray emitter such as P-32. In order to prevent detection of photons from LED lights to irradiate the plants, it was necessary to put aluminum foil (50 micron in thickness) in front of the scintillator. Therefore, middle to low energy beta-emitters, such as C-14, S-35, Ca-45, were not detected. Instead of using the aluminum foil, our new system was revised to turn off LED lights while the photon counting period. By this improvement, the detection of middle energy beta-emitters during intermittent light irradiation was achieved. Furthermore, a commercially available digital camera was equipped at the opposite side of photon counting camera, which could be operated by remote control. It allowed superimposing the autoradiogram and the real image during the growth of the plant.

Log: 226. **ELEMENTAL ANALYSIS AND CHARACTERIZATION OF LEAD WHITE PIGMENT IN 15-TH - 17-TH CENTURIES ICON PAINTINGS FROM SOUTH-EASTERN POLAND.** Panczyk E.(1); Panczyk M.(2). (1) INCT, (2) Polish Geological Institute - National Research Institute.

A physicochemical analysis of a painting is aimed at determining the exact nature of the various materials and the technical features of paint application stating their condition. An analysis of the pigments used in paintings is of the utmost importance in order to develop appropriate methods for their conservation as it provides a detailed characteristic of the materials and enables their origins to be determined. Individual features that typify a specific artist can also be extracted. The lead white pigment has been used in painting since the middle ages both as a priming ground and pigment. Purity of the lead white is directly connected with the lead purification methods that have undergone considerable changes throughout centuries. Marked progress in

this respect was noted in the 19th century. Analyses of samples of the lead white taken from genuine 15th-18th centuries icon paintings representing the so-called South- Eastern Polish school collected at the Orthodox Art Department at the Castle Museum in Łańcut were carried out. The samples were analyzed by neutron activation analysis and by thin cross-section petrography; SEM-WDS were also used to confirm obtained data . The clustering, principal components and discriminant function analyses using STATISTICA programme were carried out to identify the similarity degree of analysed objects. The distribution pattern of trace elements in lead white pigment is distinctive and unique for analysed icon paintings and is not a copy of the pattern occurring in pigments used in other European painting schools that may have influenced the Polish master's style.

Log: 227. **STABILITY OF POLYVINYL POLYPYRROLIDONE AGAINST GAMMA-RAY IRRADIATION IN HNO₃ MEDIA.** Nogami, M(1,†); Sugiyama, Y(1); Kawasaki, T(1); Harada, M(1); Kawata, Y(2,#); Morita, Y(2); Kikuchi, T(3); Ikeda, Y(1). (1) Tokyo Institute of Technology, (2) Japan Atomic Energy Agency, (3) Mitsubishi Materials Corporation. Present Address (†) Kinki University, (#) Mitsubishi Materials Corporation

Stability of polyvinylpyrrolidone (PVPP), a resin with adsorption selectivity to U(VI) in nitric acid media, against gamma-ray irradiation has been examined using HNO₃ solutions of various concentrations. Samples of PVPP slurry immersed in HNO₃ solutions in the range from 0.1 to 6 mol·dm⁻³ (= M) were irradiated by the 60Co source at 3.6 kGy·h⁻¹ for 0.9 MGy at room temperature under ambient atmosphere. The stability of the PVPP was evaluated based on the change in the adsorption capacity of U(VI) in 3M HNO₃. As the result, no clear decrease in the capacity was observed for any samples. Or rather, it was found that the capacity increased by approximately 50 % for the PVPP slurry irradiated in 6M HNO₃. The infrared spectroscopic study for the irradiated PVPP suggested that all samples had amide groups and carboxyl ones (peaks at around 3300 cm⁻¹ and 1730 cm⁻¹, respectively). In addition, a distinguished peak around 1810 cm⁻¹ appeared for the PVPP irradiated in 3 and 6 M HNO₃. This may be attributed to two adjacent C=O bonds as seen in pyruvic acid. These results indicate that PVPP degrades by gamma-ray irradiation in HNO₃ from the cleavage of the pyrrolidone ring by the addition of oxygen atom originating from HNO₃, followed by the formation of chain monoamides with multiple coordinative atoms by the continuous addition of oxygen, finally leading to the generation of primary-amine type anion exchange resin. It is also indicated that all generated functional groups possess adsorptivity to U(VI) in 3 M HNO₃.

Log: 228. **URANIUM ISOTOPE RATIO MEASUREMENTS USING LARGE DYNAMIC RANGE ION COUNTING DALY DETECTOR ON TIMS.** Palacz, Z.A(1) (1) Isotopx Limited (UK).

We present uranium isotope ratio measurements using an ion counting Daly detector on the Phoenix Thermal Ionization Mass Spectrometer. Close coupling the amplifier electronics with the photomultiplier tube result in increased linearity to 3.5 million cps using a single deadtime correction (no detectable change in isotope ratio with count rate up to a minimum of 3.5 million cps). Cross calibration between the Daly and Faraday collector can be made on count rates of >1 million cps, thereby minimizing errors due to counting statistics from the Faraday collector. Large isotope ratios can thus be analysed using a combination of Faraday and ion counter, with gain measurements performed for every data cycle, ensuring precision and calibration of any gain drift. Measurement of ratios at the 1e6 level on the Daly by itself will be explored.

Log: 229. **A MARRIAGE OF THE NBL URANIUM CRM SERIES WITH THE IRMM URANIUM CRM SERIES? A QUESTION OF METROLOGICAL TRACEABILITY.** Bürger, S (1); Balsley, SD (1); Baumann, S (1); Berger, J (1); Boulyga, SF (1); Cunningham, JA (1); Koepf, A (1); Poth, J (1). (1) International Atomic Energy Agency.

It is self-evident that the certified values of certified reference materials (CRMs) traceable to the International System of Units (SI) and produced by different national or even international metrological laboratories need to be consistent with each other within stated uncertainties. When using respective materials, any discrepancy would cast doubt on the metrological traceability. In this work, the consistency among the certified values of the two preeminent uranium isotope CRM series – New Brunswick Laboratory (NBL) and Institute for Reference Materials and Measurements (IRMM) - is probed using state-of-the-art modified total evaporation MC-TIMS. NBL CRMs with respect to IRMM CRMs were independently certified, and both sets are deemed to be traceable to the SI units. In this study, the certified reference materials IRMM-184 (natural uranium) and IRMM-187 (5 % enriched U-235) were measured against various NBL U series CRMs, ranging from depleted and natural uranium (CRM U005-A and 112-A) to low and high enriched uranium (U030-A, U200, and U500). The conclusion reached is that the certified values of those IRMM and NBL CRMs - with the exception of U005-A - agree within certificate-stated uncertainties. The conclusion underlines the quality of both sets of CRMs and provides confidence in the traceability. Using IRMM CRMs as calibration standards, a new set of values for NBL U series CRMs is presented with expanded uncertainties ($k = 2$, equal to about 95 %) of about 0.035 % for major isotope ratios and at an order of magnitude of 0.1 % to 0.5 % for minor ratios.

Log: 230. **GAMMA SPECTROMETER MEASUREMENTS OF MICROGRAM QUANTITIES OF PLUTONIUM.** Myers, SC(1); Tandon, L(1); Porterfield DR(1); Martinez, PT(1). (1) Los Alamos National Laboratory .

Nineteen groups of Marple series cascade impact filters were used to collect airborne particles associated with various operations in plutonium gloveboxes. The cascade impactor separated particles based upon particle size, while the total mass of plutonium deposited on the filters ranged from 10⁻⁴ to 10⁻⁸ grams. Gamma spectrometer measurements with an n-type high purity germanium detector (HPGe) were performed with the goal of accurately quantifying the ²³⁹Pu mass on each filter using counts from the uranium L x-rays in the 13-21 keV region. However, the mass was great enough in some of the measurements to also quantify using the ²³⁹Pu gamma ray at 129.3 keV. One-quarter portions of each filter were also analyzed using thermal ionization mass spectroscopy (TIMS) and comparisons between the two methods were performed. A significant challenge was to find a calibration source with very low self-absorption characteristics so that the detector response to the low energy x-rays could be properly characterized. This was accomplished by doping an actual Marple filter with a small NIST traceable quantity of ²³⁹Pu (7.3 ug). The comparison of gamma spectrometry to TIMS analyses yielded largely favorable results. Some notable sources of measurement bias beyond counting statistics were uncovered in the process. The particle distributions on the filters were not perfectly homogenous, and since the TIMS analyses were performed on just $\frac{1}{4}$ of a filter, a significant difference could result. Duplicate filter gamma measurements revealed potential biases in the reproducibility of sample geometry, and high concentrations of ²⁴¹Am appeared to cause L-shell fluorescence effects in a small number of samples.

Log: 232. **CHARACTERIZATION OF URANIUM ISOTOPIC ABUNDANCES IN DEPLETED URANIUM METAL ASSAY STANDARD 115.** K. J. MATHEW, G. L. SINGLETON, R. M. ESSEX, A. HASOZBEK, G. ORLOWICZ & M. SORIANO, NEW BRUNSWICK LABORATORY, DEPARTMENT OF ENERGY.

Certified reference material (CRM) 115 is a uranium metal assay standard that has a depleted U isotopic composition. Each CRM unit consists of a 70-gram strip of U metal. As part of a project to create ~1-gram units of CRM 115, the materials were analyzed to verify the U assay of the smaller units and to determine certified isotopic abundance values. Ten units of CRM 115 were analyzed to characterize the uranium isotopic abundances using a ThermoFinnigan TRITON thermal ionization mass spectrometer (TIMS). The $^{235}\text{U}/^{238}\text{U}$ ratios were characterized by the total evaporation (TE) and modified total evaporation (MTE) techniques. The $^{234}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$ ratios were characterized using the MTE technique. Characterized major isotopic abundances were verified using a modified MAT261 instrument with SPECTROMAT upgrades. Sufficient care was taken to ensure that natural and/or background uranium did not contaminate the metal samples at any time prior to analysis in the mass spectrometer. These steps included: (i) the use of pre-cleaned Teflon or quartz containers for sample handling, (ii) the use of ultra high purity acids and other reagents, and (iii) the use of carefully out-gassed zone-refined Re filaments. Uncertainties in isotopic abundances were calculated following the Guide to the Expression of Uncertainties in Measurements (GUM). The characterization of the uranium isotopic abundances makes CRM 115 a primary reference standard that can be used as a QC standard in both uranium assay determinations (e.g. titrimetry and isotope dilution mass spectrometry) and isotopic measurements using TIMS and ICPMS.

Log: 233. **THE DIRECT TOTAL EVAPORATION METHOD FOR TIMS ANALYSIS.** Michael Wegener (Spectromat GmbH), Kattathu J. Mathew (New Brunswick Laboratory) & Altug Hasozbek (New Brunswick Laboratory).

The total evaporation (TE) method is a well known analytical method for isotopic measurements using thermal ionization mass spectrometers (TIMS). As fractionation effects are minimized in this analytical method, it is a method of choice in many practical applications that require high accuracy and precision isotope ratio measurements. The speed of signal regulation is a critical parameter for a steady sample evaporation process. Standard TE methods use the data system to read the ion signal and then try to get a stable signal by setting the filament current accordingly. The new, hardware-driven proprietary DTE (Direct Total Evaporation) method uses an analog regulator in the filament power supply with direct feedback of the detector intensity. Only target values are set by the data system initially. The sample evaporation process is then carried out by the hardware. The data system just monitors, collects and calculates the data. Due to the nature of electronic regulation the ion signal is kept stable for the duration of the run until the sample is consumed. For routine uranium isotopic analyses of the major ratio ($^{235}\text{U}/^{238}\text{U}$) using a modified MAT261 instrument with SPECTROMAT upgrades, precision (relative standard deviation, expressed as a percent) and accuracy (relative difference, expressed as a percent) of 0.05 % are obtained for low enriched and high enriched uranyl nitrate solutions.

Log: 234. **URANIUM ISOTOPE DILUTION MASS SPECTROMETRY (U-IDMS) TECHNIQUE USING NBL URANIUM CERTIFIED REFERENCE MATERIALS.** A. HASOZBEK, K. J. MATHEW, G. ORLOWICZ, & U. NARAYANAN, NEW BRUNSWICK LABORATORY (NBL), DEPARTMENT OF ENERGY.

Certified Reference Materials (CRMs) and proven application techniques play a critical role in high accuracy and precision analytical measurements. An understanding of the systematic component of the uncertainties of all related measurement techniques is a prerequisite to estimation of the uncertainties of high precision analytical measurement results. Isotope Dilution Mass Spectrometry (IDMS) is an established high accuracy and precision analytical technique, which is commonly used in nuclear safeguards measurements and isotope geology applications. In this project, we evaluated the possibility of using some of the well characterized uranium

isotopic and assay CRMs as spikes in U-IDMS method. U-IDMS measurements were performed with 'U' tracer (spike) to precisely determine 'U' elemental assay compositions. Well characterized ultra-pure specialty 'U' solutions prepared from high purity metal and oxide isotopic and assay standards (CRMs) were tested as alternative spikes. Standard IDMS equation was used for the estimation of the uranium elemental assay using the measured isotopic abundances of spike, sample (mixed) and blank on a thermal ionization mass spectrometer (TIMS) instrument. Two instruments, MAT 261 TIMS instrument with Spectromat hardware and software upgrades and a ThermoFinnigan TRITON, were used for the isotopic measurements. Preliminary results indicate that ^{235}U and ^{238}U isotopes in NBL CRMs can be used as spikes for the determination of unknown U elemental assay in nuclear safeguards applications.

Log: 235. **ACTINIDE CYANOMETALLATES.** Gorden, A. E. V. (1); Maynard, B. A. (1); Lynn, K. S. (1); Sykora, R. E. (2). (1) Auburn University (2) University of South Alabama .

Simple salts of the tetracyanoplatinate (TCP) anions have been known to exist for over 200 years. Early interest lied in the differing colors of the compounds altered simply by changing the cation in the solid state. In 1985, Gliemann and Yersin compiled a table of 36 known solid state TCP compounds ranging from hydrogen as the lightest cation to thulium as the heaviest in $(\text{enH}_2)[\text{Pt}(\text{CN})_4] \cdot 0.5\text{H}_2\text{O}$ and $\text{Tm}_2[\text{Pt}(\text{CN})_4] \cdot 3.21\text{H}_2\text{O}$ compounds respectively. A major structural feature of these compounds are the pseudo 1-D Pt...Pt interactions between the square planar tetracyanoplatinate anions. Quasi one-dimensional chains form in the solid state as the square planar TCP anions tend to stack forming parallel columns. The Pt...Pt interactions result in the unique spectroscopic and emissions properties of these compounds. The lanthanides offer the ability to tune the Pt...Pt spacing of these pseudo 1-D chains, predictably by taking advantage of the lanthanide contraction. The addition of ligands coordinating to the cationic metal center allows for further tuning of the TCP chains. We recently reported the characterization of actinide salts of the tetracyanoplatinates, three new TCP compounds; $\text{Th}(\text{H}_2\text{O})_7[\text{Pt}(\text{CN})_4] \cdot 2.10\text{H}_2\text{O}$, $\text{Th}_2(\text{H}_2\text{O})_{10}(\text{OH})_2[\text{Pt}(\text{CN})_4] \cdot 3.5\text{H}_2\text{O}$ and $\text{K}_3[(\text{UO}_2)_2(\text{OH})(\text{Pt}(\text{CN})_4)_2] \cdot 2\text{NO}_3 \cdot 1.5\text{H}_2\text{O}$. Of interest, was the intense emission of the thorium compounds, while the uranyl compound, lacked any observed emission. Here, we describe the emission and structural characteristics of new actinide tetracyanometallates.

Log: 236. **VANADIUM DETERMINATION IN PERNA PERNA MUSSELS (LINNAEUS, 1758: MOLLUSCA, BIVALVIA) BY PASSIVE BIOMONITORING IN THE SANTOS COAST, BRAZIL.** Seo, D (1); Vasconcellos, MBA (1); Catharino, MGM (1); Moreira, EG (1); Sousa, ECPM (2); Saiki, M (1). (1) IPEN-CNEN/SP, (2) Oceanographic Institute of the University of São Paulo.

The determination of vanadium present in seawater is of great importance to evaluate marine contamination from industrial sources as well as to identify health hazards since mussels and other marine organisms are used as food. Besides, this evaluation in the Santos coast, SP, Brazil, is important, since this area is impacted by industrial and urban activities and discharges from ships and boats. In a previous study, V results obtained by mussel transplants (active biomonitoring) were presented. This study aimed the determination of the element V by passive biomonitoring by analyzing *Perna perna* mussels collected in natural environment located in Santos which includes the sites of Ponta de Itaipu and Ilha das Palmas and the reference area is situated in Cocanha Beach. Seasonally, ninety individuals of mussels were collected in each site between September/08 and July/09. After shell removal and sample preparation, the element V was determined by Instrumental Neutron Activation Analysis (INAA). The INAA procedure consisted in the irradiation of the samples and synthetic standard of V for 8 s, under a thermal neutron flux of $6.8 \times 10^{12} \text{ n/cm}^2\text{s}$ in the nuclear research reactor IEA-R1 of IPEN-CNEN/SP. The measurements of the gamma radioactivity of the samples and standards were done using a hyperpure semiconductor Ge detector coupled to DSA-1000 Digital Spectral Analyzer. For analytical quality control, the NIST Standard Reference Material SRM 1566b Oyster Tissue was analyzed and the results presented good accuracy. The V concentrations in mussels on dry mass basis varied from 0.760 ± 0.009 to $3.34 \pm 0.32 \text{ mg/kg}$. Statistical tests applied to the

results showed difference of V concentrations between the sampling sites and season of sample collection.

Log: 237. **AUTOCAL – A METHOD FOR AUTOMATIC ENERGY CALIBRATION OF B- Γ SYSTEMS BASED ON A ^{137}Cs SPECTRUM.** S. Biegalski, The University of Texas at Austin.

A method was developed and implemented that calculates energy vs. channel calibrations. This method utilizes a ^{137}Cs spectrum acquired in beta-gamma coincidence. Both the gamma singles and the beta-gamma coincident spectrum are utilized. The gamma singles spectrum is utilized to calculate the gamma detector energy vs. channel function. The Compton line in the beta-gamma coincidence is then used to calculate the gamma detector energy vs. channel function. Currently a linear regression ($y=mx+b$) is utilized to fit the data. However, higher order polynomials may easily be implemented if desired.

Log: 238. **SDAT IMPLEMENTATION FOR THE ANALYSIS OF RADIOXENON B- Γ COINCIDENCE SPECTRA.** S. Biegalski (1), A. Flory (2), D. Haas (2), J. Ely (2), (1) The University of Texas at Austin, (2) Pacific Northwest National Laboratory.

The Standard Deconvolution Analysis Tool (SDAT) was developed for analysis of radioxenon β - γ coincidence spectra measured as part of the International Monitoring System (IMS) as defined in the Comprehensive Nuclear-Test-Ban Treaty (CTBT). The SDAT software analyzes each β - γ coincidence spectrum by fitting library vectors of each radionuclide of interest: ^{131}mXe , ^{133}mXe , ^{133}Xe , and ^{135}Xe . Detector background and radon are incorporated as optional components of the sample solution. Results are reported in mBq m⁻³. A new graphical user interface has been developed to facilitate ease of use and improve the data visualization. Automated energy vs. channel calibration algorithms were developed and implemented based on ^{137}Cs β - γ coincidence spectra. Details on the user tool and testing are included.

Log: 239. **TRACERS FOR RADIOPHARMACEUTICAL PRODUCTION FACILITIES.** S. Biegalski (1), T. Bowyer (2), D. Haas (2), (1) The University of Texas at Austin, (2) Pacific Northwest National Laboratory.

When radiopharmaceutical production facilities irradiate uranium targets for the production of radionuclides such as ^{99}Mo , the emissions interfere with the global monitoring efforts conducted by the Comprehensive Nuclear Test-Ban Treaty Organization. While suppressing emissions from such facilities would be ideal, it is understood that radioxenon emissions may not be reduced to levels below detectable quantities. As a result, a study was conducted to investigate tracers that may be utilized for source identification upon measurement. Both radioactive and stable tracers are evaluated.

Log: 240. **THE ANALYSIS OF URANIUM-232: COMPARISON OF RADIOCHEMICAL TECHNIQUES AND AN IMPROVED METHOD BY ALPHA SPECTROMETRY.** Magen Coleman, 1; Evelyn Bond, 2; Lav Tandon, 1. (1) Actinide Analytical Chemistry Group, Los Alamos National Laboratory; (2) Nuclear and Radiochemistry Group, Los Alamos National Laboratory.

Uranium-232 is an isotope of interest for nuclear forensic studies because it can provide information on the irradiation history of a sample of uranium. It is formed in uranium materials through several pathways and is typically found in very small quantities (usually ng/g or smaller). The low abundance of this isotope in irradiated materials makes it very difficult to measure accurately and precisely. Many different methods have been proposed for the analysis of ^{232}U using radiochemical methods including alpha and gamma spectrometry. In this paper, literature methods will be discussed and an improved method using alpha spectrometry will be presented. Alpha spectrometry offers a direct analysis technique for measuring ^{232}U , with few interferents that can be removed via separations. Results from our improved method will be presented and

compared to results obtained from a non-destructive gamma spectrometry method that utilizes an indirect measurement. LA-UR 11-06755

Log: 241. **OPTIMIZATION OF GAMMA SPECTROSCOPY WITH DIGITAL PULSE ANALYZERS FOR NAA.** William D. James, Center for Chemical Characterization and Analysis, Texas A&M University, College Station, Texas 77843-3144.

Cambio is a software application well-suited for analysis of high-resolution nuclear spectral data in which the critical information resides in well-masked peaks or in which the data are sparse and good statistics cannot be obtained. Uncertainties in results are reported in terms of standard deviations. In the practical use of any analysis application, it is important to have a good understanding of the accuracy of the reported uncertainties. The authors report the results of an experiment that provides a quantitative measure of the accuracy of the reported uncertainties in analyses using Cambio. Ten calibration sources ranging in activity from 25 to 450 kBq were placed 20.5 meters from a bare, 100% relative-efficient high-purity germanium detector. Peaks in these sources occur from 20 keV to 2800 keV. Data were accumulated for 128 hours to create a parent spectrum from which 1024 8-hour statistically-equivalent child spectra were created by random sampling of the parent spectrum data. The parent spectrum was analyzed by fitting parameters which included an underlying continuum, a background of natural peaks adjusted by detector efficiency and material attenuation, and a foreground of the ten sources adjusted by detector efficiency only. Each of the child spectra were then analyzed starting with the parameters of the fit to the parent spectrum and then minimizing chi-square with respect to changes in these parameters. The standard deviations of the estimated activities of the sources from all 1024 spectra were then calculated and compared to the reported uncertainties of the activities of the sources.

Log: 242. **USING INNA TO DETERMINE CERAMIC PRODUCTION AND DISTRIBUTION DURING THE 13TH AND 14TH CENTURIES IN THE ESPAÑOLA BASIN, NEW MEXICO.** William D. James* and Suzanne L. Eckert**, *Center for Chemical Characterization and Analysis, Department of Chemistry, Texas A&M University, College Station, TX 77843, **Department of Anthropology, Texas A&M University, College Station, TX 77843.

One of the primary concerns of archaeological ceramic studies is to determine ceramic production provenance. In the Santa Fe River Valley of New Mexico, traditional techniques for determining production provenance of prehistoric black-on-white pottery have failed to provide satisfactory results. As such, chemical compositional analysis using INAA was performed on 368 prehistoric potsherds recovered from five 14th century sites located in the valley. The resultant dataset of 31 elements determined allows the samples to be partitioned into 3 major compositional groups using a standard suite of statistical techniques including bivariate scatter plots, k-means clustering, principal components analysis, and discriminant analysis. By relying on Mahalanobis distances, these compositional groups can be pared down to "Core Groups" by setting criteria for group membership at 95 percent confidence of belonging to one group and a less than 1 percent chance of belonging to any other group. Although this results in a smaller data set ($n = 353$), examination of only the Core Groups provides more confidence in the robustness of observed patterns. However, defining chemical compositional groups and defining ceramic reference groups that can be tied to the ancestral pueblo landscape are two different issues. A well-defined compositional group cannot always be tied to a geological region. In an attempt to do this, we examine each Core Group separately, relying on multiple lines of geological evidence. We argue that our three INNA Core Groups represent at least two production provenances. This project represents the largest INAA archaeological project performed at TAMU's Center for Chemical Characterization to date, and the results provide a far richer interpretation of prehistoric ceramic production in the Santa Fe River Valley than previously obtained.

Log: 243. **ENVIRONMENTAL CS ISOTOPE RATIOS: FROM GLOBAL FALLOUT TO MODERN EFFLUENTS.** Snyder, DC; Mann, NR; Delmore, JE. Idaho National Laboratory.

A procedure has been developed to measure $^{135}\text{Cs}/^{137}\text{Cs}$ ratios by thermal ionization mass spectrometry (TIMS) on femtogram levels of fission product cesium. Measurements have been made on a variety of contaminated environmental matrices. Results of these measurements constrain the utility of $^{135}\text{Cs}/^{137}\text{Cs}$ ratios as an indicator of source as well as age of environmental radioactive contamination. These results further demonstrate that the measured Cs isotopic compositions of materials can be influenced by the rate of environmental release and transport on the scale of minutes to days, in addition to the fissioning material, neutron energy and flux and age of the fission product. Specifically, fractionation of these isotopes can occur due to the 135-decay chain requiring several days to completely decay to Cs, while the 137-decay chain requires less than one hour. The ^{135}Cs can be deposited farther downwind from the source term than the ^{137}Cs because the Cs precursors are gaseous iodine and xenon. The Cs isotopic compositions of globally distributed fission product generated during the above ground nuclear testing era, aged fission product released during fuel processing and effluent from operating nuclear power reactors have been characterized. Isotope ratio measurements by TIMS have been facilitated by development of sample digestion and purification methods that minimize potential elemental and polyatomic mass interferences (e.g. Ba^+ and NaRbCN^+). Measurements were made using a triple sector mass spectrometer which provided sufficient abundance sensitivity to resolve the ^{135}Cs from naturally occurring ^{133}Cs .

Log: 244. **DETERMINATION OF COPPER IN NATURAL WATERS BY REVERSED-PHASE EXTRACTION CHROMATOGRAPHY AND NEUTRON ACTIVATION USING 1-(2-THIAZOLYLazo)-2-NAPHTHOL.** Goodwin WE; Rao, RR; Chatt, A. Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, Halifax, Nova Scotia, B3H 4J3, Canada.

In recent years, an increasing interest in the quantities of certain trace elements in potable and natural waters has been expressed, requiring the development of methods for their accurate measurement. These elements are typically present at parts-per-billion levels or lower. Their determination in natural waters such as seawater is further complicated by the presence of matrix interference. As a result, preconcentration methods have become necessary. A preconcentration neutron activation analysis (PNAA) method has been developed in this work for the determination of trace levels of copper in natural water samples, including seawater. Copper is preconcentrated by a reversed-phase extraction chromatography (RPEC) method. A chelating agent, namely 1-(2-thiazolylazo)-2-naphthol (TAN), is adsorbed onto an inert resin support, namely Amberlite XAD-4, and used to extract copper from aqueous samples. Copper is assayed by direct neutron activation of the resin, without the need for an elution step. Factors affecting the uptake of copper onto the ligand-impregnated resin have been investigated; these include flow rate, pH, column height, presence of other trace elements and interference by matrix elements. Precision and accuracy of the method have been evaluated by replicate analysis and copper determinations in certified reference materials. The method has been used for the analysis of copper in real drinking water samples. The RPEC method using TAN-XAD-4 has been found to be fast, simple, reliable, and cost-effective. Copper is quantitatively retained over a wide pH range, and can be assayed even in the presence of large excess of matrix elements such as Na, Ca, and Cl.

Log: 245. **DETERMINATION OF IODINE IN CANADIAN FOODS BY EPITHERMAL INSTRUMENTAL NEUTRON ACTIVATION AND ANTI-COINCIDENCE GAMMA-RAY SPECTROMETRY.** W. Zhang (1,2); A. Chatt (1). (1) Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, Halifax, NS, B3H 4J3, Canada; (2) Radiation Protection Bureau, Health Canada, 775 Brookfield Road, AL 6302D1, Ottawa, ON, K1A 1C1, Canada

Iodine is an element of much interest in nutritional research. The daily dietary safe and adequate intake range of iodine for adults is reported to be 150-200 µg. Iodine deficiency is considered to be a serious problem in many countries. Excessive iodine intake can also contribute to certain thyroid disorders in susceptible individuals. Therefore the accurate determination of iodine in diets and individual food items is of considerable importance. Low levels of iodine cannot be easily measured by instrumental neutron activation analysis (INAA) using thermal neutrons because of interferences from high activities of ²⁴Na, ⁵⁶Mn, ⁸²Br, and ³⁸Cl in the sample. In order to circumvent this problem, epithermal INAA (EINAA) has been used by a few researchers with some success. Background activities can be reduced to some extent by EINAA. However, the background in the region of the 443-keV photopeak of ¹²⁸I is often dominated by Compton scattering from the gamma-rays of the above interfering nuclides; so the detection limits are generally not improved that greatly by EINAA alone. Anti-coincidence counting as a background suppression technique can be used in conjunction with EINAA to further reduce the background and lower the detection limit even further. An EINAA method using anti-coincidence counting (EINAA-AC) has been developed in the present study for the determination of as low as 5 ppb of iodine in Canadian diets and food items.

Log: 246. **STUDIES ON SELENOPROTEIN SPECIES IN BOVINE KIDNEYS BY BIOCHEMICAL NEUTRON ACTIVATION ANALYSIS** . Jayawickreme, CK; Chatt, A. Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, Halifax, Nova Scotia, B3H 4J3, Canada .

Neutron activation analysis (NAA) methods in conjunction with biochemical separation procedures can be developed for the studies of metalloproteomics. The analytical technique used for the detection, identification and measurement of elements in protein fractions must meet certain requirements such as the ability to analyze a few milligram quantities of solid and liquid samples non-destructively and to determine trace amounts of several elements in a matrix largely composed of carbon, hydrogen, nitrogen, oxygen and sulfur. Moreover, the technique should provide excellent precision, accuracy, sensitivity, and detection limits among other quality assurance parameters. Rapidity of analysis and high throughput of samples are also desirable features. Instrumental NAA (INAA) meets all of the above requirements and is ideally suited for studying metalloproteins and protein-bound trace elements. Emphasis will be placed in this paper on our studies of selenoproteins in bovine kidneys. Selenoproteins are generally known to concentrate in the cytosol fraction of animal tissues. We found an unusual enrichment of about 75% of Se in the nuclei fraction of bovine kidneys. About 98% of this Se is bound to proteins. An elaborate extraction method has been developed at different pH to fractionate several selenoproteins from the nuclei fraction. A cyclic INAA (CINAA) method has been developed using the ^{77m}Se nuclide with a half-life of 17.4 s for the rapid determination of picogram levels of selenium in milligram amounts of protein fractions with high precision and accuracy. Extensive studies of the fraction at pH 9.5 show the presence of three new selenoproteins.

Log: 247. **ESTIMATION OF COMBINED UNCERTAINTIES ASSOCIATED WITH THE DETERMINATION OF IODINE IN FOODS BY THE RELATIVE AND K0 INAA STANDARDIZATION METHODS.** Nyarko, BJB (1,2); Akaho, EHK (2); Fletcher, JJ (3); Chatt, A (1). (1) Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, Halifax, Nova Scotia, B3H 4J3, Canada; (2) Ghana Atomic Energy Commission, P.O. Box LG80, Legon-Accra, Ghana; (3) Physics Department, University of Cape Coast, Cape Coast, Ghana.

A combination of different types of instrumental neutron activation analysis (INAA) techniques was developed for the determination of iodine in various food items from Ghana. The methods involved conventional reactor flux INAA in conjunction with conventional and Compton suppression gamma-ray spectrometry using both the relative and the k0 standardization methods. The other methods were epithermal INAA (EINAA), pseudo-cyclic INAA (PC-INAA) and pseudo cyclic EINAA (PC-EINAA) with conventional and Compton suppression gamma-ray spectrometry using the relative and k0 standardization methods. The uncertainties associated with the determination of iodine using both the relative and the k0 standardization methods were evaluated following ISO GUM procedures. The trueness of the methods were checked by analyzing several reference materials (RM) and standard reference materials (SRM) provided by the U.S. National Institute of Standards and Technology (NIST). Our results were in good agreement with the certified values.

Log: 248. **A NEW NEUTRON DEPTH PROFILING SYSTEM USING MULTIPLE DETECTORS AND DIGITAL SPECTROSCOPE.** Padhraic Mulligan, Lei Cao (Nuclear Engineering Program, The Ohio State University).

Neutron depth profiling (NDP) is a non-destructive characterization technique for determining the concentration vs. depth for certain sparse isotopes in a material. NDP operates on the principle of isotopic emission of charged particles at a known energy following absorption of a neutron. A collimated beam of thermal neutrons impinges upon a sample containing certain isotopes with large cross sections (Li-6, B-10, He-3, etc.) These isotopes absorb a neutron and emit charged particles with known kinetic energies. As the charged particles travel out of the sample, the particles lose energy through interactions with electrons in the matrix. By detecting the energy of these emerging charged particles, the depth of the original isotope can be determined using stopping range tables specific to the material. An NDP instrument also provides an ideal experimental platform in understanding the fundamental physics of neutron detection. Our variation to the existing NDP technology seeks to improve on the efficiency, using multiple detectors and digital data acquisition. For a given isotope, the time required to achieve reasonable counting statistics is inversely proportional to the solid angle of the detector and the reaction rate in the sample. The reaction rate is a function of neutron beam energy and intensity. Since a beam's characteristics are fixed for most facilities, the only method for analyzing a sample with less counting time is to increase the solid angle of the setup. We will show that by increasing the number of detectors in our NDP instrument, we can achieve a reasonably good depth profile of B-10 in a silicon wafer using a low power University Research Reactor.

Log: 249. **CHARACTERISATION OF A SPECTROMETER FOR MEASUREMENTS OF ALPHA-PARTICLE AND X-RAY/GAMMA-RAY IN COINCIDENCE.** M. Eriksson, I. Levy, J-F. Comanducci, IAEA-EL, Monaco.

A novel alpha-particle gamma X-ray spectrometer setup, recording the acquisition in list mode has been tested for mixed radioactive sources. The list mode results have been compared with those obtained when the spectrometer used a hardware configuration for the coincidence acquisition. The spectrometer records the coincidence events between an alpha particle and characteristic L X-ray. In addition, a comparison of the performance of the spectrometer using either an HPGe detector or a Si(Li) detector has been made. The results show the list mode advantage compared to the coincidence hardware configuration with the flexibility to perform time and energy coincidences in both the detectors of the spectrometer. The paper will describe the set-up in detail and also show the results from some applications where the spectrometer has been

used, e.g. background reduction in characteristic X-ray analysis and isotopic ratio determination of alpha particle emitting radionuclides.

Log: 250. **ENHANCING CAPABILITIES OF THE SAFEGUARDS ANALYTICAL SERVICES: THE NEED OF HUMAN RESOURCES TO MEET FUTURE ANALYTICAL CHALLENGES** .

Gabriele Voigt, PhD, International Atomic Energy Agency, Steven D. Balsley, PhD, International Atomic Energy Agency, David Donohue, PhD, International Atomic Energy Agency, Christian Schmitzer, PhD, International Atomic Energy Agency

The International Atomic Energy Agency (IAEA) has established the project 'Enhancing Capabilities of the Safeguards Analytical Services' – ECAS, in order to modernize and replace the IAEA's aged nuclear analytical laboratory and infrastructure in Seibersdorf, Austria, and to expand its capability, inter alia, for sensitive environmental sample analyses. A large geometry SIMS (secondary ion mass spectrometer) has recently been installed and is now in routine operation for uranium particle analysis, with expanded analytical capabilities for Pu and other minor actinides planned for the future. The contingency for "finger printing" of uranium materials through impurity analysis, as well as the handling and analysis of samples with very low quantities of uranium, has been accommodated in the design of the new nuclear material laboratory. In parallel, the IAEA's Network of Analytical Laboratories is being expanded to help meet future analytical requirements arising from safeguards' implementation. Within the fields of chemistry, radiochemistry, mass spectrometry, materials science, etc. the IAEA and its partner Member State laboratories must seek and train new scientists, as well as retain existing expertise, in order to meet the challenges of safeguards implementation that is driven by a wide variety of information sources. Implementing new analytical capabilities will require a commensurate enhancement of human resources to match new job requirements. This means that as the Department of Safeguards transitions from traditional facility-based safeguards implementation to a more holistic State-level approach, new job profiles and skills will become essential. This paper describes the planned new features, analytical capabilities and staff required to meet the future needs of the safeguards laboratories.

Log: 251. **OPTIMIZATION OF PULSE PROCESSING PARAMETERS FOR HPGE GAMMA-RAY SPECTROSCOPY SYSTEMS USED IN EXTREME COUNT RATE CONDITIONS AND WIDE COUNT RATE RANGES.** Ronald M. Keyser and Timothy R. Twomey, ORTEC, 801 South Illinois Avenue, Oak Ridge, TN 37831.

The need to perform gamma-ray spectroscopy measurements at high count rates with HPGE detectors is more common than many believe. Examples exist in safeguards, radiochemistry, nuclear medicine, and neutron activation analysis. In other applications wide dynamic ranges in count rate may be encountered, for example samples taken after a nuclear accident are counted on a system normally used for environmental monitoring. In a real situation, it may not be possible to reduce count-rates by increasing the distance or using collimators. The challenge is to obtain the "best" data possible in the given measurement situation. "Best" is a combination of statistical (number of counts) and spectral quality (peak width and position) considerations over a wide range of count rates. The development of MultiChannel Analyzers (MCA) using Digital Signal Processing (DSP) has made possible a much wider range of values for shaping times as well as the processing of the detector signal in various ways to improve performance with pulse-by-pulse adjustments. The pulse processing time is directly related to the shaping time. The throughput is related to the pulse processing time and the duration of the detector signal. Longer shaping times generally produce better peak resolution. However, the longer shaping times mean larger dead times and lower throughput. The ability to select the best compromise between throughput and resolution is possible with DSP MCAs. In addition, the dead-time-per-pulse can be reduced by changing the digital filter without significant impact on the FWHM. To evaluate the improvements and to suggest an approach to optimization of system performance, a small and a large GEM (P-type) coaxial HPGE detector were selected for measurements to determine the performance at various input count rates and wide range of rise times and flattops in the DSpec 50 MCA. Results will be presented for the throughput measured at dead times from 30 to 99.9% with and without the use of the ORTEC Enhanced Throughput Mode.

Log: 252. **DETERMINATION OF ACTINIDES BY ICP-MS.** Y. Shi, R. Collins, C. Broome, Atomic Energy of Canada Limited, Chalk River Laboratories, Chalk River, ON Canada K0J 1J0.

Quantitative and isotopic measurement of actinide elements is required in many circumstances in the nuclear industry. For example, determination of very low levels of these alpha emitters in urine samples is used to assess the internal dose for nuclear workers. Quantifying actinide isotopes in radioactive waste from nuclear processing and nuclear facility decommissioning provides important information for waste management. Accurate determination of the uranium isotopic ratios in reactor fuels provides fuel burnup information. Inductively coupled plasma mass spectrometry (ICP-MS) has been used for the determination of Th, U, and Pu in various samples including urine, nuclear waste, and spent nuclear fuel in our laboratory. In order to maximize the capability of the technique and ensure quality analyses, ICP-MS was used to analyze samples directly, or after pre-treatment to separate complicated matrices or to concentrate the analyte(s). High-efficiency sample introduction techniques were investigated. Spectral interferences caused by peak tailing and hydrides of major actinide isotopes were studied in detail using solutions prepared in both water and heavy water. The quality of the isotopic ratio measurement was monitored using standard reference materials (SRM). ICP-MS is a proven rapid, sensitive, and accurate mass spectrometric technique routinely used to analyze actinides in various samples related to the nuclear industry.

Log: 253. **PHYTOSANITARY IRRADIATION FOR EXPORT OF FRESH PRODUCE FROM HAWAII: COMMERCIAL ADOPTION AND CURRENT ISSUES.** Peter A. Follett, USDA-ARS, 64 Nowelo St., Hilo, Hawaii, USA.

The export of many of Hawaii's fresh fruits and vegetables to the U.S. mainland is regulated due to the presence of quarantine pests. During the past 12 years, the commercial x-ray irradiation facility, Hawaii Pride LLC, has shipped papaya and other tropical fruits and vegetables to the United States mainland using irradiation treatment to control quarantine insect pests. Hawaiian

purple sweet potato is presently the highest volume product with annual exports of more than 12 million lbs. The advent of generic radiation doses for tephritid fruit flies (150 Gy) and other insects (400 Gy) will accelerate new commodity export approvals and facilitate worldwide adoption. Lowering doses for specific pests and commodities can lower treatment costs and increase capacity owing to shorter treatment times, and will minimize any quality problems. Current impediments to wider adoption include the 1 kGy limit for fresh horticultural products, the labelling requirement, and non-acceptance of phytosanitary irradiation in Japan, the European Union and elsewhere. Irradiation has potential as a treatment for unregulated imports to prevent new pest incursions. A video will be shown of sweet potato production and irradiation treatment at Hawaii Pride.

Log: 254. **VALIDATION OF AN ISOTOPE-DILUTION METHOD FOR AGE DATING ^{90}Sr .** Jennifer L. Steeb, Donald G. Graczyk, Alice M. Essling, Vivian S. Sullivan, Yifen Tsai, David B. Chamberlain.

The time since a radioactive source material was last separated from its decay daughters is designated the "age" of the material, and is useful in deducing the material's origins and history for nuclear forensics investigations. Age dating a ^{90}Sr source requires accurate determination of the ^{90}Zr daughter and the residual ^{90}Sr parent in a sample of the source material. Because the target nuclides are isobaric, the Sr and Zr from the sample must be chemically separated for mass spectrometric determination of the isotope amounts. Incomplete recovery of either element during separation introduces significant uncertainty in its determination by direct analysis. Adding known amounts of low-abundance Sr and Zr isotopes prior to processing a sample and using the technique of isotope dilution (ID) permits accurate measurement of the ^{90}Sr and ^{90}Zr isotopes irrespective of chemical recovery. This paper describes the validation of an isotope-dilution method for determining ^{90}Zr and ^{90}Sr in a ^{90}Sr source material spiked with ^{86}Sr and ^{92}Zr . The elements are separated with a miniature gas pressurized extraction chromatography (GPEC) system using a column containing Eichrom Sr ResinTM. Zirconium is rinsed through the column with 3M HNO_3 /trace HF and strontium is eluted with 1% acetic acid. Two mass spectrometric techniques, inductively coupled plasma mass spectrometry (ICP-MS) and thermal ionization mass spectrometry (TIMS), were used for measuring isotope ratios. Data were obtained with ^{90}Sr from a blood-irradiator source dissolved by Idaho National Laboratory and with assay standards of non-radioactive strontium and zirconium. Measured $^{90}\text{Zr}/^{90}\text{Sr}$ ratios and corresponding ages determined by ID-ICP-MS and ID-TIMS are presented.

Log: 255. **ORGANICS IN A HANFORD MIXED WASTE REVISITED: MYRIAD ORGANICS AND CHELATOR FRAGMENTS UNMASKED.** Toste, AP(1)1, Lechner-Fish, TJ(2), and Scheele, RD(3). 1. Missouri State University, 2. Gardere, 3. Pacific Northwest National Laboratory.

The analysis of a highly radioactive mixed waste, a volume reduction, or double-shell slurry (DSS) waste, from the U.S. DOE's Hanford Site, originally published in 1988, yielded only a 1.2% accounting of its total organic content (TOC), whereas the analysis of another mixed waste yielded 94.9%. Of particular import, evidence of organic degradation was unearthed, notably the presence of chelator fragments, presumably derived from radiolytically and/or chemically mediated degradation of complexing and chelating agents used in nuclear operations. Soon thereafter, our lab moved to academia, and focused on studying the chemodynamics of organic degradation in a simulant of a Hanford mixed waste, in a controlled laboratory setting. Much has been learned about the genesis of chelator fragments. In this report, the reanalysis of the DSS waste, which was carried out immediately, but previously unpublished, using an organic analysis scheme specifically adapted to the waste's physicochemical properties, is described. The TOC accounting climbed to 72.3%, unmasking myriad organics. A fairly complex mixture of semivolatile organics was identified, including organophosphates like tri-n-butylphosphate (TBP), a mixture of alkyl and hetero-substituted benzenes and phthalate esters, and a mixture of catboxylic acids, notably oxalate. Reanalysis of the DSS data, based on subsequent insights, including a few which are relatively recent, now reveals the presence of several nitrosated

chelator fragments. The nitrosation may, or may not, be an artifact of a derivatization procedure used in the organic analysis scheme.

Log: 256. **ADVANCES IN ^{235}U - ^{231}Pa AGE DATING OF URANIUM PURIFICATION.** Gary R. Eppich, Lawrence Livermore National Laboratory; Ross W. Williams, Lawrence Livermore National Laboratory.

The ^{235}U - ^{231}Pa chronometer is an important tool in a nuclear forensic investigation of uranium. It is used in combination with the ^{234}U - ^{230}Th chronometer for the determination of the last date of purification of uranium. Accurate measurement of ^{231}Pa , a notoriously difficult analysis, is required for the calculation of the ^{235}U - ^{231}Pa model age. For isotope dilution mass spectrometry a ^{233}Pa ($t_{1/2} = 26.967$ d) spike is added to a sample of uranium and protactinium is purified using an anion exchange column followed by two quartz-wool columns containing no ion exchange resin. The quartz-wool column takes advantage of the nature of Pa to hydrolyze as $\text{Pa}(\text{OH})_5$, which readily sorbs to SiO_2 . Pa is eluted using 2 % HNO_3 + 0.05M HF. For a single quartz-wool column, the separation factor of Pa from U is an order of magnitude greater than can be obtained using a traditional anion exchange column, allowing for a more rapid and complete separation. The ^{233}Pa spike is prepared by milking ^{237}Np , purified using similar techniques, and calibrated with the Table Mountain Latite rock standard, which is widely accepted to be in secular equilibrium. The average working life-time of the ^{233}Pa spike is three months, after which time a fresh spike must be prepared and calibrated. U and Th concentrations are measured in aliquots of the uranium by isotope dilution mass spectrometry using ^{233}U and ^{229}Th spikes, respectively. Isotope ratios are measured using a Nu Plasma MC-ICPMS. Concordant model-ages using the ^{235}U - ^{231}Pa and ^{234}U - ^{230}Th chronometers are measured for CRM U100, which is used for quality control, and for several samples of uranium under forensic investigation.

Log: 257. **APPLICATION OF MG-28 FOR CHARACTERIZATION OF INFLUX AND EFFLUX OF MG IN THE ROOTS OF RICE SEEDLINGS UNDER LOW PH AND HIGH SALINITY.**

Kobayashi, NI (1); Iwata, N (1); Saito, T (1); Suzuki, H (2); Iwata, R (3); Tanoi, K (1); Nakanishi, TM (1). (1) the University of Tokyo, (2) National Institute of Radiological Sciences, (3) Tohoku University.

Low pH and high salinity in soil affect plant growth through various factors including cation deficiency, such as calcium and magnesium etc. In this study, the effect of low pH and high salinity on the Mg uptake was investigated by determine the net influx (uptake) and efflux of Mg. Measurement of the net Mg influx and efflux amount was possible by the application of a radioactive isotope Mg-28 (half-life is 21.1 hours), which was generated by Al-27 (, 3p) Mg-28 reaction. Rice seedlings were grown in the full-nutrient solution until one long main root and four short crown roots were developed. Then, rice roots were put in the full-nutrient solution containing Mg-28 and adjusted either to pH 4.5 (low pH) or pH 5.6 (normal pH). After 15 min, the roots were washed with the cold solution for 15 min to remove the Mg-28 adsorbed on the surface of the roots. Distribution profile of Mg-28 along the root and the net Mg influx were analyzed using an imaging plate. Regardless of pH condition, Mg-28 was found to be accumulated at the root tip part (0 ~ 1 mm). Meanwhile the net influx was decreased almost by 70% under the low pH condition compared to that under normal pH. Effect of low pH on the efflux activity as well as the effect of salinity were also studied and discussed in the presentation.

Log: 258. **EFFECTS OF ORGANIC COMPOUNDS ON THE AIR-WATER PARTITIONING OF THE GROUNDWATER RADON.** Lee, KY; Yoon, YY; Cho, SY; Ko, KS. KIGAM.

Effect of organic compounds on air-water partitioning of groundwater radon (Rn-222) was studied by a modified equilibrium partitioning in closed system (EPICS) technique. The organic compounds were benzene, toluene, xylene and ethyl benzene. A radioactive standard solution (SRM4967, NIST) of radium (Ra-226) was used as a radon source of groundwater. A simple method for the determination of the partitioning of the groundwater radon was developed using a liquid scintillation counter (LSC). The radon activity of the gaseous phase in a closed container

was measured 35 days after sealing and used to calculate the effect. The effect of temperature on the radon partitioning was investigated at 0, 10, 20, 30 and 40 C. It was found that there is a linear correlation between temperature and the radon partitioning.

Log: 259. **DISTRIBUTION OF ^{137}Cs AND ^{134}Cs IN THE NORTH PACIFIC OCEAN : IMPACTS OF TEPCO FUKUSHIMA-DAIICHI NPP ACCIDENT.** Michio Aoyama, Meteorological Research Institute; Daisuke Tsumune, Central Research Institute of Electric Power Industry; Yasunori Hamajima, Kanazawa University.

Before TEPCO Fukushima-daiichi NPP accident, distribution and inventory of ^{137}Cs which originated from atmospheric weapons tests have been studied in the Pacific Ocean since late 1950s and ^{137}Cs inventory in the North Pacific Ocean was 290 \pm 30 PBq and it decreased to 69 PBq in 2011 because its radioactive decay and inter-basin transport from the North Pacific Ocean to Indian Ocean and the South Pacific Ocean. Impact of TEPCO Fukushima-daiichi NPP accident to the North Pacific Ocean occurred through two pathways, namely direct release and atmospheric deposition to wide ocean surface. We collected more than 100 seawater samples in the North Pacific Ocean in April and May 2011 by 7 VOS. Since the sample volume was 2 liter each, we measured radiocaesium activity at Ogoya underground facility to obtain reliable activity. ^{134}Cs was detected east of data line north of 40 deg. N in the Pacific Ocean in April 2011 of which activity ranged from 2 Bq m⁻³ to 12 Bq m⁻³. Total amount of direct release of ^{134}Cs was estimated to be 3.5 \pm 0.7 PBq and that by atmospheric deposition was tentatively estimated to be 10.5 \pm 2.5 PBq together with equivalent amount of ^{137}Cs , respectively. Just before Fukushima accident, about 69 PBq of ^{137}Cs was in the North Pacific Ocean, therefore, newly added ^{137}Cs is estimated to be about 20 % of total inventory of ^{137}Cs in the North Pacific Ocean.

Log: 260. **SEPARATION CHEMISTRY IN THE NUCLEAR FUEL CYCLE.** T. A. Todd, Idaho National Laboratory.

Advances in separation chemistry are providing new insights into actinide and fission product separation technology for recycle of nuclear fuel. The U.S. Fuel Cycle Research and Development (FCR&D) program, as well as international research programs, are making progress in the difficult challenge of separating trivalent actinides from lanthanides, separating americium from curium and/or separation of fission products from reprocessing raffinates or nuclear waste. Existing technology for the separation of trivalent actinides from lanthanides has been successfully demonstrated at laboratory-scale, but the scale-up of such processes to production-scale would be difficult due to the complex chemistry and large dependence on pH. Research into novel approaches including: Development and understanding of new soft donor extractants, combined hard and soft donor extractants, stereognostic approaches, utilization of the higher oxidation states of americium and inorganic ion exchange methods are currently in progress. Additionally, a new extractant to improve fission product extraction has been developed. An overview of the recent progress in minor actinide and fission product separation will be provided.

Log: 261. **ATOMIC LAYER DEPOSITON A- Al_2O_3 DIFFUSION BARRIERS TO ELIMINATE THE MEMORY EFFECT IN BETA-GAMMA RADIOXENON DETECTORS.** Warburton, WK; Hennig, W. XIA LLC.

A typical design for a beta-gamma radio-Xenon places the Xe in a fast plastic scintillator cell surrounded by a larger NaI(Tl) or CsI(Tl) crystal and looks for coincidences between beta particles captured in the plastic and gamma rays stopped in the crystal. An issue in this approach is that Xe diffuses into the plastic during the measurement so that radioactivity from one sample can contribute to the count rate in the next (the "memory" effect). In this work we report on efforts to develop a transparent Al_2O_3 barrier layer for the plastic surface that is deposited by Atomic Layer Deposition (ALD). Layers from 2 to 30 nm were deposited on wafer blanks and initially tested by exposure to Radon. We describe our experimental setup and present a model for

interpreting total Radon countrate versus time to extract the amount absorbed into the sample in 24 hours. Early results showed that at 10 nm we could reduce the memory effect to only 4% of the initial dose. Results with thicker samples will be presented and described, together with a discussion of the effects of deposition temperature. Since we find that film quality degrades with decreasing deposition temperature, while the plastic melts at the optimum temperature, we will be considering modifications of the ALD process to retain film quality at temperatures below 70 C.

Log: 263. **NEAR INFRARED REFLECTANCE SPECTROSCOPY AS A PROCESS SIGNATURE IN URANIUM OXIDES.** Plaue, JW (1, 2, 3); Klunder, GL (2); Hutcheon, ID (2); Czerwinski, KR (1); (1) Radiochemistry Program, Department of Chemistry, University of Nevada, Las Vegas, (2) Chemical Sciences Division, Lawrence Livermore National Laboratory, (3) Defense Nuclear Facilities Safety Board.

Near-infrared (NIR) reflectance spectroscopy was examined as a potential tool for the determination of forensic signatures indicative of the chemical process history of uranium oxides. The ability to determine the process history of nuclear materials is a desired, but underdeveloped, area of technical nuclear forensics. Application of the NIR technique potentially offers a quick and non-destructive tool to serve this need; however, few data have been published on the compounds of interest. The viability of NIR was investigated through the analysis of a combination of laboratory-derived and real-world uranium precipitates and oxides. A set of reference uranium materials was synthesized in the laboratory using the commonly encountered aqueous precipitation reactions for uranium ore concentration and chemical separation processes (ammonia, hydrogen peroxide, sodium hydroxide, ammonium carbonate, and magnesia). NIR spectra were taken on a range of samples heat treated in air between 85-750 °C. X-ray diffraction patterns were also obtained to complement the NIR analysis with crystal phase information. Similar analyses were performed on a set of 25 real-world samples, with process information obtained from the literature, to provide a comparison between materials synthesized in the laboratory and samples representative of industrial processes. LLNL-ABS-514352

Log: 265. **DEVELOPMENT OF A HIGH TEMPERATURE TREATMENT DEVICE FOR SPENT NUCLEAR FUEL.** Niko Kivel (1), Natalia Shcherbina (1), Ines Günther-Leopold (1), (1) Paul Scherrer Institute.

Novel reprocessing schemes and techniques are the focus of the Euratom FP7 project "Actinide Recycling for Separation and Transmutation" (ACSEPT), where the Paul Scherrer Institute (PSI) is represented in the pyrochemical domain. The subject of investigation is the selective separation of fission products (FPs) from spent nuclear fuel as a head-end step to either classical hydro based or pyro processes which are not applied on large scale by now. The selective removal of FPs which poses as major contributor the overall radiation or bear great potentials in terms of radiotoxicity i.e. Cesium or Iodine, is advantageous for further processes. At the PSI a device was developed to release volatile FPs by means of inductive heating. The heating up to 2500 K promotes the release of material that is further transported by a carrier gas stream into an inductively coupled plasma mass spectrometer for online detection. The carrier gas can be either inert (Ar) or can contain reducing or oxidizing components like Hydrogen or Oxygen, respectively. The presentation will provide insight into the development of the device by computer aided engineering approaches. The commissioning and evaluation of the device as well as a comparison to predictions by a computer model for the release will be presented.

Log: 266. **FUNDAMENTAL STUDIES OF MASS DISCRIMINATION IN MULTI-COLLECTOR INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY.** Niko Kivel (1), Bodo Hattendorf (2), Detlef Günther (2), Frank Vanhaecke (3), (1) Paul Scherrer Institute, (2) ETH Zürich, (3) Gent University.

A full understanding of the fundamental processes causing mass discrimination in multi-collector inductively coupled plasma - mass spectrometry (MC-ICP-MS) is still lacking. Despite the fact that various empiric models exist to correct isotope ratio measurement results for mass discrimination

and the origin of the main contributors is known, the quantification of the contributing effects is not possible yet. In the work presented, the impact of instrumental parameters, such as interface pressure and cone design, on the extent of mass discrimination will be demonstrated for different m/q ranges. Especially the development of the "Jet Interface" by Thermo Fisher Scientific has proven that the extraction of the ions into the mass spectrometer is a crucial contributor to mass discrimination. A further effect often associated with mass discrimination is the space-charge effect. The effect of space-charge is strongest at the lowest kinetic energies and it should be manifested in different radial distributions of low and high energy ions after the first ion lens. For this experiment, implantation of the extracted ions into a metal target positioned at the end of the first acceleration ion lens was opted for. The ions have a kinetic energy of less than 2 keV at this stage and should have been isotopically fractioned radially with respect to the flight direction by the space-charge effect. The radial distribution of the isotopes implanted in the targets is determined by laser ablation MC-ICP-MS. First results of the implantation experiments, as well as an outlook to further experiments and modeling based on the IBSimu software will be presented.

Log: 267. **DETERMINATION OF PLUTONIUM, AMERICIUM/CURIUM ISOTOPES AND RDIO-STRONTIUM IN ENVIRONMENTAL SAMPLES WITH TRU AND SR RESIN.** M. H. Lee*, J. H. Park, K. Song
Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute.

This study presents a rapid and quantitative sequential radiochemical separation method for the Pu, Sr and Am/ Cm isotopes with a TRU resin and Sr resin in environmental samples. After the radionuclides were leached from samples with 8M HNO₃, the Pu and Am/Cm isotopes were adsorbed on the TRU column and Sr isotopes were adsorbed the Sr column connected with TRU column. The Pu and Am/Cm isotopes were purified from other nuclides through TRU column, respectively. Also, ⁹⁰Sr was separated from other hindrance elements with the Sr Spec column. Finally, for removing rare earth elements, Am/Cm fractions eluted from TRU column were purified with the anion exchange resins. After α source preparation for the purified Pu and Am/Cm isotopes with the micro-coprecipitation method, the Pu and Am/Cm isotopes were measured by an alpha spectrometry. Strontium-90 was measured by a low level liquid scintillation counter. The radiochemical procedure for Pu, Am/Cm and Sr nuclides investigated in this study has been applied to environmental samples after validating it with IAEA Reference soils.

Log: 268. **A NEW ROUTE OF PRODUCTION OF POLONIUM-210.** A. Younes (1); C. Alliot(2); M. Mokili(2); G. Montavon(1); J. Champion(1). SUBATECH (1) ,ARRONAX(2).

Po-210 is obtained after the decay of At-210 (8.1 h) produced by the nuclear reaction Bi-209 (α , 2n)At-210. A Bi foil was irradiated for two hours by a 40 MeV alpha external beam delivered by the ARRONAX cyclotron in Nantes with an average beam current of 0.1 μ A . First tests were done with Po-210 activities ranging from 1 to 10 KBq. After 1 week (about 21 periods of At-210) , Po-210 and the byproducts (Bi-207 and Bi-206) were dissolved in 10 M nitric acid. The solution was then evaporated to dryness and the residue was dissolved in 7 M HCl. Po-210 was extracted in 10% TBP/p-xylene mixture and finally selectively back-extracted in the presence of 9 M HCl. The recovery yield amounted to 90%.

Log: 269. **RADIOCHEMICAL MEASUREMENTS OF NATURAL RADIONUCLIDES AS A SENSITIVE TOOL IN ENVIRONMENTAL MONITORING.** Smodiš, B; Štok, M; Černe, M; Planinšek, P; Benedik, L. Jožef Stefan Institute.

Uranium mining and milling activities are one of important sources of radioactive contamination of the environment. Uranium decay products deposited onto U-mill tailings may be transported into the nearby soil and water and consequently into vegetation where they may accumulate. Transfer of radionuclides thus represents radiological risk to humans and wildlife due to accumulation of radionuclides in target organs and ionizing radiation. Therefore, continuous surveillance of potentially exposed areas is not just a logical requirement need but also a legal obligation. As radiological monitoring represents additional cost involved in rehabilitating contaminated areas,

relatively inexpensive analytical tools such as gamma spectrometric measurements are usually applied. However, the detection limits obtained by such methods often do not fit for the purpose. Consequently, more sensitive techniques need to be considered, in particular when biological materials should be analyzed. One of appropriate options is radiochemical determination involving alpha-spectrometric or beta-counting detection, as well as measurement following neutron activation. Radiochemical procedures for the determination of U-238, Ra-226 and Pb-210 are presented and critically compared with the gamma spectrometric measurements, as exemplified by a case study of the Žirovski vrh mine and milling facility in Slovenia. The mining/milling influential area has been under continuous radiological monitoring aimed at assessing additional radiation doses due to the on-going activities since starting industrial excavations in 1982. Besides radiological assessment, the radiochemical measurements allow also for assessing mobility and bioavailability of the contaminating radionuclides.

Log: 270. **URANIUM AND RADIUM GROUNDWATER MONITORING AT INFN-GRAN SASSO NATIONAL LABORATORY, ITALY.** Wolfango Plastino (1, 2) on behalf of ERMES Collaboration. (1) Dept. Phys. - Univ. Roma Tre, (2) INFN - Roma Tre and Gran Sasso National Laboratory.

Within the framework of the Italian National Institute of Nuclear Physics (INFN) scientific program ERMES (Environmental Radioactivity Monitoring for Earth Sciences) groundwater environmental radioactivity measurements were performed inside the INFN-Gran Sasso National Laboratory (INFN-LNGS), Italy: particularly, radon, radiocarbon, tritium, uranium, and radium. For uranium as well as isotopic ratios, the measurements were performed using inductively coupled plasma mass spectrometry (ICP-MS). Some samples were filtered and analyzed by means of gamma-ray spectroscopy with high purity germanium (HPGe) detectors as ICP-MS cannot be used to analyze the radioisotopes following uranium in the natural radioactive decay chain due to their extremely small concentration. Indeed, using this technique the gamma active radionuclides in the uranium chain can be assessed, and in particular the subchain of radium: for the different chemical behaviour of uranium and radium in water, it is important to know whether these two radionuclides are in secular equilibrium. Now, ERMES contributed to several interesting results: (a) the repeated short-term enrichments of uranium in groundwater, which can be directly associated with the geodynamics of the earthquake (L'Aquila, Italy - April 6th, 2009), are a much more precise strain-meter than radon, whose presence is modulated by the radium content; (b) the radioactivity concentration in the ground water has spatial-temporal variations showing that the neutron flux background at INFN-LNGS is not steady-state, but can vary by a few percent, and for a better characterization of the neutron flux background this new piece of information should be considered.

Log: 272. **COMBINED APPROACH OF MICROSENSOR AND EF-TEM/EELS STUDIES TO STUDY THE SPECIATION OF URANIUM IN BIOFILMS AND IN THE SURROUNDING BULK SOLUTION.** Krawczyk-Bärsch, E (1); Lünsdorf, H (2); Pedersen, K (3); Arnold, T (1); Bok, F (1); Steudtner, R (1); Lehtinen, A (4); Brendler, V (1). (1) HZDR Dresden, Germany, (2) Helmholtz Centre for Infection Research Braunschweig, Germany, (3) University of Gothenburg, Sweden, (4) POSIVA OY, Eurajoki, Finland.

The effects of a microbial habitat (biofilm) on the transport behavior of uranium in a simulated incident in an underground repository were studied by pH and Eh microelectrodes and energy-filtered transmission electron microscopy (EF-TEM), and electron energy-loss spectroscopy (EELS). Biofilm samples were collected in the underground rock characterization facility tunnel ONKALO (Finland) and positioned in a rectangular flow cell for immobilization experiments by adding uranium to the bulk solution to a final uranium concentration of 4.25×10^{-5} M. The bulk solution was pumped through the flow cell in a closed circuit, simulating in situ conditions on the tunnel wall. The use of microelectrode with a tip diameter of 10 μ m to measure pH and Eh in the biofilm proved to be essential to indicate that the biofilm has formed its own microenvironment. EF-TEM and EELS studies indicated that uranium in the biofilm was immobilized intracellularly in microorganisms by the formation of metabolically mediated uranyl phosphate, similar to Autunite.

In contrast, time resolved laser fluorescence spectroscopy (TRLFS) in the bulk solution identified aqueous uranium carbonate species, likely $(\text{Ca}_2\text{UO}_2[\text{CO}_3]_3)$, formed due to the high concentration of carbonate. The results were further confirmed by thermodynamic calculations of the predominant field of uranium species, formed in the bulk solution. Our combined approach clearly showed that uranium was removed from solution and immobilized exclusively in biofilm microorganisms. In contrast, the aqueous uranyl carbonates species, formed in the bulk solution, may contribute to the migration of U(VI) and consequently to environmental hazard.

Log: 274. **RAPID DETERMINATION OF SILVER IN CULTIVATED JAPANESE AND KOREAN OYSTERS AND JAPANESE ROCK OYSTERS USING THE SHORT-LIVED NEUTRON ACTIVATION PRODUCT 110-AG.** Fukushima, M (1); Chatt, A (2). (1) Department of Basic Sciences, Faculty of Science and Engineering, Ishinomaki Senshu University, Minamisakai, Ishinomaki, Miyagi, 986-8580, Japan; (2) Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, Halifax, Nova Scotia, B3H 4J3, Canada.

Silver is one of the trace elements whose levels are generally low in most biological materials. However, rather high levels of silver can be found in marine bivalves. Oysters are known to accumulate silver in their soft tissues. Japanese people consume a fair amount of oysters. In order to assess the levels of silver as well as their distribution in various organs, cultivated oysters from Korea and Japan and Japanese rock oysters were analyzed using the 110-Ag (24.6 s) nuclide by instrumental neutron activation analysis (INAA). Cultivated oysters were collected from Korea and Matsushima in Miyagi prefecture of Japan in 2004 November. The outer shells of these oysters were first washed with tap water followed by the separation of the soft tissues, namely hepatopancreas, gill, and mantle. The samples were freeze dried and pulverized in a mill. About 0.2-0.4 g of the powdered samples were heat-sealed in precleaned polyethylene vials and irradiated for 15 s in a neutron flux of $5 \times 10^{11} \text{ cm}^{-2} \text{ s}^{-1}$ at the Dalhousie University SLOWPOKE-2 Reactor facility in Canada. After a cooling period of 20 s, gamma-ray spectra were recorded using a high-resolution Ge detector system. The 657.76-keV photopeak of 110-Ag was used for assaying silver. About 10-50 times higher levels of silver were found in Japanese cultivated oysters compared to the Korean ones. The silver levels in organs did not differ much from one another but they did vary with depth. The details of method and result will be presented.

Log: 275. **DETECTIONS BY THE CTBT RADIONUCLIDE MONITORING NETWORK AFTER THE FUKUSHIMA ACCIDENT.** Matthias Auer (1), Mika Nikkinen (1), Robert Werzi (1), Emerencia Duran (1); (1) Comprehensive Nuclear-Test-Ban Treaty Organization.

In order to monitor compliance with the Comprehensive Nuclear-Test-Ban Treaty, the International Monitoring System, a global network of seismic, hydroacoustic, infrasound and radionuclide sensors is currently being set up. The radionuclide component of this network, which will eventually comprise eighty stations, is to more than 75% completed and is designed to monitor radionuclide emissions into the atmosphere from any location on earth with high sensitivity. The station operation follows a network wide standardized operation and maintenance scheme and undergoes regular quality control checks, which makes this network a unique source of highly reliable measurements of concentrations of aerosol borne radionuclides (particulates) and of Xenon isotopes (noble gases). At the time of the accident at the Fukushima Daiichi nuclear power plant in March 2011, sixty stations for monitoring particulates and 25 noble gas stations were operational and releases were detected at all stations in the northern hemisphere but also at several stations south of the equator. In general, good agreement was found between the detections predicted by atmospheric transport models and the actual measurements at IMS stations. Based on the measured activities and isotopic composition, the nature of the release during the accident is investigated.

Log: 276. **QUANTITATIVE STUDY OF NON-STIMULATED HUMAN WHOLE SALIVA USING NAA.** H R Lewgoy(1), C. B. Zamboni(2), S. Metairon(2), I. M. M. A Medeiros(2). (1)Universidade Anhanguera Bandeirante, UNIBAN; (2)Instituto de Pesquisas Energéticas e Nucleares, IPEN - CNEN/SP.

In the last years, the use of saliva has increased as a way of diagnosing. Investigations of flow rates, pH, molecular components, hormones and proteins have been presenting a significant progress in clinical testing for diagnostics. The inorganic elements also have an important correlation that can assist, especially in periodontal disease, but these salivary components are still poorly investigated. The major advantage for using saliva in diagnosis, comparatively to serum and plasma, is the easy access and facility of collection (non-invasive). We investigated the whole non-stimulated saliva of 54 healthy subjects and 12 patients with periodontal disease using NAA technique. The samples were obtained from donors at São Paulo city (Brazil). The analyses were performed in the nuclear reactor IEA-R1 (3.5-4.5MW, pool type) at IPEN/CNEN-SP (Brazil). We found considerable metabolic changes, mainly in Ca and I.

Log: 277. **ELEMENTAL ANALYSIS OF BIOLOGICAL TISSUES OF DMDMDX/J AND C57BL/6J MICE STRAINS INVASTIGATED BY NAA.** S Metairon(1), C B Zamboni(1), M F Suzuki(1), C R Bueno Jr (2) O A Sant'Anna (3). (1)Instituto de Pesquisas Energéticas e Nucleares, IPEN - CNEN/SP, (2) Centro de Estudos do Genoma Humano, Instituto de Biociências, USP, (3) Instituto Butantan.

Biological tissues from DMDmdx/J dystrophic mice (male = 9 and female = 10) as well as C57BL/6J control group mice (male = 12) were investigated using NAA. These animals were obtained from Jackson Laboratory (Maine, USA) and later inbred at IPEN – CNEN/SP (São Paulo, Brazil). All the analyses were performed in the nuclear reactor IEA-R1 (3.5MW, pool type) at IPEN. The DMDmdx/J mouse strain is relevant as an experimental model for Duchenne Muscular Dystrophy (DMD), which is the most severe and prevalent type of muscular dystrophy. To show in more details the alterations that this disease may cause in blood, tibia, quadriceps and heart, correlations matrixes of the DMDmdx/J mouse strain were generated and compared with control group. A significant change was observed in the analysis of the heart of dystrophic mice suggesting that this dysfunction affects severely the heart muscle. In blood, the content of K is lower in dystrophic strains as well as Mg and S contents in quadriceps when compared with control group. Other than that, these results may help the researchers to evaluate the efficiency of new treatments and to compare the advantages of different treatment approaches before performing tests in patients with muscular dystrophy.

Log: 278. **A SIMPLIFIED APPROACH TOWARDS OPTIMIZATION OF TIMS EFFICIENCY FOR SAFEGUARDS PURPOSES AT ROKKASHO ON-SITE LABORATORY: MEASUREMENT OF NON-SEPARATED U-PU SAMPLES..** K. Raptis, G. Duhamel, R. Ludwig, S. Balsley, S. Bürger, V. Mayorov (IAEA), S. Hara, Y. Itoh (NMCC), J. Ninagawa(JSGO).

The Rokkasho Reprocessing Plant (RRP) in Japan is the largest plutonium facility under IAEA safeguards. Following a commissioning period, RRP is expected to go into commercial operation in the near future with a nominal annual throughput of 8000 kg Pu. Several unattended non-destructive analysis instruments are installed in the nuclear material stream at strategic key points of the facility to provide real-time information about material flows and inventory verification. In addition, a dedicated on-site laboratory (OSL) is located on the RRP premises which houses a spectrum of instrumentation needed to independently verify (1) material flow, (2) inventory declarations, (3) the operator's measurement system, and (4) the facility status. The main instrumentation used at the OSL are hybrid K-edge densitometry, alpha and gamma spectrometry, Pu(VI) spectrophotometry and thermal ionization mass spectrometry (TIMS). At OSL there are two strategic uses of TIMS: (1) to analyse the most important sample types with the highest accuracy and precision, and (2) to serve as a benchmark for equipment calibration and control the performance of other analytical systems. TIMS measurement performance is regularly verified and assessed by a diversity of quality control measures such as quality charting

performance using certified reference materials, duplicate measurements of samples and regular inter-comparison exercises within and outside the RRP. In order to comply with the timeliness and authentication aspects in OSL, rapid reporting of analytical results is required for certain sample types. OSL staff therefore continuously seeks new ways to improve measurement performance and decrease analysis reporting times. One particular processing step has been the focus of recent attention. The chemical preparation needed for isotope dilution analysis, including chemical separation of Pu from U, adds a significant amount of time to the overall analytical process for certain sample types. Exclusion of the U-Pu separation step therefore presents a potentially significant time saving. However, the consequence of analyzing mixed U-Pu samples by TIMS is not fully understood. A series of experiments is being carried out to explore the impact of non-separated U-Pu samples on TIMS measurement performance. Initial tests with plutonium product samples of very low uranium content have generated good results and indicate no adverse effect on U or Pu measurement quality by TIMS. Expansion of the experimental scope to include MOX and samples with higher U/Pu ratios is underway and will quantify the impact of larger U amounts on TIMS measurements of Pu, specifically on the major $n(\text{Pu-240})/n(\text{Pu-239})$ ratio. The influence on Pu ratios of minor isotopes will be considered as well. We will report on the latest progress made in measuring non-separated U-Pu samples by TIMS and will discuss the implications of excluding the U-Pu separation step for some sample types in OSL.

Log: 279. **ADVANCES IN NON-PROLIFERATION AND NUCLEAR FORENSICS ANALYSIS AT CEA/DIF.** F. Pointurier, A. Hubert, A.L. Fauré, A.C. Pottin, O. Marie, A. Salaün, M. Mendes and J. Aupiais (CEA, DAM, DIF, F-91297 Arpajon, France).

CEA/DIF trace analysis laboratory is an active member of the IAEA Network of Environmental Laboratories (NWAL) since 2001, both for bulk and particle analysis of environmental samples. First, various improvements carried out on existing analytical methods to obtain more precise and more accurate measurements, and/or faster results, will be presented. Then, current and planned developments of new analytical capabilities, for instance for uranium radio-chronometry, geo-location of uranium-ores, will be presented and discussed.

Log: 280. **COLLABORATION BETWEEN DOE/NNSA AND CEA IN THE AREA OF BULK ANALYSIS OF ENVIRONMENTAL SAMPLES.** F. Pointurier (CEA, DAM, DIF, F-91297 Arpajon, France), R.W. William (DOE/LLNL), S.P. LaMont (DOE/LANL, NNSA), R.E. Steiner (DOE/LLNL), D.A. Bostick (DOE/ORNL), K.B. Olsen (DOE/PNNL).

Since 2007, DOE/NNSA and CEA have collaborated on improving their respective laboratory procedures for bulk analysis of environmental samples, i.e. swipe samples collected by inspectors in nuclear facilities. The participating laboratories (CEA/DIF in Arpajon, France, Lawrence Livermore, Los Alamos, Pacific Northwest, and Oak Ridge National Laboratories) are all active members of the IAEA Network of Environmental Laboratories (NWAL) and make analyses of similar samples for the IAEA in support of nuclear safeguards. In the frame of this collaboration, all involved laboratories exchanged procedures covering both chemical purification and mass spectrometry isotopic measurements, toured each other's NWAL laboratories to observe procedures, and participated in analysis of two sets of Quality Control samples prepared by Lawrence Livermore NL with a variety of isotopic compositions containing 50-500 nanograms of uranium and 20-200 picograms of plutonium. In this presentation, laboratory procedures and practises are compared, results obtained during the two exercises are given and discussed. Recommendations for improving bulk analysis procedures are proposed.

Log: 281. **APPLICATION OF NANOSECOND-UV LASER ABLATION - ICP-MS FOR THE ISOTOPIC ANALYSIS OF SINGLE SUB-MICROMETER-SIZE URANIUM PARTICLES.** F. Pointurier, A.C. Pottin, A. Hubert (CEA, DAM, DIF, F-91297 Arpajon, France).

For the first time, laser ablation - Inductively Coupled Plasma - Mass Spectrometry (LA-ICP-MS) was used to carry out isotopic measurement on single sub-micrometer-size uranium particles. The analytical procedure was applied on several particle-containing samples already analyzed in

the same laboratory by established techniques for particle analysis: combination of the fission track technique to thermo-ionization mass spectrometry (FT-TIMS) and secondary ions mass spectrometry (SIMS). Particles were extracted from their initial matrix with ethanol and deposited on polycarbonate disk where they were fixed in a layer of an organic compound (collodion). Beforehand the isotopic analysis, particles were precisely located on the disk's surface by scanning electron microscopy (SEM) for one sample and by using the fission track technique for the other sample. Most of the particles were smaller than 1 μm and their U-235 contents were in the femtogram range. U-235/U-238 ratios were successfully analyzed for all located particles using a nanosecond-UV laser (Cetac LSX 213 nm) coupled to a quadrupole based - ICP-MS (Thermo "X-Series II"). In this presentation, LA-ICP-MS results and performance are discussed and compared with the ones obtained by FT-TIMS and SIMS.

Log: 282. **INVESTIGATION OF TRINITITE - A COMBINED RADIOMETRIC AND MICRO-ANALYTICAL APPROACH.** Wallace, C.M. (1); Koeman, E. (1); Simonetti, A. (1); Burns, P.C. (1). (1) University of Notre Dame.

Recent publications about trinitite and melt glass from other nuclear tests have demonstrated their heterogeneous nature (Fahey et al., 2010) and the inhomogeneous distribution of radionuclides within this material (Eaton and Smith, 2001). Hence, we are undertaking a detailed investigation of trinitite with spatial resolution on the order of tens to hundreds of microns, which includes a multitude of micro-analytical techniques. Gamma spectroscopy was conducted on individual trinitite samples to identify the major radionuclides present in the bulk samples. 75 μm petrographic thin sections were then cut from the samples, and the distribution of alpha activity within the thin sections was mapped using alpha track radiography on CR-39 plastic detectors. Optimized exposure time was determined for samples in contact with the detectors, and the plastic was subsequently etched with 6.25M NaOH or KOH to reveal alpha tracks. Subsequent to identification of areas with high alpha activity, laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) spot analyses were performed in order to determine trace element/radionuclide compositions while maintaining spatial resolution. Initial LA-ICP-MS results confirm the ^{239}Pu -rich nature of the Trinity device; however, the relationships between abundances of Pu and fission/activation products remain somewhat ambiguous. The findings of this preliminary study emphasize the importance of utilizing a combination of radiometric techniques and high spatial resolution mass spectrometry when characterizing post-detonation nuclear material.

Log: 283. **DETERMINATION OF ^{41}Ca WITH LSC AND AMS: METHOD DEVELOPMENT, MODIFICATIONS AND APPLICATIONS.** D. Hampe, B. Gleisberg, M. Köhler, Verein für Kernverfahrenstechnik und Analytik Rossendorf e. V.; S. Akhmedaliev, G. Rugel, S. Merchel, Helmholtz-Zentrum Dresden-Rossendorf.

The isotope ^{41}Ca is produced by neutron capture of the stable and most abundant calcium nuclide ^{40}Ca in concrete of the bioshield of nuclear reactors. Because of its long half-life (1.04*10⁵ a) the declaration of ^{41}Ca in concrete is often requested for radioactive waste disposal. The radioanalytical ^{41}Ca determination by liquid scintillation counting (LSC) is still a reasonable option for laboratories involved in decommissioning of nuclear installations despite the emission of only low-energy Auger electrons (ca. 3.6 keV) and the difficulty of obtaining a certificated standard. Besides accelerator mass spectrometry (AMS), being the most sensitive analytical technique not only for ^{41}Ca , is increasingly gaining broader accessibility and applicability. Herein, we present a radiochemical separation procedure developed for ^{41}Ca determination with LSC and AMS in varying materials. The radioanalytical isolation consists of anion exchange and extraction chromatography as well as carbonate precipitation and recrystallization from organic solvents. Thereby, disturbing radionuclides as ^{55}Fe , ^{60}Co , ^{90}Sr , ^{137}Cs , ^{152}Eu or ^{241}Pu are removed with decontamination factors of 102-104. Quench curves for determining the measurement efficiency are generated with a ^{41}Ca solution gained from the $^{41}\text{Ca}/^{40}\text{Ca}$ certified reference material ERM-AE701. In routine application the procedure is characterized by chemical yields of 25-80%, measurement efficiencies of 1-10% and detection limits of 0.05 Bq*g⁻¹ ash and 0.3 Bq*l

¹. Aliquot solutions of LSC can be easily converted into CaF_2 -AMS-targets by successive oxalate and fluoride precipitation. Pros and cons for both measurement techniques are addressed based on ^{41}Ca results from LSC and AMS for the same material.

Log: 284. **COMBINING CONFOCAL LASER SCANNING MICROSCOPY AND LASER-INDUCED FLUORESCENCE SPECTROSCOPY TO STUDY THE INTERACTIONS OF BIOFILMS WITH URANIUM.** Arnold, T(1).

Knowledge of the molecular-level speciation of uranium in microbially influenced systems is essential for predicting its environmental properties, i.e., its stability, mobility, toxicity, and potential bioavailability to humans and other organisms, and, in addition, its reactivity with surfaces and ligands.

Laser-induced fluorescence spectroscopy (LIFS) techniques have superior sensitivity for uranium(VI) and delivers direct evidence on uranium speciation. The detection limit for aqueous uranium is currently $0.2 \mu\text{g L}^{-1}$, which makes it suitable for coupling with a microscopic system. Confocal laser scanning microscopy (CLSM) is the microscopic technique that has contributed most significantly to gaining new insights into biofilms. It has been preferred to other types of microscopy, because it enables the study of live, fully hydrated biofilms. It enables three-dimensional reconstruction of biofilm architecture by optically sectioning the sample. CLSM usually concentrates on tagging cellular components (or biofilm constituents) by use of fluorescence probes and visualisation of these by use of an appropriate excitation wavelength. However, studying the interactions of metals with biological systems requires, beside its visualization, also its simultaneous spectroscopic identification. CLSM coupled to a sophisticated detection device for LIFS provides such an alternative and links visual information with chemical information. Thereby, it enables the study of metal interactions, at environmentally relevant concentrations, in situ in a noninvasive fashion in biological samples, including biofilms. Every voxel out of which CLSM images are made of carries both visual information and chemical information and so information on uranium speciation and uranium redox states can be obtained.

Log: 285. **MEASUREMENT OF RADON AND THORON PROGENY SIZE DISTRIBUTION AND DOSE ASSESSMENTS AT THE MINERAL TREATMENT INDUSTRY IN THAILAND.** S. Chanyotha, C. Kranrod, N.Chankow, R. Kritsanuwat, P. Sripoy, K. Pangza, Department of Nuclear Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok, 10330, Thailand.

A new portable type cascade impactor has been developed to determine the activity size distribution of radon and thoron progeny in a natural environment more efficiently. The modified impactor consists of 4 stages with a back up filter stage for the collection of aerosol samples. The aerosol cut points in the impactor are set for 10, 2.5, 1 and $0.5 \mu\text{m}$ at a flow rate of 4 L min^{-1} . Five CR-39 chips were used as alpha detectors for each stage. In order to separate α particles emitted from radon and thoron progeny, CR-39 detectors are covered with aluminum-vaporized Mylar films. The thickness of each film is adjusted to allow α particles emitted from radon and thoron progeny to reach the CR-39 detectors. The technique has been successfully tested in field studies, particularly inside a Mineral Treatment industry in Thailand to estimate doses in the working environment. The dose calculations by Lung Dose Evaluation Program (LUDEP) showed that activity median aerodynamic diameters (AMAD) played a significant role in determining the particle size distributions of the attached radon and thoron progeny. The dose conversion factor determined from short term measurements due to exposure from the inhalation of thoron and its progeny was found to be four times higher than comparable values for radon and its progeny. The effective dose for workers exposed to radon is about four to six times higher than thoron.

Log: 286. **DISTRIBUTION OF LANTHANIDE AND ACTINIDE ELEMENTS BETWEEN BIS-(2-ETHYLHEXYL)PHOSPHORIC ACID AND BUFFERED LACTATE SOLUTIONS CONTAINING SELECTED COMPLEXANTS**. Rudisill, TS(1); DiPrete, DP (1); Thompson, MC(1). (1) Savannah River National Laboratory.

With the renewed interest in the closure of the nuclear fuel cycle, the TALSPEAK process is being considered for the separation of Am and Cm from the lanthanide fission products in a next generation reprocessing plant. However, an efficient separation requires tight control of the pH which likely will be difficult to achieve on a large scale. To address this issue, we measured the distribution of lanthanide and actinide elements between aqueous and organic phases in the presence of complexants which were potentially less sensitive to pH control than the diethylenetriaminepentaacetic (DTPA) used in the process. To perform the extractions, a rapid and accurate method was developed for measuring distribution coefficients based on the preparation of lanthanide tracers in the Savannah River National Laboratory neutron activation analysis (NAA) facility. The complexants tested included DTPA, aceto-, benzo-, and salicylhydroxamic acids (AHA, BHA, and SHA), N,N,N',N'-tetrakis(2-pyridylmethyl)ethylenediamine (TPEN) and ammonium thiocyanate (NH₄SCN). The hydroxamic acids were the least effective of the complexants tested. The separation factors for TPEN and NH₄SCN were higher, especially for the heaviest lanthanides in the series; however, no conditions were identified which resulted in separations factors which consistently approached those measured for the use of DTPA.

Log: 287. **EVALUATION OF COMBUSTION TECHNIQUE FOR ACCURATE 3H ANALYSIS BY COMPARISON WITH ACID DISSOLUTION TECHNIQUE**. Daeji Kim(1), Phillip E Warwick(2). (1) Korea Institute of Nuclear Safety, (2) GAU-Radioanalytical, National Oceanography Centre, University of Southampton.

Determination of tritium requires specialized methods of extraction and measurement because of the low-energy pure β emission of the radionuclide. A number of limited non-destructive methods have been used for quantifying 3H in metals. However, these methods are relatively insensitive because the detection of 3H at low activity concentration is difficult. For quantitative analysis and where the 3H is present as a species other than free HTO a more aggressive approach is required to liberate the 3H. Typically, this involves the combustion of the sample and conversion of all liberated 3H species to HTO which is subsequently trapped and counted by liquid scintillation counting. Sample oxidation has been effected using a range of techniques including chemical oxidation and combustion in O₂ atmospheres at elevated pressures. The most widespread technique is ambient pressure combustion where the sample is thermally oxidised in air or O₂-enriched air within a tube furnace assembly. The combustion technique is capable of extracting any tritium fraction both weakly bound and strongly bound in solid material. However, sample oxidation rate is significant concern because it will directly affect to isolation of 3H from a specific samples. A metal is one of the difficult materials to decompose by thermal treatment because it is relatively slowly oxidized at high temperature and it will be variable depended on metal types. Alternative method decomposing metals completely for 3H analysis is dissolution in acids. The aim of this study is evaluation of combustion technique for accurate 3H analysis by comparison with acid dissolution technique.

Log: 289. **RA-226, PB-210, BI-210 AND PO-210 DEPOSITION AND REMOVAL FROM SURFACES AND LIQUIDS**. Wojcik, M; Zuzel, G. Institute of Physics Jagiellonian University.

In the frame of the BOREXINO experiment a question, which fraction of Ra-226 present in water can plate out on the nylon inner vessel surface during the water filling of the detector was investigated. Three tests with water of different pH and Ra concentrations resulted in similar absolute activities of the deposited radium. Extrapolating this result to the full-scale BOREXINO detector for its inner vessel we expect a total surface Ra-226 activity of about 90 micro-Bq. The corresponding rate of about 3 Rn dpd in the fiducial volume of the detector was still in an acceptable background range. Another problem studied in the frame of BOREXINO was removal

of Pb-210 from the liquid scintillator by its distillation and water extraction. After several tests had been performed for both methods the general outcome of this research was that for the water extraction the initial Pb content in the scintillator was reduced only accordingly to the volume ratio of the applied liquids (simple dilution). In contrast, distillation was very effective providing Pb reduction factor of more than 100. Removal and deposition efficiencies of the long-lived Rn-222 daughters during etching from and onto surfaces of standard and high purity germanium were investigated in the frame of the GERDA experiment. The standard etching procedure of Canberra-France used during production of high purity n-type germanium diodes was applied to germanium discs, which have been exposed earlier to a strong radon source for its progenies deposition. In opposition to copper and steel, they were removed from germanium very efficiently. However, the reverse process was also observed. Considerable amounts of radioactive lead, bismuth and polonium isotopes present initially in the artificially polluted etchant were transferred to the clean high purity surface.

Log: 290. **PRODUCTION RATES AND COST ESTIMATES OF ^{99}mTc FROM RHODIUM TARGETS USING ELECTRON ACCELERATORS.** LOWE, D (1); CZERWINSKI, K (1); SUDOWE, R (1); (1) - UNIVERSITY OF NEVADA LAS VEGAS.

The production of ^{99}mTc from ^{99}Mo has been widely studied and replacement techniques for ^{99}Mo are being thoroughly investigated. Through photodisintegration processes such as (γ, α) reactions, it is possible to create ^{99}mTc from a solid rhodium target that is subjected to high energy photons. Since the cross section for this type of reaction is low, it is desirable to know if the amount of ^{99}mTc that can be made from this reaction is enough to create at least one patient dose of ^{99}mTc , which is approximately ^{30}mCi . If so, how much would each patient dose cost and how does this compare to the current market price for ^{99}mTc ? This paper shows the production rates of ^{99}mTc from a rhodium target at a variety of end point energies on a Varian K15 linear electron accelerator for end point energies between 15 and 25 MeV. Furthermore, cost estimates based on accelerator cost, end point energy and shielding costs will also be presented as well as the broader implications for CTBT verification.

Log: 291. **BETA-DELAYED NEUTRON SPECTROSCOPY USING TRAPPED RADIOACTIVE IONS.** Yee, RM (1,2); Scielzo, ND (2); Bertone, PF (3); Buchinger, F (4); Caldwell, S (3,5); Clark, JA (3); Deibel, CM (3); Fallis, J (3,6); Li, G (3,4); Gulick, S (4); Lascar, D (3,7); Levand, AF (3); Norman, EB (1,2); Pedretti, M (2); Savard, G (3,5); Segel, RE (3,7); Sharma, KS (6); Sternberg, MG (3,5); Sun, T (3); Van Schelt, J (3,5); Zabransky, BJ (3). (1) UC Berkeley - Dept. of Nuclear Engineering, (2) Lawrence Livermore National Laboratory, (3) Argonne National Laboratory, (4) McGill University - Dept. of Physics, (5) University of Chicago - Dept. of Physics, (6) University of Manitoba - Dept. of Physics, (7) Northwestern University - Dept. of Physics.

The properties of beta-delayed neutron emission are of interest to both the basic and applied nuclear physics communities. For example, branching ratios are needed to determine how the short-lived neutron-rich isotopes synthesized in the astrophysical r process decay back to stability to become the isotopes we observe today. Also, neutron energy spectra and branching ratios are required for the design of nuclear reactors. Reliable measurements of the beta-delayed neutron properties can be performed with unprecedented precision using an ion trap surrounded by radiation detectors. When a radioactive ion decays in the trap, the recoil-daughter nucleus and emitted particles emerge from the $\sim 1 \text{ mm}^3$ trap volume with minimal scattering. These properties allow the momentum and energy of the emitted neutron to be precisely reconstructed from the nuclear recoil. Spectroscopy of beta-delayed neutrons can be performed with high efficiency, energy resolutions approaching $\sim 3\%$, and virtually no background. Results from a recent proof-of-principle experiment will be discussed. Prepared by LLNL under Contract DE-AC52-07NA27344 and ANL under Contract DE-AC02-06CH11357.

Log: 292. **GEANT4 MODEL VALIDATION OF COMPTON SUPPRESSED SYSTEM FOR PROCESSES MONITORING APPLICATION.** Bender, SE (1); Unlu, K (1); Orton, CR (2). (1) Pennsylvania State University, (2) Pacific Northwest National Laboratory.

Material accountancy and process monitoring in a reprocessing facility is uniquely challenging due to the complexity of the highly radioactive process stream. To overcome these difficulties, the Multi-Isotope Process (MIP) monitor project was initiated at PNNL. The goal is to develop a system that detects subtle changes in the distribution of elements in a reprocessing stream using gamma ray spectra coupled with multivariate analysis techniques autonomously in near-real time. The high concentration of ^{137}Cs in the aqueous process stream before the separation stage masks potential valuable minor gamma-ray lines because of the dominant 661.7 keV peak and subsequent Compton scattering effects. Incorporating a Compton suppression system into the MIP design may help to resolve some of the challenges. A Monte Carlo model using the simulation toolkit Geant4 is being developed to predict the expected suppressed spectrum to estimate reduction in the Compton continuum of complex samples at the Penn State University Radiation Science and Engineering Center. Despite the lack of timing information between decay events in the particle management of Geant4, encouraging results were recorded utilizing only the information within individual decays without accounting for accidental coincidences. The model has been validated with single and cascade decay emitters in two steps: as an unsuppressed system and with suppression activated. The results of the unsuppressed model for a complex source term were very promising with the peak-to-Compton ratios of the real detection system and model within 0.7%. The suppressed model shows good agreement from single photon emitting nuclides but more development is underway for complex cases like spent fuel. In this paper, preliminary results of the suppressed Geant4 model of complex source terms will be presented.

Log: 293. **EXPERIMENTAL COMPARISONS OF THE UNCERTAINTIES IN ANALYSIS OF NUCLEAR SPECTRA WITH THE CAMBIO SOFTWARE APPLICATION.** Coldwell, RL; Lasche, GP; Sandia National Laboratories.

In this paper, we present a digital gamma-gamma coincidence and anticoincidence spectrometry designed to read out waveform signals from preamplifier of gamma-ray detectors. The analogue signal digitalization is realized using DGF/Pixie-4 software and card package (XIA LLC). It has been demonstrated that the spectrometry provides an effective method of assessing the content of uranium isotopes for nuclear materials with advantages of graphically charactering ^{235}U enrichment by its unique coincidence "fingerprints". This spectrometry also provides a more sensitive and effective way for environmental trace-level cosmogenic ^{22}Na monitoring and quantification with critical detection limit of 2 mBq. An Ortec Compton suppression system has been converted in the study. Instead of using timing electronic chain to determine the coincidence event, the analog signals from primary and guard detectors were fed directly into a Pixie-4 card for pulse height and time coincident measurement. The data acquired in list-mode include coincidence and anticoincidence events consisting of records of energy and timestamp from primary and guard detectors. Every event is processed and stored in a text file for offline analysis. The advantages of list-mode data acquisition include individually logging and time stamping each electronic pulse, which allows the simultaneous creation of several different single, summed, coincident and anticoincident spectra with a single measurement. One of the consequences of list-mode data file offline processing is a spectrum with Compton suppression and without any gamma-ray peak sensitivity loss. A sophisticated computer simulation was also created with the goal of obtaining a better understanding of the experimental results.

Log: 294. **A NOVEL METHOD FOR THE MEASUREMENT OF TRACE ELEMENTS IN URANIUM RICH MATERIALS.** Zimmer, M.M.1, Kinman, W.S.1, Baldwin, S. M.1, 1.Los Alamos National Laboratory, Chemistry Division, Nuclear and Radiochemistry.

A new method for the measurement of trace elements in uranium ore concentrates (UOC) was developed. The trace element composition of a UOC, particularly the abundance of rare earth elements (REE), provides a useful tool for analysis of material source and process history. A number of UOC trace element methods have been described in the literature. The analytical approach described here, however, differs in the choice of dilution and analysis scheme, use of

interference correction, internal standard choice for drift correction, and the rinsing and calibration methodologies employed. Trace element concentrations are determined using two sets of external calibration standards that are matrix matched to sample solutions containing 100 ug/g U. The NIST standard reference material U960 (U metal) is the cleanest source of U identified for matrix matching purposes. The use of two sets of calibration standards, each containing select trace elements, allows contributions from interferences to be calculated and subtracted. Interference correction is important for the measurement of accurate REE concentrations, as the REE are particularly prone to being impacted by polyatomic oxide interferences. A two-stage rinsing routine consists of 0.16M HNO₃-0.01M HF then 0.32M HNO₃ is used between each sample. Trace element measurements are made using a Thermo Element XR high resolution ICP-MS equipped with a cooled spray chamber. The method described provides a means for accurate and precise measurement of low abundance UOC trace elements and opens the door for discovery of hitherto unrecognized source and process signatures.

Log: 295. **INTRINSIC DOSIMETRY OF GLASS CONTAINERS: A POTENTIAL INTERROGATION TOOL FOR NUCLEAR FORENSICS AND WASTE MANAGEMENT.** Clark, RA(1,2); Robertson, JD(2); Walter, ED(1); Miller, SD(1); Schwantes, JM(1). (1) Pacific Northwest National Laboratory (2) University of Missouri–Columbia.

Intrinsic dosimetry is the method of measuring total absorbed dose received by the walls of a container holding radioactive material. By considering the total absorbed dose received by a container in tandem with the physical characteristics of the radioactive material housed within that container, this method has the potential to provide law enforcement officials or waste management personnel with enhanced pathway information regarding the history of the container and its radioactive contents. We report the latest in a series of experiments designed to validate and demonstrate this newly developed tool. Thermoluminescence (TL) dosimetry was used to measure dose effects on the raw stock material of borosilicate container glass with time up to 60 days after gamma ray, x-ray, beta particle or ultraviolet irradiations at doses from 0.15 to 20 Gy. Two peaks were identified in the TL glow-curve when irradiated with Co-60, a relatively unstable peak around 115°C and a more stable peak around 225°C. Signal strength of both peaks decayed with time. The minimum detectable dose using this technique was estimated to be 0.15 Gy, which is roughly equivalent to a 24 hr irradiation at 1 cm from a 50 ng source of Co-60. After fading, this dose would still be observable approximately 1 year post-irradiation. Similar doses with beta particles and x-rays produced TL glow-curves of lower intensity. UV light created a TL signal with only one readily observable peak near 115°C. Samples from different geographical origins showed differences in TL glow-curve shape and intensity.

Log: 296. **SPECIATION OF TECHNETIUM IN BOROSILICATE GLASS.** Bradley Childs(1), Frederic Poineau(1), Ken Czerwinski(1). (1) University of Nevada Las Vegas.

Technetium-99 is an important fission ($T_{1/2} = 2.13 \times 10^5$ y) product of the nuclear industry. Technetium in its highest oxidation state (VII) is highly mobile and can represent a threat to the environment. There are over 55 million gallons of high level mixed waste located at the Hanford site. Waste tanks at the Hanford site contain Tc that could potentially leak, and in the context of management of technetium, it is envisaged to incorporate it into a very resistant glass for long term storage. A waste immobilization plan has been made in order to look at how Tc will behave when vitrified. The main concern in this process is the volatility of ⁹⁹Tc. In the work, the speciation of technetium and its resulting behavior was evaluated in glass samples. Samples were prepared from a frit that was based on formers that were used to make borosilicate glass. The behavior of technetium in glass samples was monitored based on experimental conditions (temperature, gas, glass composition). Once the glass was prepared, volatility studies were conducted in a tube furnace from 600 °C to 1100 °C. Glass sample preparation methods, characterization techniques (EXAFS, and SEM) will be presented. Preliminary results indicate that Tc(VII) is reduced to Tc(IV) in the glass at 1100 °C under a nitrogen atmosphere.

Log: 297. **RADIOCHRONOLOGICAL AGE OF A URANIUM METAL SAMPLE FROM AN ABANDONED FACILITY.** Meyers, LA(1), Williams, RW(2), Glover, SE(1), Stalcup, AM(1), Spitz, HB(1). (1) University of Cincinnati, (2) Lawrence Livermore National Laboratory.

A piece of scrap uranium metal bar buried in the dirt floor of an old, abandoned metal rolling mill was analyzed using a multi-collector inductively coupled plasma mass spectroscopy (MC-ICP-MS). The mill rolled uranium rods in the 1950s and 1960s. Samples of the dirt in which the bar was buried were also analyzed. The isotopic composition of uranium in the bar and dirt were both the same as natural uranium. The time elapsed since the uranium metal bar was last purified can be determined by the in-growth of the isotope ^{230}Th from the decay of ^{234}U , assuming that only uranium isotopes were present after purification. The age of the metal bar was determined to be 61 years at the time of this analysis and corresponds to a production date of July, 1950 \pm 1.5 years.

Log: 298. **STUDY OF THE OCTAHALODITECHNETATE $\text{Tc}_2\text{X}_8\text{N}^-$ ($\text{X} = \text{Cl}, \text{Br}$; $\text{N} = 2, 3$) ANIONS AND THEIR PHOSPHINES DERIVATIVES..** F. Poineau(1), P. Forster(1), T.Todorova(2), L. Gagliardi(3), A. Sattelberger(4), K. Czerwinski(1). (1) UNLV, (2) Ecole Polytechnique Federale de Lausanne, (3) University of Minnesota, (4) Argonne National Laboratory.

The technetium dimers $(n\text{-Bu}_4\text{N})_2\text{Tc}_2\text{X}_8$ ($\text{X} = \text{Cl}, \text{Br}$), $[\text{Cs}(2+x)][\text{H}_3\text{O}(1-x)]\text{Tc}_2\text{Br}_8$ ($x = 0.22$) and $\text{Tc}_2\text{X}_4(\text{PMe}_3)_4$ ($\text{X} = \text{Cl}, \text{Br}$), were synthesized and studied by a number of physical and computational techniques. Single crystal XRD of the acetone solvate $(n\text{-Bu}_4\text{N})_2[\text{Tc}_2\text{Br}_8]$ revealed a Tc-Tc distance (2.1625(9) Å) and average Tc-Br distances (2.4734(7) Å). The metal-metal separation in $\text{Tc}_2\text{Br}_8^{3-}$ (2.1261(9) Å) is identical to that in $\text{Tc}_2\text{Cl}_8^{3-}$ while it is significantly shortened compared to $\text{Tc}_2\text{Br}_8^{2-}$. The structure and bonding in the $\text{Tc}_2\text{X}_8\text{N}^-$ ($\text{X} = \text{Cl}, \text{Br}$; $n = 2, 3$) systems has been investigated using multiconfigurational quantum calculations. The structural parameters calculated for the four anions are within 3 % of the experimental values. Effective bond order analysis demonstrates that the four dimers exhibit similar bond multiplicity and to possess an effective triple Tc-Tc bond. The change of electronic configuration does not affect total bond order while it affects the metal-metal bond. Reactions of $(n\text{-Bu}_4\text{N})_2\text{Tc}_2\text{X}_8$ with trimethylphosphine were performed in dichloromethane; the new technetium(II) dimers, $\text{Tc}_2\text{X}_4(\text{PMe}_3)_4$ ($\text{X} = \text{Cl}, \text{Br}$), were isolated and characterized by single crystal XRD, UV-Visible spectroscopy, and cyclic voltammetry. The metal-metal distances are 2.1317(1) Å for $\text{X} = \text{Cl}$ and 2.1315(2) Å for $\text{X} = \text{Br}$. The UV-Visible spectra were recorded in benzene. Assignment of the bands as well as computing their excitation energies and intensities were performed at the CASSCF/CASPT2 level of theory. Calculations predict that the lowest energy band corresponds to the $d^* \rightarrow s^*$ transitions.

Log: 299. **HARDWARE AND SOFTWARE UPGRADES FOR MAT260/261/262 AND MAT281 INSTRUMENTS FOR NUCLEAR SAFEGUARDS MEASUREMENTS.** Michael Wegener, H.-J. Wust, Robert Schmidt (Spectromat GmbH).

Older MAT260/261/262 thermal ionization mass spectrometers (TIMS) and MAT281 UF6 gas mass spectrometers (GSMS) have aged electronic systems that may be prone to fail. Spare part supplies needed for the maintenance of these instruments are becoming scarce. Spectromat has great experience with upgrading of the TIMS and GSMS instruments for users ranging from research laboratories to safeguards analytical facilities. Partial TIMS upgrades using the RunIt26x software or complete upgrades (replacing all electronic components by modern units) for the RunIt26xPRO software package are available. Both software packages are extremely user friendly and are tailored to the needs of the nuclear safeguards community. For TIMS instruments, either choice offers the use of the unique Direct Total Evaporation (DTE) Method. For both TIMS and GSMS instruments, overall sample and data handling is improved significantly. Signal/noise ratios are reduced by optically decoupled Voltage-to-Frequency converters for the amplifiers. MAT281 UF6 mass spectrometers are upgraded completely for running the UF6/PRO software package. Instrument and user safety of the MAT281 is enhanced

by changes in the inlet system electronics. Both instrument types are workhorses with excellent performance after rejuvenation by Spectromat.

Log: 300. **STUDY OF SILICON DETECTORS FOR HIGH RESOLUTION RADIOXENON MEASUREMENTS.** Wolfgang Hennig, Christopher E. Cox, Stephen J. Asztalos, Hui Tan, Patrick J. Franz, Peter M. Grudberg, and William K. Warburton
XIA, LLC.

The radioxenon detectors currently used in applications related to nuclear explosion monitoring are typically either coincidence beta/gamma systems or high resolution gamma only systems. In the work reported here, we explored silicon-based detectors as a possible alternative to existing systems. Silicon detectors are sensitive to X-rays, gamma rays, beta radiation, and mono-energetic conversion electrons; have very high resolution for X-ray lines that the existing systems cannot clearly separate; have very low background; and as small solid state devices, can be assembled into compact systems with no or moderate cooling. Systems with multiple detectors can use X-ray/beta and X-ray/CE coincidences for higher sensitivity. On the other hand, the detection efficiency is low for higher-energy photons, and available detector sizes are small. To evaluate performance, resolutions at key energies were measured with a variety of sources and Si detectors. Interactions of characteristic electrons and photons were modeled with Monte Carlo simulations to determine detection efficiencies. The results from measurements and simulations were used to estimate the minimum detectable concentration for Xe isotopes in several possible detector geometries: in the order of 1.5 mBq/m^3 for most isotopes with a simple 1 cm^3 cube detector with two active sides and $\sim 0.1 \text{ mBq/m}^3$ for a cube with six active sides.

Log: 301. **THERMAL AND STRUCTURAL PROPERTIES OF BINARY TECHNETIUM CHLORIDES.** Erik Johnstone (1); Dr. Frederic Poineau (1); Dr. Alfred Sattelberger (1, 2); and Dr. Ken Czerwinski (1). (1) University of Nevada - Las Vegas (2) Argonne National Laboratory.

Low-valent binary technetium chlorides were synthesized in the solid state and analyzed using various characterization methods. Technetium dichloride was synthesized from the stoichiometric reaction of the elements as a novel compound with new structure-type containing a Tc-Tc triple bond. The α -phase of technetium trichloride was prepared from the reaction of $\text{Tc}_2(\text{O}_2\text{CCH}_3)_4\text{Cl}_2$ with passing HCl (g) at elevated temperatures and characterized as a novel compound isostructural to ReCl_3 with a triangular Tc^{3+} core structure. The β -phase of the trichloride was identified as a second product from the synthetic procedure of TcCl_2 , and exhibited structural characteristics comparable to the trichlorides of ruthenium and molybdenum. Technetium tetrachloride was synthesized from the metal and excess chlorine gas in sealed Pyrex tubes and used as a starting material for decomposition to TcCl_2 and TcCl_3 . For each species, thermal behavior and synthetic mechanisms were correlated. The low-valent technetium chlorides synthesized exhibit interesting chemical and physical properties, and may serve as potential synthetic inorganic and radiopharmaceutical precursors, as well as waste form-type material in the nuclear fuel cycle.

Log: 302. **NUCLEAR DEVICE DEBRIS: IMPLICATIONS FROM TRINITITE-HOSTED INCLUSIONS.** Bellucci, J.J.(1) and Simonetti, A. (1) (1) University of Notre Dame.

This study documents the three-dimensional morphology of trinitite-hosted inclusions and the first observations of alloys consisting primarily of Pb, Ta, Ga, and W. Scanning electron and backscatter electron imaging, as well as energy dispersive x-ray spectra (EDS) chemical composition data are reported for heavy metal inclusions in 14 different samples of trinitite. Grains composed of Fe-Ti-Si are the most abundant and presumably derived mainly from the explosion tower. Grains of Cu, Pb, Ta+Ga+W were also observed and are likely derivatives of the trinitite device wiring, tamper, and tamper and core, respectively. Additionally, a grain of a Ba dominant phase and multiple zircons were observed in a large majority of samples. Identifying these inclusions is the first step in preparing the samples for a more detailed in situ chemical and isotopic investigation using LA-(MC)-ICP-MS. The spherical morphology and the ubiquitous

positioning of the heavy metal inclusions on the crater walls of the glassy trinitite surfaces indicate a two-step formation. Stage one involves formation of the glassy trinitite, while the second stage involved the precipitation of the inclusions that were incorporated on the surface of the trinitite. Furthermore, the precarious positioning of these inclusions further emphasizes the need for analysis using non-destructive techniques prior to methods employing a bulk sample digestion approach.

Log: 303. **CURRENT STATUS OF A NEW GAMMA-GAMMA COINCIDENCE ANALYSIS SYSTEM.** AUTHORS: Martin E. Keillor, Craig E. Aalseth, Luke E. Erikson, Brian D. Glasgow, Eric W. Hoppe, Todd W. Hossbach, Leila K. Mizouni, Allan W. Myers, Cory T. Overman, Allen Seifert, and Timothy Stavenger ORGANIZATION: Pacific Northwest National Laboratory .

The Pacific Northwest National Laboratory is currently developing a custom software suite capable of automating many of the tasks required to accurately analyze coincident signals within gamma spectrometer arrays. The analysis system is designed to intelligently identify which algorithm best applies to a dataset in order to quantify the activities of isotopes observed in the sample. This capability is required to enable rapid analysis of data collected with a new low-background intrinsic germanium (HPGe) array at PNNL. The HPGe array is designed for high detection efficiency, ultra-low-background performance, and sensitive gamma-gamma coincidence detection. Traditional methods for constructing ultra-low-background detectors were followed during construction of the array, including use of materials known to be low in radioactive contaminants, use of ultra pure reagents, and clean room assembly. The cryostats were constructed mainly from copper electroformed into near-final geometry at Pacific Northwest National Laboratory. The first half of the array, a single cryostat containing 7 HPGe crystals, was recently installed into a new shallow underground laboratory facility with approximately 35 meters water equivalent overburden. This update will present an overview of the new facility, the detector assembly, initial background and measurement results, the status of the 2nd half of the array, and the CASCADES analysis development.

Log: 304. **RECENT DEVELOPMENTS IN THE MANUFACTURE OF LUTETIUM-173 TARGETS.** Taylor, WT(1); Bach, H(2); Nortier, FM(3).

To better model nuclear processes there is an interest in measuring neutron capture cross sections of lanthanide radioisotopes. Natural hafnium targets have been irradiated with 100 Mev protons at the Los Alamos Isotope Production Facility at the Los Alamos Neutron Science Center (LANSCE) to produce neutron poor lutetium radioisotopes. After irradiation, the targets were allowed to cool to allow shorter lived lutetium isotopes to decay. This left predominately Lu-173 with small amounts of Lu-174. The hafnium targets were then chemically processed to isolate the lanthanide fraction through ion exchange techniques. Recent efforts have focused on the separation of lanthanide species to produce an elementally pure lutetium product and the manufacture of small high density Lutetium targets.

Log: 305. **FURTHER DEVELOPMENTS OF A ROBUST ABSOLUTE CALIBRATION METHOD UTILIZING BETA/GAMMA COINCIDENCE TECHNIQUES..** Justin I McIntyre, Mathew W. Cooper, James Ely, Derek Haas, Brian Schrom.

Performing accurate and verifiable measurements is often the most challenging goal for any radiation detector and is especially challenging for the radioxenon detectors deployed by the IMS. Often the accuracy of the measurement is directly tied to how well the detector is calibrated, in both energy and efficiency. Standard methods often rely on using certified sealed sources to determine the absolute efficiency. Similarly, efforts to calibrate the absolute efficiency of radioactive gas cell detectors utilize a number of methodologies which allow adequate calibration but are time consuming and prone to a host of difficult to determine uncertainties. Utilizing methods developed in the 1960's it has become clear that it is possible to achieve much better results that are consistent across a range of isotopes and activities. Even more compelling is the ease with which this process can be used on routine samples to determine the total activity

present in the detector. Additionally, recent advances in the generation of isotopically pure radioxenon samples of Xe-131m, Xe-133, and Xe-135 allow these measurement techniques to achieve much better results than have previously been possible when using mixed isotopic radioxenon sources. This paper will discuss the beta/gamma absolute detection efficiency technique and compare the results obtained using previous methods with these new methods using pure sources of Xe-133, Xe-131m, Xe-135 and Rn-222 as well as a mixed sample of Xe-133/Xe-133m.

Log: 306. **INVESTIGATIONS OF XENON DIFFUSION IN PLASTIC SCINTILLATORS, AND SATURATION AS A SOLUTION TO THE MEMORY EFFECT IN RADIOXENON DETECTION SYSTEMS.** Bläckberg, L(1); Fritioff, T(2); Mårtensson, L(2); Ringbom, A(2); Sjöstrand, H(1); Klintenberg, M(1). (1) Uppsala University, (2) Swedish Defence Research Agency (FOI).

Radioxenon detection systems are used worldwide within the framework of the verification regime of the Comprehensive Nuclear-Test-Ban Treaty. Many of the used systems incorporate a plastic scintillator cell, which acts as a container for the xenon sample during the measurement. One drawback with this setup is that part of the xenon sample diffuses into the plastic scintillator material during the measurement. The result is an unwanted memory effect consisting of residual activity in the detector, even after it has been evacuated and cleaned. One approach to solve this issue is to saturate the plastic detector with stable xenon, or another stable gas, taking the place of radioxenon in the polymer matrix. In this work a plastic scintillator cell was filled with stable xenon, and the pressure in the cell monitored during 250 hours. A pressure drop was observed during the first 100 hours, but after this time the pressure was stabilized. The pressure remained stable when the cell was evacuated and filled again to the initial pressure, indicating that the plastic material was saturated with xenon. From the measured pressure data, both the solubility and diffusivity of xenon in the plastic scintillator material can be estimated, by looking at the total pressure drop and the time evolution of the pressure inside the cell. To evaluate if saturation could be a viable solution to the memory effect problem it is planned to expose a saturated detector to radioactive xenon and measure the residual activity left in the cell after the exposure.

Log: 307. **TITANIUM-BACKED TARGETS PRODUCED VIA THE POLYMER-ASSISTED-DEPOSITION METHOD.** Gothe, OR (1)(2); Nitsche, H (1)(2); Gregorich, KE (2); Gates, JM (2). (1) University of California - Berkeley, (2) Lawrence Berkeley National Laboratory.

With recent advances in ion-source technology, increasingly intense beams are becoming possible. These higher intensities are important to heavy element science, especially given the current trend in exploring ultra low cross section nuclear reactions. Such advances in source technology, as well as attempts to perform measurements at the sub picobarn cross section level, make it increasingly important to create stronger and more resilient targets. We have recently reported on a new method of producing targets that are crack-free and homogenous via polymer-assisted-deposition [PAD]. The oxide layer of these targets is highly controllable and has excellent properties for nuclear reactions. The original method was published using a silicon nitride backing. In recent experiments, we tested lead targets produced using our procedure by irradiating them with a $1\text{-}\mu\text{A }^{40}\text{Ar}$ beam. However, the silicon nitride showed significant damage. We report our continued progress of altering the procedure toward the production of ultra smooth, thin titanium-backed targets produced by polymer assisted deposition. Titanium will increase the durability of the targets because, as a metal, it is able to repair beam damage by self annealing.. Such targets are able to withstand up to a factor two higher beam intensities as due to lower energy absorption and increase the energy resolution at the center of target.

Log: 308. **3-DIMENSIONAL DISTRIBUTION OF CHEMICAL CONSTITUENTS OF TRINITITE.** Koeman, E.C. (1), Wallace, C.M. (1); Simonetti, A. (1) University of Notre Dame.

Trinitite is the melt glass produced from the first nuclear bomb test on July 16, 1945. It is composed of the arkosic sand located at the detonation site (White Sands, Alamogordo, NM) that was fused with the radioactive debris from the bomb. The surface and topographical features of

selected hand samples of trinitite were examined using scanning electron microscopy (SEM), back scattered electron imaging, and energy dispersive x-ray spectral (EDS) analysis. Subsequently, detailed mineralogical and petrographic examination of thin sections for the identical samples ensued also using both SEM and EDS techniques. Location of Pu “hot spots” within thin sections was facilitated using alpha track radiography on CR-39 plastic detectors. These regions were then investigated further using laser ablation-inductively coupled plasma mass spectrometry (LA-ICP-MS) in order to document the distribution of chemical constituents, including those inherent to the “gadget” and from the natural geological environment, at high spatial resolution. Preliminary LA-ICP-MS time-resolved results indicate that samples of trinitite are chemically heterogeneous on a vertical (depth) scale; in particular chemical constituents seem to define two distinct layers, a top section (~20 microns thick) that is enriched in incompatible (natural) elements, and a lower section (~20 microns thick) that is depleted in relation to these same elements. However, the vertical distribution of ^{239}Pu does not follow that of the natural chemical constituents. This study reports transmission electron microscopy (TEM) and focused ion beam (FIB) findings, for the first-time, on the vertical (3-Dimensional) distribution of radionuclides within trinitite at the nanometer scale.

Log: 309. MACRO TALSPEAK SEPARATIONS OF AM-CM FROM FISSION PRODUCTS -- IMPLICATIONS FOR A SINGLE STEP SEPARATION AND ADVANCED FUEL CYCLE IRRADIATION OF AM-CM. E.D. Collins, D.E. Benker, R.J. Vedder, G.D. Del Cul, and R.J. Ellis, Oak Ridge National Laboratory.

Correlations of TALSPEAK distribution coefficient (K_d) dependencies on pH have enabled selection of favorable conditions for macro-scale (multi-g/L) separations of Am-Cm from lanthanide fission products obtained by high burn-up irradiation of plutonium. Preliminary feed solution denitrations were made for simulated PUREX raffinate, using the Modified Direct Denitration process for converting nitrate salts to finely divided oxide powder. This process is similar to the evaporation, denitration, and calcination used for high level waste calcination-vitrification at the LaHague Reprocessing Plant. Redisolution of oxide powder in dilute nitric acid can enable removal of insoluble zirconium, molybdenum, and iron. Non-lanthanide fission products can be removed by extraction of actinides and lanthanides into di-2-ethylhexyl phosphoric acid (HDEHP), the extractant used for the reverse TALSPEAK process. Actinides were partitioned from lanthanides by back-extraction using 1.5M lactic acid containing 0.05M DTPA at pH 3.5 in a mixer settler bank with 12 stripping and 4 solvent back-scrub stages. The correlations for K_d vs. pH were used to select conditions to minimize Am-Cm losses to the lanthanide waste stream. Measured Am-Cm losses were ~ 0.2% (wt). Measured decontamination factors (DF) for the Am-Cm varied from ~ 20 for neodymium to > 100 for lanthanum and > 50 for europium. The overall DF was ~ 40, which is sufficient for Am-Cm recycle in “burnable poison” fuel rods in either heavy water reactors or light water reactors. Reactor physics calculations have shown that the presence of lanthanide fission products in burnable poison fuel rods does not degrade the transmutation of Am-Cm.

Log: 310. CALCULATING RELATIVE IONIZATION PROBABILITIES OF ACTINIDE ISOTOPES BY RESONANCE IONIZATION MASS SPECTROMETRY: A CASE STUDY OF U-235 AND U-238. Isselhardt, BH (1); Prussin, SG (2); Hutcheon, ID (1); Knight, KB (1); Savina, MR (3). (1) Lawrence Livermore National Laboratory, (2) University of California at Berkeley, and (3) Argonne National Laboratory.

Resonance Ionization Mass Spectrometry (RIMS) has been developed as a method to measure uranium isotope abundances. In this approach, RIMS is used as an element selective ionization process to provide a distinction between uranium atoms and potential isobars without the aid of chemical purification and separation. The use of broad bandwidth lasers with automated feedback control of wavelength was applied to the measurement of the $^{235}\text{U}/^{238}\text{U}$ ratio to decrease laser-induced isotopic fractionation. Isotope standards can be used to identify and correct bias in measured isotope ratios, but understanding the source of the laser induced bias from first-principles can be used to improve the precision and accuracy of experimental measurements. A

rate equation model for predicting the relative ionization probability has been developed to study the effect of variations in laser parameters on the measured isotope ratio. The model uses atomic data and empirical descriptions of laser performance to estimate the laser-induced bias expected in experimental measurements of the $^{235}\text{U}/^{238}\text{U}$ ratio. Empirical corrections are included to account for ionization processes that are difficult to calculate from first principles with the available atomic data. The model has been used to predict the measured isotope ratio given the laser power and wavelength distributions. The methodology of this model can be expanded to other elements and can be used to design analyses for trace isotopes that have not been studied before. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL-ABS-516114.

Log: 311. **PROMPT GAMMA CHARACTERIZATION OF ACTINIDES.** Christoph Genreith (1); Matthias Rossbach (1); Eric Mauerhofer (1); Tamás Belgya (2); (1) Institute for Energy- and Climate Research 6 – Nuclear Waste Management and Reactor Safety, Forschungszentrum Juelich GmbH, 52425 Juelich, Germany; (2) Institute of Isotopes HAS. Dept. of Nuclear Research, H-1525 Budapest, POB 77.

We investigate the potential of prompt gamma neutron activation analysis as an alternative to passive and active neutron interrogation to non-destructively determine transuranic isotopes in nuclear waste. Hence, exact knowledge of prompt g-ray energies and intensities and neutron capture cross sections of transuranic isotopes is required. The most comprehensive database on radiative neutron capture cross sections is the prompt g-ray spectrum catalog, developed at the Institute of Isotopes in Budapest and published by the IAEA. It includes prompt g-ray energies and intensities measured over a large energy range from a few keV up to several MeV at once. Therefore a normalization of different experiments is not necessary. This database contains data for isotopes up to uranium, only. Consequently, experiments to study the basic neutron capture data of several transuranic isotopes are required. Measurements of the basic neutron capture data of ^{237}Np and ^{242}Pu samples were performed at the Budapest Research Reactor and latest results will be presented and discussed.

Log: 312. **OPTIMIZATION OF A SEQUENTIAL EXTRACTION PROCEDURE FOR ANALYSIS OF MULTIPLE ACTINIDE ELEMENTS.** Faye, SA(1); Georgetown, BA(2); Clanton, BC(3); Gallardo, AM(1); Sudowe, R(1). (1) University of Nevada, Las Vegas, (2) University of Houston, (3) Florida Memorial University.

To obtain a better understanding of actinide element contaminant mobility in the environment, the association of the contaminants with specific host phases of soils and sediments must be investigated. This investigation is most commonly conducted using sequential extraction techniques, which employ the selective dissolution of soil phases with increasingly aggressive chemical treatment. A sequential extraction procedure by Outola et al (2009) will be the basis for this research. The procedure contains five operationally defined soil fractions: exchangeable, carbonate, Fe/Mn oxides, organic and residual. Variables such as reagent contact time, temperature and reagent concentration will be altered to optimize the procedure for maximum dissolution of actinide elements. Analysis of the resulting leachates containing multiple actinide elements will require sample separation and purification prior to alpha spectroscopy. Extraction chromatography based procedures by Maxwell et al (2006) and Thakur et al (2011) will be investigated as separation methods due to their quick and efficient separation of actinide elements. Reagents used in the sequential extraction procedure will be coupled with each of the two extraction chromatography based separation procedures to examine the recovery and separation efficiency for each fraction of the sequential extraction procedure. Samples will be prepared for alpha spectroscopy using a cerium fluoride microprecipitation method. The optimized sequential extraction/extraction chromatography procedure will be of use for investigating fractionation in varying soil and sediment samples. Information on bioavailability, mobility, origin and mode of occurrence will be furnished from this method. Results will be presented together with recommendations.

Log: 313. **PLUTONIUM(VI) SORPTION TO NANOCAST ORDERED MESOPOROUS CARBON.** Tashi Parsons-Moss, Harun Tüysüz, Deborah Wang, and Heino Nitsche; University of California, Berkeley, CA 94720-1460, USA; Lawrence Berkeley National Laboratory, Nuclear Science Division, Berkeley, CA 94720-8169, USA.

Nanocast ordered mesoporous carbons (OMCs) have a high surface area, good thermal and chemical stability, well-defined pore sizes, and are electrically conductive. Such desirable properties make OMCs attractive in the areas of adsorption, separations, catalysis, energy storage, and electrochemical sensing. The present work compares Pu(VI) sorption to both untreated and chemically oxidized CMK-type OMCs of cubic Ia3d symmetry with Pu(VI) sorption to an amorphous activated carbon. The OMC material was synthesized via nanocasting against a KIT-6 silica template, and characterized by tunneling electron microscopy, infrared spectroscopy, low-angle X-ray diffraction, and nitrogen adsorption isotherms. The oxidized OMC was prepared by treating the OMC material with nitric acid to increase the density of carboxyl groups on the surface. The activated carbon is commercially available and was chosen for its high surface area. Batch studies of all three materials with Pu(VI) solutions in a perchlorate matrix were performed to investigate pH dependence, sorption kinetics, and Pu(VI) capacity, as well as Pu desorption and material reusability. The OMC-Pu interaction was also probed via X-ray absorption spectroscopy. Both OMC materials greatly outperformed the amorphous activated carbon in terms of Pu(VI) sorption kinetics and capacity. In all cases, sorption increased with pH in the range of pH 1-4, and the oxidized OMC showed the fastest binding and greatest capacity in that range. These results indicate that deprotonated carboxyl groups are responsible for binding plutonyl cations to carbon materials under slightly acidic environments.

Log: 314. **AEROSOL DATA PROCESSING PIPELINE.** Brian Schrom PNNL.

Software is being developed at Pacific Northwest National Laboratory for the U.S. National Data Center to handle International Monitoring System data that is part of the Comprehensive Test Ban Treaty Organization network of noble gas stations. A process pipeline consisting of email, an Oracle database, an Apache Tomcat server, a file store, a dynamic link library, and custom Java code handle the data processing and archiving needs for SAUNA and Spalax data. A Java GUI is the primary interface for interacting with the data store and includes functionality for reviewing the data, viewing station histories, and administering the configuration. Current and future concepts will be presented.

Log: 315. **TRAINING IN NEUTRON ACTIVATION ANALYSIS: PROBLEMS AND SUGGESTIONS FOR SOLUTIONS.** Bode, P(1); Bacchi, MA(2); De Nadai Fernandes, EA(2). (1) Delft University of Technology, (2) CENA-USP..

Neutron activation analysis (NAA), either instrumental or radiochemical, is a valuable method of trace element determination in many fields of science for both undergraduate as well as graduate and PhD research programs. NAA is often in the hands of experienced groups at specialized laboratories for see themselves facing the challenge of either acting as a service center to all requests from non-nuclear skilled scientists from the applied fields, or to train these scientists in operating NAA on themselves. And the latter causes many problems in the practice. Within a minimized timeframe (in order to have the users practicing NAA quickly on their own), the NAA laboratory has to pass part of its long time experience on the requirements of, and potential sources of error insample preparation, neutron flux monitoring, geometrical errors and spectral interference problems to newcomers; as well as to make them understand how to monitor and observe such deficiencies. It is worrying to conclude that in this, NAA has not made any progress for half a century: it has remained a technique from and by experts. There is an immediate need for a worldwide concentrated action to improve this situation. In this contribution, suggestions for improvement are given.

Log: 316. **OPTIMIZATION IN NEUTRON ACTIVATION ANALYSIS.** De Nadai Fernandes, EA (1); Bode, P(2); Bacchi, MA (1). (1) CENA-USP, (2) Delft University of Technology.

It is at least remarkable that analysis protocols in neutron activation analysis almost indifferently show the use of counting times of 1-5 min for measurements shortly after irradiation, and 1-4 hours for measurements 1-3 weeks after irradiation. It is not entirely clear what initiates the reasons for such a consistency except for the fact that laboratories may have start copying each others protocols and do so for already more than 50 years. It is barely convincing that such 'standard' counting times of , e.g., 1 hour, provides the best result for any customer of any type of material to be analyzed. It also indicates that possibly the analytical question –and the way the end-user will eventually draw conclusions on basis of the result- has been properly assessed. Often NAA laboratories install their analysis protocol (sample size, irradiation-, decay-, counting time and counting geometry) on basis of optimal performance rather than on basis of optimally providing an answer to the actual question. The latter could be an indicative value at the decimal scale (1-10-100 mg.kg-1) which requires a different approach then for providing a question in which the degree of uncertainty has to be minimized. In this contribution, the performance of NAA when operating less commonly analytical protocols will be compared to its performance at conventional standard conditions.

Log: 317. **INTERACTIONS OF PU(VI) WITH NANOPARTICULATE AND MESOPOROUS FERRIHYDRITE.** L. Kestrel Brogan, Deborah L. Wang, Harun Tüysüz, and Heino Nitsche; Department of Chemistry, University of California, Berkeley, Berkeley, CA 94720, USA; Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA.

2-line ferrihydrite is a common nanoparticulate iron(III) oxyhydroxide, and is well known for its high surface area and reactivity. These properties, paired with the material's low cost and environmental compatibility, make ferrihydrite attractive as a sorbent material for actinide remediation, sequestration, and sensing applications. The disadvantage of ferrihydrite is its low stability due to low crystallinity and the tendency to age to more crystalline iron oxyhydroxide phases. This disadvantage can be mitigated by creating a nanocast mesoporous ferrihydrite, which also has high surface reactivity, but gains stability from the mesoporous structure and the silica nanocasting template. While the addition of silica may also impart some stability to nanoparticulate ferrihydrite, the micron sized mesoporous material may be much more suitable for most applications. Key to this endeavor is a full comparison of the surface reactivities of the two ferrihydrite materials. They have been characterized with respect to the point of zero charge, crystal and particle structure, and surface area. Plutonium(VI) sorption as a function of pH in an aqueous perchlorate matrix has been measured across a wide pH range, and the surface complexes characterized using X-ray absorption spectroscopy (XAS). While no plutonium redox chemistry was expected due to the oxidation state of iron in the system, it was discovered that exposure to light could influence the oxidation state of the plutonium, resulting in Pu(IV). The capacity of the two ferrihydrite materials as well as their redox reactivity with respect to the plutonium(VI) ion will be discussed.

Log: 318. **DETERMINING HPGE TOTAL DETECTION EFFICIENCY USING F-F COINCIDENCE.** Erikson L., Keillor M., Aalseth C., Hossbach T., Mizouni L., Stavenger T., McDonald B., Pacific Northwest National Laboratory.

Both peak and total detection efficiencies are generally needed to calculate sample activity from a gamma spectroscopic measurement, except in the case of isotope specific calibration. This is particularly true when the sample is in close proximity to the detector and corrections for cascade summing effects are required to avoid significant inaccuracy in the result. These corrections use the total detection efficiency to correct for summing-in and summing-out events, and the extent of the correction depends on the geometry and the gamma cascade for the isotope in question. Experimentally determining the total efficiency is labor intensive and requires multiple measurements with a set of single-gamma-emitting standards. Modeling the total efficiency vs. energy may be less time consuming, but is also likely to produce less confidence in the final

result. Pacific Northwest National Laboratory's Radiation Detection and Nuclear Sciences group has constructed a low background 14-crystal HPGe array for sample measurement; in all measurements, samples will be in close proximity to the germanium crystals. This close geometry and the sheer number of efficiency calibrations required for the system have led us to investigate methods to simplify the efficiency calibration procedure. One method we are developing uses the γ - γ coincidence plane to isolate Compton scattering event populations, allowing experimental determination of total detection efficiency values from the measurement of a single mixed isotope standard. The results of Monte Carlo simulation of this method, as well as related experimental results from the detector system, are presented.

Log: 319. **IMPACT OF MEASUREMENT UNCERTAINTIES ON MULTIVARIATE GAMMA-RAY SPECTRUM ANALYSIS.** Orton, CR(1); Jordan, DV(1); Rutherford, CE(1); Coble, JB(1); Fraga, CG(1); Schwantes, JM(1). (1) Pacific Northwest National Laboratory.

Pacific Northwest National Laboratory (PNNL) is developing an on-line, non-destructive analysis, process monitoring technology called the Multi-Isotope Process (MIP) Monitor. The MIP Monitor uses gamma spectroscopy and pattern recognition software to identify and potentially quantify off-normal conditions in gamma ray emitting process streams. Gamma spectroscopy has long been used to measure radioactive nuclide concentration in samples, but using calibrated multivariate techniques to detect and characterize systematic, process-induced changes in radioactive nuclide concentrations is not as well developed. Research is underway at PNNL to explore the basic performance capabilities and uncertainties associated with using multivariate analysis techniques on gamma-ray spectra to quantify nuclide activities and other sample characteristics. We have collected spectra using different detector types viewing a sample set comprising a mixture of radioactive nuclides and applied multivariate analysis methods to compare the spectra. An overview of the multivariate methods and the results from this experiment will be presented.

Log: 320. **ALPHA AND GAMMA RADIOLYSIS OF NUCLEAR SOLVENT EXTRACTION LIGANDS USED FOR AN(III) AND LN(III) SEPARATIONS.** Mezyk, SP(1); Cullen, TD(1); Mincher, BJ (2); Nilsson, M(3). (1) California State University at Long Beach, (2) Idaho National Laboratory, (3) University of California at Irvine.

The separation of the minor actinides from dissolved nuclear fuel remains one of the more formidable challenges in this regard the development of a quantitative understanding of the fundamental chemistry of separations processes. In the USA the most developed process for this separation is TALSPEAK based on the competition between HDEHP (bis(2-ethylhexyl) phosphoric acid) in the organic phase and lactate-buffered diethylenetriaminepentaacetic acid (DTPA) in the aqueous phase. In Europe and Japan, focus has been placed on the use of dithiophosphinic acids, or DMDOHEMA/ bis(triazinyl)pyridine-diamide mixtures. However, all large-scale separation processes must be robust under high-radiation dose conditions. The effects of both gamma and alpha irradiation on solvent systems using steady-state irradiations to measure extraction performance changes (distribution and stripping ratios) and identify decomposition products, and electron pulse radiolysis measurements to determine the kinetic parameters for the important transient species reactions, has been performed. Gamma irradiations have been performed using standard ^{60}Co irradiators, while three approaches have been initiated to identify the best measurement methodology for alpha irradiations: internal isotope irradiation (^{244}Cm , ^{211}At), ion-beam measurements and nuclear reactor induced alpha radiolysis. Results will be presented on our investigations into the radiolysis of the CMPO and TBP ligands in dodecane, where we have now demonstrated that the radiolytic decomposition efficiencies are approximately the same for both gamma and alpha radiolysis, and that the overall decomposition is significantly less when this formulation is irradiated in contact with acidic water. This work is being performed under NEUP Contract DE-AC07-05ID14517.

Log: 321. **INTERACTIONS OF PLUTONIUM(VI) WITH GOETHITE AND ALUMINUM SUBSTITUTED GOETHITE.** Wang, DL (1, 2); Brogan, LK (1, 2); Nitsche, H (1, 2). (1)

Department of Chemistry, University of California, Berkeley (2) Nuclear Science Division, Lawrence Berkeley National Laboratory.

For risk assessment and to demonstrate good stewardship at radioactively contaminated and nuclear waste storage sites, it is important to understand the molecular-level interactions of plutonium with environmental matrices such as mineral surfaces. Because Pu is highly redox active, contact with minerals found in nature could reduce the more soluble oxidation states of Pu(V) and Pu(VI) to the less soluble Pu(IV) oxidation state, thus limiting Pu mobility in the environment. In this study, the interactions of Pu(VI) with two different mineral surfaces were compared: the iron oxide mineral goethite (Fe(III)OOH) and a 1% aluminum-substituted goethite. The Al-substituted goethite was synthesized because minor elemental substitutions are often present in naturally-occurring minerals. Both minerals were characterized with respect to the powder X-ray diffraction (XRD) pattern, point of zero charge, surface area, and elemental content. The Pu(VI) sorption as a function of pH was measured for both mineral surfaces via batch experiments with aqueous perchlorate Pu solutions. X-ray absorption spectroscopy (XAS) was used to investigate the redox chemistry that occurs between Pu(VI) and the mineral surfaces, as well as the local coordination environment of the Pu ions. While reduction to Pu(IV) was not expected for either mineral surface, reduction to Pu(V) and Pu(IV) for both systems was observed from the X-ray absorption near-edge structure (XANES) measurements. Data analysis of the extended X-ray absorption fine structure (EXAFS) is currently underway to determine whether the sorbed Pu is bound to the mineral surface in an inner- or outer-sphere coordination.

Log: 322. **ELUCIDATING THE RADICAL KINETICS INVOLVED IN THE RADIOLYTIC DESTRUCTION OF LANTHANIDE-COMPLEXED DTPA.** Thomas D. Cullen (1); Stephen P. Mezyk (1); Leigh R. Martin (2); Bruce J. Mincher (2). California State University, Long Beach (1), Idaho National Laboratory (2).

The recycling of nuclear fuels to extract the remaining useful actinides is one of the most important strategies required for viable nuclear power in the future, as geologic waste disposal of high-level radioactive waste has considerable negative impacts on the environment. As many of these strategies are based on solvent extraction processes, our aim is to establish the radiolytic stability of the extraction ligands. The oxidative stability of the lanthanide metal-complexed extraction ligand DTPA (diethylenetriaminepentaacetic acid) has been investigated through its reaction rate constant with the hydroxyl radical under TALSPEAK acidic pH conditions for Eu³⁺, Lu³⁺ and Gd³⁺. Specific rate constants were obtained over a range of pH conditions using thiocyanate competition kinetics. These values are slightly greater than the corresponding rate constants determined for only the DTPA protonated ligands at these pH's, determined by the same methodology. In addition based on literature pKa values and composite rate constant measurements at multiple pH values, individual rate constants for the species DTPA-H5, DTPA-H4- and DTPA-H32- of 2.69×10^9 , 3.40×10^9 and $3.26 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$ have been determined. Using these values, we have now also extended this approach to determine the temperature-dependent hydroxyl radical reaction rate constants with the Eu-DTPA.

Log: 323. **MEASUREMENT OF ¹³⁷CS IN SOIL IN KOREA BY LOW-LEVEL BACKGROUND GAMMA-RAY SPECTROMETER.** Lee, SH; Lee, SA; Lee, JM; Park, TS; Lee, KB. Korea Research Institute of Standards and Science.

Cs-137 in the soils collected from Korea from 2006 to 2008 was measured by using the low-level background gamma-ray spectrometer which was designed and developed by KRISS (Korea Research Institute of Standards and Science). The objectives of this study are to evaluate the new developed low-level background gamma-ray spectrometer and consequently to provide information on the horizontal and vertical distribution of Cs-137 in the soil in Korea. Cs-137 concentrations in surface soil varied from 12.8 ± 0.9 to $108 \pm 4 \text{ Bq/kg}$ and the vertical profiles of Cs-137 from Nari basin in Ulleung Island in the East Sea/Sea of Japan and Seongsan Ilchulbong Peak, Jeju Island showed a higher concentration at the surface layer and gradually decreased. On the other hand, the Cs-137 concentration in the soils collected from Bukhan Mountain

National Park in Seoul showed a subsurface maximum and decreased with depth. The Cs-137 inventories in the soil column were calculated to be 1830 to 4360 Bq/m² with a mean of 2770 Bq/m² which was the same order of magnitude as the global fallout inventories in the mid-latitude region of the Northern Hemisphere.

Log: 324. **IMAGING OF COSMIC-RAY INDUCED FISSION FOR TREATY MONITORING.** Borozdin, KN (1); Morris, CL (1); Perry, JO (1); Bacon, JJ (1). (1) Los Alamos National Laboratory.

We describe a novel method of detecting, identifying and imaging nuclear materials for treaty verification. Our technique is based on tracking cosmic-ray muons into the volume and detecting neutron and gamma emissions generated by the muons. The n/γ measurements provide us with detection and identification of the materials, while cosmic-ray tracking enables their localization. Combining these sources of information allows us to improve nuclear material detection including: 1) material isotope identification; 2) imaging with one-sided access to the volume; 3) n/γ background rejection with temporal coincidence and imaging. We performed initial demonstration of fission signal detection at Los Alamos using Mini-Muon Tracker in coincidence with neutron and gamma-ray detectors. With our detector we have demonstrated imaging of a fissile material based on cosmic-ray muon tracking. In our experiments we have used 1-liter (~20 kg) cubes of both depleted uranium and low-enriched uranium, as well as non-fissile materials (including aluminum, iron and lead) of different shapes. Comparison of the signals from DU and EU allows us to demonstrate how we can distinguish uranium from lower-Z materials and also perform uranium isotope identification with our technique. We discuss capabilities of our measurements and potential application of our technique for treaty monitoring.

Log: 325. **CHALLENGES IN DEVELOPING RADIOCHEMICAL METHODOLOGY FOR SRS HIGH ACTIVITY WASTE MATRICES.** DiPrete, DP (1); DiPrete C.C. (1); Coleman C. J. (1); Malek M. A. (1); White T. L.(1); Pennebaker F. M. (1); Jones M. A. (1). (1) Savannah River National Laboratory.

Waste cleanup currently underway at the Savannah River Site have created a need to characterize the inventories of various waste tank heels prior to tank closure. The heels of each tank slated for closure have unique chemical constituents as well as unique radiological distributions compared to previously characterized heels. Continuous programmatic evaluations of allowable closed tank radiological inventories result in steadily decreasing detection limit targets for isotopes required for characterization. Numerous target isotopes are often required to be characterized to levels as much as 10 orders of magnitude below radioactive concentrations of interfering isotopes. New isotopes continue to be added to the characterization lists. A recent tank closure program required characterization of 54 radioisotopes on 36 high activity waste samples with a turn-around of several months. Dose rates of samples required initial manipulations to be carried out remotely in the Shielded Cells Facility, followed by additional hands-on radiohood operations. Methodology for 6 isotopes had to be developed from the ground up, while the existing methodology for a number of previously analyzed radioisotopes had to be greatly modified in order to meet the data quality objectives in the unusual matrix. An overview of methodology and available resources used to rapidly develop protocols capable of meeting these Tank Closure radiological characterization requirements will be discussed as well as some specifics arising from recent program efforts.

Log: 326. **THE SPATIOTEMPORAL VARIATIONS OF RADON IN GROUNDWATER AND ITS RELATIONSHIP TO GEOLOGY.** Ko, KS (1); Ha, KC (1); Kao, DC (1), Lee, GY (1), Yoon, YY (1), (1) Korea Institute of Geoscience & Mineral Resources (KIGAM).

Radon is generally produced through the radioactive decay of uranium and radium, which is naturally present in soil and minerals in bedrock. There is no regulation for radon in Korea, but US EPA proposed 4,000pCi/L of radon in drinking groundwater as potential carcinogen to reduce

the public health risks. The basic groundwater survey has been executed to secure safe groundwater as sustainable water resources in Korea since 1970s. In this study, we investigated the spatial and temporal variation of groundwater radon in Asan city which has several hot springs and is located at the central region of Korea. The geology of Asan city is mainly composed of Jurassic granite and Precambrian schist. To investigate the spatial and temporal variation of radon in groundwater, total 176 groundwater samples were collected from 159 wells installed at bedrock or alluvial aquifer. Twelve and fifteen percents of samples exceeded 4,000pCi/L of radon in bedrock and alluvial wells, respectively. It is considered that high radon of groundwater in Asan is correlated to geology of granite. High concentrations of radon in schist area may be produced by the effect of biotite granite below schist, which play a role as radon enrichment in groundwater. Fluoride also showed high values as radon in schist area, which is also caused by the weathering of biotite granite with fluoride bearing minerals. The investigation results of the study will be helpful to manage and provide safe and clean water resources plan using groundwater.

Log: 327. **DISTRIBUTION OF ^{137}Cs , ^{131}I AND $^{239,240}\text{Pu}$ CONCENTRATIONS IN THE RAINWATER IN KOREA AFTER FUKUSHIMA NUCLEAR ACCIDENT.** Heo, DH; Lee, SH; Kang, HB; Oh, PJ; Lee, JM; Park, TS; Lee, KB; Suh, JK. Korea Research Institute of Standards and Science.

Radionuclides such as ^{137}Cs , ^{131}I and $^{239,240}\text{Pu}$ in rainwater have been analyzed by KRISS in Korea after Fukushima nuclear accident in March 2011, to identify their behavior in the atmosphere and to assess radionuclide contamination through atmospheric input. The preliminary results are showing that the observed concentrations of these radionuclides in the rainwater samples were extremely higher than those measured before Fukushima accident, and gradually decreased with time. In this study, the most recent results will be discussed

Log: 328. **TRIVALENT ACTINIDE AND LANTHANIDE SEPARATIONS USING DGA EXTRACTION CHROMATOGRAPHY.** Jennifer Shusterman (1), Julie Champion (1), and Heino Nitsche (1). (1) University of California, Berkeley.

The effective separation of trivalent actinides and lanthanides is one of the key steps in successfully partitioning spent nuclear fuel. Thus far the process has been hindered by the chemical and physical similarities of these elements. We have explored the separation properties of the DGA (N,N,N',N'-tetra-n-octyldiglycolamide) extraction chromatography resin from Eichrom. Although the trivalent actinides and lanthanides are similar in most respects, they do exhibit slightly different complexation behavior with the DGA ligand in acidic media, making the resin a promising choice for successful procedures. Both hydrochloric and nitric acid of varying concentrations have been explored as eluents to determine the role of the acid in binding and the cause of the binding differences. Based on these differences in complexation, we have designed a procedure to separate Eu(III), Am(III), and Cm(III) from La(III), and have achieved successful separations of all three metals from the La(III). Experiments are currently being conducted for a separation of Eu(III) from Am(III), a process that directly models the fuel processing stage to remove trivalent actinides from trivalent lanthanides, which are some of the heavy fission products present in the spent fuel. In parallel, we are developing a separation of Cm(III) from Am(III) as a final partitioning stage for the trivalent actinides.

Log: 329. **FUKUSHIMA RADIONUCLIDES IN THE BRATISLAVA AIR – A COMPARISON OF MONITORING AND MODELING RESULTS.** P.P. Povinec, I. Šýkora, M. Gerab, K. Holýa, L. Brešťáková, A. Kováčik, J. Bartočík, M. Gažák, aDepartment of Nuclear Physics and Biophysics, Faculty of Mathematics, Physics and Informatics, Comenius University, 84248 Bratislava, Slovakia, bDepartment of Astronomy, Physics of the Earth and Meteorology, Faculty of Mathematics, Physics and Informatics, Comenius University, 84248 Bratislava, Slovakia, cMicroStep, Monitoring and Information Systems, Bratislava, Slovakia.

Lagrangian dispersion modeling was applied to trace radionuclide transport from Fukushima to Europe. A reasonable agreement was achieved when comparing predicted values with radionuclide concentrations measured in the Bratislava air which showed three concentration maxima, associated with different air masses present in the Central Europe. A comparison with data obtained during the Chernobyl accident showed negligible radionuclide levels obtained during the Fukushima accident. The ^{131}I and ^{137}Cs vs. ^7Be aerosol activity records showed that the increases in ^{131}I and ^{137}Cs activity concentrations were accompanied by ^7Be increases, however, no significant correlation was obtained. This would indicate that both processes, i.e. a horizontal transport of the radioactive plume from Fukushima at higher altitudes, as well as a vertical transport of Fukushima radionuclides and ^7Be within the troposphere were responsible for observed radionuclide concentrations. The observed $^{131}\text{I}/^{137}\text{Cs}$ activity ratios in the Bratislava air also indicated a presence of different air masses in the sampling site.

Log: 330. **STUDIES ON BATCH SORPTION METHODOLOGIES: EU SORPTION ONTO KIVETTY GRANITE.** Holgersson, S., Chalmers University of Technology, Division of Nuclear Chemistry.

Nuclear power is a source for energy production that have almost no greenhouse gas emissions compared with the burning of fossil fuels, yet also have the capacity to replace fossil fuel plants on the large scale which is demand from industrial production needs. However, the handling and storage of nuclear waste is often considered as the achilles heel of nuclear industry. The most viable concept today for handling nuclear waste is the final repository storage, usually to be located in a deep geological formation. To be able to calculate the future course for a final repository for nuclear waste may be one of the more challenging tasks in computer modeling. The batch sorption method for conducting experiments, with radionuclide sorption onto crushed and sieved solid phases, is commonly used for obtaining sorption coefficients (R_d) needed in different hydrogeochemical transport models for estimating a possible future radionuclide release. It is therefore of importance that the methodology for batch sorption experiments is robust and standardized so that same results can be obtained by any laboratory responsible for delivering the data. In this work six different methodological features of the batch sorption experiment were investigated: total metal concentration, solid:liquid (S:L) ratio, phase separation method, wall sorption measurement method, pre-treatment of solid phase and phase mixing method. The system studied was ^{152}Eu sorption onto crushed Kivetty granite of the 0.045-0.2mm fraction, using synthetic Olkiluoto saline water for reducing conditions with three initial pH values set to 8.5, 9 and 10. The standard conditions were $[\text{Eu}]_{\text{tot}}=5 \cdot 10^{-7}\text{M}$, S:L = 1:20 and samplings were made at 3 days, 1 week, 1, 3 and 6 months. The major causes on data spread have been identified as pH drift, sorption onto filters used for phase separation and precipitations of radiotracer.

Log: 332. **NUCLEAR SPALLATION REACTIONS IN YTTRIUM WITH 120 GEV PROTONS.** Suzuki, H (1); Sekimoto, S (1); Yashima, H (1); Shibata, S (1); Matsumura, H (2); Toyoda, A (2); Oishi, K (3); Matsuda, N (4); Kasugai, Y (4); Sakamoto, Y (4); Nakashima, H (4); Nakamura, T (3); Boehnlein, D (5); Coleman, R (5); Lauten, G (5); Leveling, A (5); Mokhov, N (5); Ramberg, E (5); Soha, A (5); Vaziri, K (5). (1) Kyoto University Research Reactor Institute, (2) High Energy Accelerator Research Organization, (3) Shimizu Corporation, (4) Japan Atomic Energy Agency, (5) Fermi National Accelerator Laboratory.

The production cross sections by high energy protons are important in the field of cosmo-chemistry for deciphering the cosmic-ray irradiation history. These data also serve as a nuclear database for the estimation of residual radioactivity in the accelerator facilities. Those cross sections are indispensable for studying the formation mechanism of nuclides by nuclear spallation, fission reactions and fragmentation processes, especially in the field of nuclear chemistry. Although the cross section data for the production of nuclides via nuclear spallation reactions that are close to target nuclides have been well-studied, proton cross sections in the energy range above 100 GeV have rarely been measured, except for those produced from copper and gold targets. In this paper, we are reporting the first measurements of the reaction

cross section for yttrium production induced by 120-GeV protons. The irradiations were performed at the Meson Test beam facility in Fermi National Accelerator Laboratory (FNAL). The experimental results obtained for yttrium target are consistent with the excitation functions which were estimated from literature values. We intend to discuss high energy nuclear reaction mechanism using the data obtained in this work.

Log: 332. **ISTC ACTIVITIES ON NUCLEAR FORENSICS** . Svetlana Genisaretskaya, ISTC.

ISTC is developing a new targeted initiative entitled Scientific and Technical Support Against the Illicit Trafficking of Nuclear and Radioactive Materials (NRM TI). The objective of this Targeted Initiative is to provide scientific and technical support for projects that can be fulfilled in CIS countries and Georgia, which will support measures against the illicit trafficking of nuclear and radioactive materials. ISTC activities under NRM TI may include research work such as Partner Projects, support for workshops, training programs, and equipment and material purchases necessary to implement described activities. In addition, support of CIS consortia and clusters working in the area of nuclear forensics, support for upgrades to regulations to meet national and international standards including national and international training, certification and third party audits, and facilitation of academic and industry partnerships. The following actions are also to be considered within the framework of the NRM TI a) to develop a set of legal norms that may assist and enable quick actions of government bodies to detect illegally stored or transported nuclear and/or radioactive material, as well as detection of loss of such materials in the legal places of storage, and b) to facilitate further coordination of activities between various stakeholders active in the area. The NRM TI is being undertaken in cooperation with ISTC partner - U.S. Department of Energy's Global Initiatives for Proliferation Prevention Program (GIPP). New Partners from ISTC's funding Parties who are interested in developing nuclear forensics projects in CIS countries are welcome to participate in this TI

Log: 333. **SPATIAL AND SEASONAL VARIATIONS OF ^{210}Po AND ^{210}Pb ACTIVITY CONCENTRATIONS IN MYTILUS GALLOPROVINCIALIS FROM CROATIAN COAST OF THE ADRIATIC SEA.** Martina Rožmarić Mačefat(1), Matea Rogić(1), Ljudmila Benedik(2), Marko Štrok(2), Delko Barišić(1); 1-Ruđer Bošković Institute, Zagreb, Croatia 2- Jožef Stefan Institute, Ljubljana, Slovenia.

Results of two years monitoring of ^{210}Po and ^{210}Pb activity concentrations in soft tissue of the species *Mytilus galloprovincialis* from Croatian part of the Adriatic coast are presented. The samples were collected at thirteen coastal stations (some of which are also a part of Mediterranean Mussel Watch Project) in spring and autumn of 2010 and 2011. All the collected mussels were ranging between 4 cm and 6 cm in shell length. After sample pretreatment lead and polonium were radiochemically separated on Sr resin. ^{210}Po was determined by alpha-spectrometry after selfdeposition on a silver disc and ^{210}Pb was determined by a low-level gas proportional counter after PbSO_4 precipitation. The results of ^{210}Po activity concentrations were found to vary between $(115.4 \pm 6.9) - (1228.0 \pm 146.1) \text{ Bq kg}^{-1}$ dry weight while ^{210}Pb activity concentrations were much lower and in range $(15.2 \pm 7.3) - (68.3 \pm 15.5) \text{ Bq kg}^{-1}$ dry weight. Higher ^{210}Po and ^{210}Pb activity concentrations were determined in spring period while the inter-site differences seen in ^{210}Po oncentrations can be due to natural background levels of sites. The $^{210}\text{Po}/^{210}\text{Pb}$ activity concentration ratios in all cases exceeded unity for all mussel samples and ranged between 5.9 and 30.7.

Log: 334. **A REINTERPRETATION OF C5-BTBP EXTRACTION DATA, PERFORMED IN VARIOUS ALCOHOLS.** Elin Löfström-Engdahl, Emma Aneheim, Christian Ekberg, Nuclear Chemistry, Chalmers University of Technology, Sweden.

The used nuclear fuel taken from a reactor has to be stored for a long time. The long storage time is mainly due to the high radiotoxicity of the actinides. If, however, these long lived actinides could be separated from the rest of the used fuel and transmuted into less radiotoxic or stable nuclides, the radiotoxicity of the used fuel would decrease. However, especially the separation of the

trivalent actinides from the trivalent lanthanides is hard to achieve. One way of achieving this separation is through solvent extraction. A well investigated extractant for achieving this separation is the C5-BTBP. C5-BTBP has shown to be able to perform the extraction of An(III) while leaving mostly of the Ln(III) in the aqueous phase. This extraction has been reported in literature to be possible from several aqueous phases and into several organic solvents. Anyway, the extraction as well as the separation is changed when the diluent is varied. Any clear explanation of these diluent effects are not stated in literature. Anyway, in this paper already presented extraction data is reinterpreted in a novel way. The data used is an extraction series where the only parameter varied is the carbon chain length of the diluent (straight chained alcohols). However, this variation in chain length gives a variation in several physical parameters, therefore, the extraction and separation data is here correlated with several physical parameters of the diluent. The conclusion is that the same trend is seen when correlating the data with several parameters.

Log: 335. **ON THE CATEGORIZATION OF URANIUM MATERIALS USING LOW RESOLUTION GAMMA RAY SPECTROMETRY.** A. Vesterlund, T. Ulvsand, K. Lidström, G. Skarnemark, C. Ekberg, H. Ramebäck.

In order to characterize uranium materials during e.g. nuclear safeguard inspections and in initial stages of nuclear forensic investigations, hand-held low resolution gamma ray detectors such as NaI(Tl) with automatic radionuclide identification as well as uranium categorization capabilities may be used. However, to be able to characterize uranium correctly by low resolution gamma spectrometry using such instruments, both matrix and shielding of the material must be known. In this paper, simulated response curves for a number of matrices applied on NaI(Tl) scintillation detector spectra, show that the result of the characterization is strongly dependent of the physical properties of the uranium material. Recommendations of how to minimize the possibility of misclassification are discussed.

Log: 336. **EFFICIENT SEPARATION AND DETERMINATION OF TECHNETIUM IN CONTAMINATED ROCKS.** W. Lixiong, T. Lei, Y. Tongzai.

Fission products are very important signature of many nuclear activities, such the reprocessing of spent nuclear fuels and nuclear weapons tests, as well as in the case of nuclear accidents. Moreover, the characterization of environmental samples collected in the vicinity of the nuclear facilities is necessary in order to control the presence of the radionuclide in the environment. Technetium-99 is one of several long-lived fission products, and when detected in the environment, they can give an indication of specific nuclear activities. This work reports a approach that can efficiently separate and determine the order of technetium-99 in quantities order of gram contaminated rock. The samples were decomposed by potassium hydroxide/potassium nitrate, and then, the technetium was isolated from the sample by technique combining: solvent extraction and exchange. After separation, technetium-99 was measured by isotope dilution mass spectrometry with technetium-97 as a spike. With this method, may be find more general application in studies of environmental contamination by nuclear materials.

Log: 337. **RADIOCESIUM IN HONEY SAMPLES COLLECTED AT AN URBAN SITE IN TOKYO IN THE YEAR 2011.** Nogawa, N(1); Minai, Y(2,3); Maruhashi, T(2); (1) University of Tokyo, (2) Musashi University, (3) Nedzu Chemical Laboratory.

Concentrations of radio-caesium in some honey samples collected in Tokyo were determined to evaluate the path of the radioactivity in urban ecosystem after the severe accident at the Fukushima-daichi nuclear power plant. The honey samples were collected in the year 2010-2011 from the honeycombs set at the roof of a hall at Musashi University, locating in the western area of the metropolitan Tokyo. It is estimated that the honeybees take nectar from flowers in some public gardens and private properties within several hundred meters around the school in the residence area for middle-to-high class citizens in Tokyo. Sixty grams of honey was taken to

determining the radioactivity by means of a Ge detector-based gamma-ray spectrometer. Efficiency for the counting in the whole range of the energy spectrum was calibrated by measurement of the in-house honey standard containing the known amount of Eu-152. Cesium-134 and Cs-137 were determined for the samples collected after the Fukushima accident in March, 2011. Radioactivity of those nuclides reached about 10 Bq/L for the honey samples collected in April, 2011 whereas that for the samples in 2010 was under the detection limit. The radioactivity is much lower than the tentative value for foods allowed for distribution. The amount of radio-caesium in the honey samples decreased gradually from spring to summer. This indicates that radio-caesium were removed from the urban ecosystem for honey production, possibly involving rain-fall, soil, plants, and honeybees.

Log: 338. **TECHNETIUM CHEMISTRY IN A NOVEL GROUP ACTINIDE EXTRACTION PROCESS.** Emma Aneheim, Christian Ekberg, Mark Foreman, Alexander Littley, Gunnar Skarnemark - Nuclear Chemistry & Industrial Materials Recycling, Department of Chemical and Biological Engineering, Chalmers University of Technology, SE 412 96 Gothenburg, Sweden.

A newly developed method for advanced reprocessing of used nuclear fuel is the Group ActiNide Extraction (GANEX) process. It is a liquid-liquid extraction process that aims at extracting all the actinides as a group, after the removal of bulk uranium from dissolved used nuclear fuel. At Chalmers University of Technology in Sweden a solvent that utilizes TriButyl-Phosphate (TBP) and a molecule from the Bis-Triazine BiPyridine (BTBP) class of ligands dissolved in cyclohexanone has been developed for the use in a GANEX process. Previously the system has not been tested with the presence of technetium that is one of the major fission products. Technetium is also often a problem within reprocessing as well as a fission product which chemistry differs from many other metals by the formation of anionic species. Therefore, this work focuses on how the described GANEX extraction system performs on technetium under different conditions. It was shown that technetium is readily extracted by the GANEX solvent and that cyclohexanone is the main extractant. Technetium extraction was also tested with the presence of uranyl nitrate and it was found that the presence of uranium inhibited the technetium extraction. It was also discovered that an increased nitrate concentration in the aqueous phase and an addition of other fission/corrosion products both inhibited the technetium extraction. Besides this it was found that irradiation of the GANEX solvent to large doses (>1MGy) increased its technetium extraction capability. In addition to these findings potential methods of preventing the technetium extraction with the GANEX solvent was also explored.

Log: 339. **APPLICATION OF MG-28 TO THE KINETIC STUDY OF MG UPTAKE BY A RICE PLANT.** Keitaro Tanoi (The Univ. of Tokyo), Takayuki Saito (The Univ. of Tokyo), Naoko Iwata (The Univ. of Tokyo), Natsuko I. Kobayashi (The Univ. of Tokyo), Atsushi Hirose (The Univ. of Tokyo), Yoshimi Ohmae (The Univ. of Tokyo), Ren Iwata (Tohoku Univ.), Hisashi Suzuki (NIRS), Tomoko M. Nakanishi (The Univ. of Tokyo).

We present the preparation of Mg-28 to study Mg uptake manner by a rice root. Since there is no conventional Mg tracer available, we prepared Mg-28 by irradiating a pure Al target by an alpha-particle beam. As a result, approximately 4 MBq of carrier-free Mg-28 was produced. To study the Mg uptake manner of rice, the time-dependent uptake manner was studied. The radioactivity of Mg-28 in the rice sample was measured by a NaI(Tl) scintillation counter. It was found that the amount of Mg-28 in rice root was increased constantly during 30 min of application. Then the Mg loaded root for 15 min was successively rinsed with the non-Rl Mg solution to remove the Mg-28 in the free space or the apoplastic space and leave only Mg-28 inside the root cells. Within about 8 min, almost all of the Mg-28 kept in the free space or apoplastic space was found to be washed away. After that, the Mg-28 efflux from the symplast was observed at constant rate. The Mg-28 tracer was shown to be an indispensable tool for kinetic analysis of Mg uptake and translocation in plants.

Log: 340. **CONTROLLING LOW-RATE SIGNAL PATH MICRODISCHARGE FOR AN ULTRA-LOW-BACKGROUND PROPORTIONAL COUNTER.** EK Mace, CE Aalseth, RM Bonicalzi, AR Day, EW Hoppe, ME Keillor, AW Myers, CT Overman, A Seifert, Pacific Northwest National Laboratory.

Pacific Northwest National Laboratory (PNNL) has developed an ultra-low-background proportional counter (ULBPC) made of high-purity copper. These detectors are part of an ultra-low-background counting system (ULBCS) in a newly-constructed shallow underground laboratory at PNNL. To control backgrounds, the current preamplifier electronics are located outside the ULBCS shielding. Thus the signal from the detector travels through almost one meter of cable and is potentially susceptible to high voltage microdischarge and other sources of electronic noise. Based on initial successful tests, commercial cables and connectors were used for this critical signal path. Subsequent testing across different batches of commercial cables and connectors, however, showed unwanted (but still low) rates of microdischarge noise. To control this noise source, two approaches were pursued: first, to carefully validate cables, connectors, and other commercial components in this critical signal path, making modifications where necessary. Second, to develop a custom low-noise, low-background preamplifier that can be integrated with the ULBPC and thus remove most commercial components from the critical signal path. This integrated preamplifier approach is based on the Amptek A250 low-noise charge-integrating amplifier module. The initial microdischarge signals observed are presented and characterized according to suspected source via pulse-shape analysis. Each of the approaches for mitigation is described, and the results from both compared with each other and with the original performance seen with commercial cables and connectors.

Log: 341. **APPLICATION OF SELF-ORGANIZING MAPS TO AN ARCHAEOMETRIC STUDY OF AMAZONIAN CERAMICS.** Hazenfratz R.; Munita, C. - Nuclear and Energy Research Institute, IPEN-CNEN/SP.

This paper comprises the application of one artificial neural network technique to the analysis and interpretation of archaeometric data. The main objective of this work was to compare the results obtained by some multivariate statistical techniques, which may rely on hypothesis of normality and homoscedasticity of data, to a pattern recognition technique which does not rely on any underlying hypothesis regarding the data distribution. The self-organizing map was chosen for such an aim. This technique is useful to visualize multidimensional data in two or three dimensions, as in the case of data produced by instrumental neutron activation analysis (INAA), which may yield several chemical elements determined for each sample. An elemental concentration data set of 24 elements (As, K, La, Lu, Na, Nd, Sb, Sm, U, Yb, Ba, Ce, Co, Cr, Cs, Eu, Fe, Hf, Rb, Sc, Ta, Tb, Th and Zn) obtained by INAA of 169 samples was analyzed in self-organizing maps. The results were compared with some multivariate statistical techniques, cluster, principal component and discriminant analysis. The archaeological ceramic fragments were excavated in the archaeological sites Lago Grande and Osvaldo in the central Amazon, Brazil. The preliminary interpretations showed that, in general, the self-organizing maps were in accordance with the multivariate patterns identified by other techniques.

Log: 342. **INVESTIGATIONS OF DIFFERENT SAMPLE MATRICES IN URANIUM ABUNDANCE ANALYSIS USING THE MTE METHODOLOGY.** Black CK; Zuleger E; Horta-Domenech J; Vargas Zuniga M. Institute for Transuranium Elements (ITU).

The ITU in Karlsruhe routinely analyses a multitude of samples from a wide range of internal and external customers. High through-put analysis techniques are employed using meticulous care to ensure accurate, precise and timely results are provided. Whilst the samples are generally the same there is a diversity of sample matrices, each of which requires individual chemical processing for analysis. This can give rise to differences in analytical run performance. TIMS is used for isotopic analysis of Uranium and Plutonium to determine abundance and concentration information. More recently there has been an increase in requests for Uranium minor isotope analysis for the U^{234} and U^{236} isotopes. At ITU we employ the MTE methodology[1] for minor

isotope analysis and have observed varying run quality between samples of differing matrix. This presentation will provide some insight into the investigations carried out into the differences observed and how they can be improved for consistent quality routine analysis. [1] S. Richter et al, J. Anal. At. Spectrom., 2011, 26, 550-564

Log: 343. **AN IMPROVED MEASUREMENT METHOD FOR ^{89}Sr AND ^{90}Sr AFTER A REACTOR ACCIDENT OR NUCLEAR DETONATION, BASED ON STRONTIUM SEPARATION CHEMISTRY.** S. Holmgren, A. Tovedal, S. Jonsson, U. Nygren, H. Ramebäck.

Measurement of ^{89}Sr and ^{90}Sr after nuclear power plant accident and nuclear explosions will be limited by short lived radionuclide interferences. In this paper an improved separation method, which takes these interferences into consideration is presented. The method is based only on strontium separation chemistry and the radioactivity for both ^{89}Sr and ^{90}Sr , via ^{90}Y , is determined by Cherenkov counting. By using radioactive ^{85}Sr as the only yield tracer this offers the advantage of determining the total chemical yield of the analysis by gamma spectrometry alone. The method development is based on theoretical calculations of potential interfering fission products present after a nuclear weapon detonation or a nuclear power plant accident. The method was applied and validated on relevant reference material, spiked strontium standard solutions and samples containing radionuclides, who are known interferences, at a composition representing that from e.g. a reactor accident. By evaluating the relative combined uncertainties of the yield determination a measurement of the reliability of the method was given.

Log: 344. **DETERMINATION OF URANIUM FISSION INTERFERENCE FACTORS FOR INAA.** Ribeiro Junior, IS (1); SAIKI, M (1); Genezini, FA (1); ZAHN, GS (1). (1) Instituto de Pesquisas Energéticas e Nucleares - IPEN.

Instrumental neutron activation analysis (INAA) is a very suitable technique for the determination of several elements in different kinds of matrices. However when a sample contains high uranium concentrations this method presents interference problems from uranium fission products. The same radioisotopes used in INAA in the determination of the elements such as lanthanides, Mo, Zr among others are formed in the uranium fission. These radioisotopes are La-140, Ce-141, Ce-143, Mo-99, Nd-147, Sm-153 and Zr-95. This study aimed to the determination of uranium fission interference factors to be used in the correction of the contribution of fission products. The experimental procedure consisted of irradiating synthetic standards of the elements to be determined and of U in IEA-R1 nuclear research reactor followed gamma ray spectrometry using HPGe detector. The thermal and epithermal neutron flux were determined using gold monitor. Experimental and theoretical interference factors obtained were statistically evaluated and comparison made with literature data demonstrates good agreement. The findings of this preliminary study suggest the application of factors in the INAA of geological materials containing high levels of uranium.

Log: 345. **FISSION YIELD MEASUREMENTS FROM HIGHLY ENRICHED URANIUM IRRADIATED INSIDE A BORON CARBIDE CAPSULE.** LA Metz1, JI Friese1, E Finn1, LR Greenwood1, RF Payne1, CC Hines2, MD King2, KM Henry2, and DE Wall2 1. Pacific Northwest National Laboratory, 902 Battelle Blvd, P.O. Box 999, Richland WA 99352 2. Nuclear Radiation Center Dodgen Research Facility, Washington State University, Pullman WA 99164.

A boron carbide capsule was previously designed and tested by Pacific Northwest National Laboratory and Washington State University for spectral-tailoring in mixed spectrum reactors. The presented work used this B_4C capsule to create a fission product sample from the irradiation of highly enriched uranium with a fast fission neutron spectrum. An HEU foil was irradiated inside of the capsule in WSU's 1 MW TRIGA reactor at full power for 200 min to produce 5.8×10^{13} fissions. After three days of cooling, the sample was shipped to PNNL for radiochemical separations and analysis by gamma, beta, and alpha spectroscopy. Fission yields for products were calculated from the radiometric measurements and compared to published evaluated fast-

pooled nuclear data. Reactor dosimetry measurements were also completed to fully characterize the neutron spectrum and total fluence of the irradiation.

Log: 347. **THE DEVELOPMENT OF RADIOACTIVE SAMPE SURROGATES FOR TRAINING AND EXERCISES**. Martha Finck, Bevin Brush, Dick Jansen, David Ceci, Kevin Carney, Don Dry, George Brooks, David Chamberlain, Margaret Goldberg.

Source term information is required for to reconstruct a device used in a dispersed radiological dispersal device. Simulating a radioactive environment to train and exercise sampling and sample characterization methods with suitable sample materials is a continued challenge. The Idaho National Laboratory has developed and permitted a Radioactive Response Training Range (RRTR), an 800 acre test range that is approved for open air dispersal of activated KBr, for training first responders in the entry and exit from radioactively contaminated areas, and testing protocols for environmental sampling and field characterization. Members from the Department of Defense, Law Enforcement, and the Department of Energy participated in the first contamination exercise that was conducted at the RRTR in the July 2011. The range was contaminated using a short lived radioactive Br-82 isotope (activated KBr). Soil samples contaminated with KBr (dispersed as a solution) and glass particles containing activated potassium bromide that emulated dispersed radioactive materials (such as ceramic-based sealed source materials) were collected to assess environmental sampling and characterization techniques. This presentation summarizes the performance of a radioactive materials surrogate for use as a training aide for nuclear forensics.

Log: 348. **CONTROLLING CO-60 IN THE MEASUREMENT OF NI-63**. S.Eriksson, A. Vesterlund, H. Ramebäck.

In methods for quantification of Ni-63, in e.g. reactor coolant water, a chemical separation is required due to Ni-63 being a pure beta emitter with limited means of quantification. Co-60 is a common interfering radionuclide, present in e.g. reactor coolant water, which is not completely separated using the commonly used separation procedure, and the two radionuclides are not resolved in the beta spectrum. The separation procedure used consists of TRU resin (Eichrom) and Ni resin (Eichrom). After running through the separation procedure, there may still remain enough interfering Co-60 to cause a large measurement uncertainty associated with the Ni-63 activity. The Co-60 interference is corrected for by using gamma spectrometry to determine the activity of Co-60 in the sample. This correction may, depending on the level of Co-60 in the sample, introduce a large contribution to the combined measurement uncertainty. In this work a second Ni separation was performed in order to decrease the radioactivity of Co-60 in the sample to be measured, and therefore reducing the combined measurement uncertainty in the Ni-63 measurement result.

Log: 349. **RADIONUCLIDES IN REDISTRIBUTED SEDIMENTS AND ASH FROM THE LAS CONCHAS FIRE IN NORTHERN NEW MEXICO**. Matteson, BS; Miller, JL; Oldham, WJ. Los Alamos National Laboratory.

In June of 2011, a tree fell onto a power line on residential property, igniting the 156,000 acre Las Conchas fire within Northern New Mexico's Jemez Mountains. In the weeks following the fire, heavy rainfall caused extensive flooding and erosion of surface sediments and ash from the affected areas. Samples from these mud and ash flows were collected from along a local highway that bisected the burned area and were analyzed for certain anthropogenic and natural radionuclides. These collections were analyzed for ^{137}Cs and Pu, which were originally deposited as global fallout during the era of atmospheric nuclear testing, and were compared to the concentration of natural U and ^{210}Pb isotopes. A striking and consistent correlation was found between elevated levels of Pu and unsupported ^{210}Pb . The latter isotope is continuously deposited onto the surface layers of the forest as a result of radioactive decay of the noble gas, ^{222}Rn . This observation suggests that prior to the fire, both these nuclides had been immobilized into the organic-rich surface of the forest floor. This study provides insight into the mobility,

redistribution, and concentration of anthropogenic radionuclides as a result of forest fires. Specifically, measurements of Pu in eroded sediments may ultimately provide a useful tracer of the organic-rich surface material that was lost during the wildfire event.

Log: 350. **HIGH SENSITIVE ANALYSIS OF AM AND PU ISOTOPES IN AEROSOL SAMPLES AFTER THE CHERNOBYL AND FUKUSHIMA ACCIDENTS.** Lujanienė, G (1); Aninkevičius, V (1); Povinec, PP (2). (1) SRI Center for Physical Sciences and Technology, Vilnius, Lithuania, (2) FMPI, Comenius University, Bratislava, Slovakia .

Analyses of airborne radioactive aerosols were carried out after the Chernobyl accident, during the post Chernobyl period, and after the Fukushima accident in daily samples in Vilnius (Lithuania) with special emphasis on better understanding of Am and Pu behaviors in the atmosphere. High activities of $^{239, 240}\text{Pu}$ and ^{241}Am found in the sample collected on 30 April 1986 were explained by the presence of “hot” particles in aerosol. The observed $^{238}\text{Pu}/^{239, 240}\text{Pu}$ activity ratio ranged from 0.44 to 0.50 while $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio varied from 0.41 to 0.42. During the post Chernobyl period the $^{239, 240}\text{Pu}$ and ^{241}Am activity concentrations in monthly samples ranged from 1 to 500 and from 0.3 to 500 nBq/m³, respectively. The exponential decrease in the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio from 0.30 to 0.19 observed in 1995-2003 was explained by decrease in the amount of the Chernobyl originated plutonium in the environment. In order to check the presence of Pu isotopes in samples collected after the Fukushima accident between 23 March and 15 April, 2011 (N=30, sampling air volume of ~ 2000000 m³) all filters were combined together to form one sample, and Pu isotopes were separated and measured by means of alpha-spectrometry. The activity concentration of $^{239, 240}\text{Pu}$ in this integrated sample was found to be 44.5 ± 2.5 nBq/m³ – very close to the background value. However, the activity of ^{238}Pu was higher than that of $^{239, 240}\text{Pu}$ (by a factor of 1.2) indicating a presence of the spent fuel of different origin than that of the Chernobyl accident.

Log: 351. **^{137}Cs , $^{239, 240}\text{Pu}$, ^{241}Am BEHAVIOR IN THE BALTIC SEA - CURONIAN LAGOON SYSTEM.** Lujanienė, G (1); Garnaga G (2); Remeikaitė-Nikienė, N (2); Jokšas, K (3); Garbaras, A (1); Skipitytė, R (1); Ščiglo, T (1); Barisevičiūtė, R (1); Valiulis, D (1); Šilobritienė, B (4). (1) SRI Center for Physical Sciences and Technology, Vilnius, Lithuania, (2) EPA, Marine Research Department, Klaipėda, Lithuania, (3) SRI Nature Research Centre, Vilnius, Lithuania, (4) EPA, Vilnius, Lithuania.

Redistribution of ^{137}Cs , $^{239, 240}\text{Pu}$, ^{241}Am activity concentrations in the coastal zone of the Curonian Lagoon and Baltic Sea was studied. Water, suspended particle, bottom sediment and soil samples were collected during different sampling campaigns in 1997–2011. Characterization of samples with different analytical techniques was performed. Total carbon and total organic carbon (TOC) were determined using a LECO CS-125 analyzer. Grain size distribution was measured by the gravimetric pipette method. Clay minerals were identified by X–ray diffraction (D8 (Bruker AXS) X-ray diffractometer). Humic substances were separated using conventional methods and characterized by FTIR and ^1H , ^{13}C NMR (Spectrum BX II (Perkin Elmer) FTIR and Varian Inova spectrometer). $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ were determined using the elemental analyzer (FlashEA 1112) connected to an isotope ratio mass spectrometer (Thermo Finnigan Delta Plus Advantage). ^{137}Cs activities were measured with an intrinsic germanium detector (resolution 1.9 keV/1.33 MeV and efficiency 42%). For measurement of Am and Pu activities the elements were first separated by the TOPO / cyclohexane extraction and then purified using UTEVA, TRU and TEVA resins (100–150 μm). $^{240}\text{Pu}/^{239}\text{Pu}$ ratio was determined using ICP-MS (ELEMENT-2). This study indicated that clay particles responsible for the redistribution of ^{137}Cs in the system while $^{239, 240}\text{Pu}$ and ^{241}Am activities in bottom sediments correlated well with the TOC (RPu=0.98 and RAm=0.83). The obtained results showed that traces of Pu and Am complexed by natural organic substances were washed out from the soil surface and transported by river flow to the Baltic Sea.

Log: 352. **FACTORS AFFECTING THE QUALITY OF PLUTONIUM DEPOSITS BY ELECTRODEPOSITION.** Bond, EM (1); Moody, WA (1); Dry, DE (1), Rabin, MW (1). (1) Los Alamos National Laboratory.

Ultra-high resolution alpha spectrometry by microcalorimetry has demonstrated a dramatic improvement in alpha energy resolution over silicon based detectors. To characterize the optimal resolution obtained by the microcalorimeter alpha spectrometers, high quality deposits that are virtually massless are required; electrodeposition is the preferred method for the preparation of high quality deposits. In order to better understand the factors that contribute to lower alpha energy resolution and deposit yield, we have conducted a study to determine the effect of some of the parameters that are used for preparing electrodeposits. We have compared four different electrodeposition methods and four different substrate materials to determine the effect on the deposit yield and alpha energy resolution of plutonium as measured by full width at half maximum (FWHM) using silicon based detectors. Furthermore, we wanted to understand the effect of sample contaminants from environmental samples on electrodeposits. Therefore, we have studied the effect on deposit yield and alpha energy resolution with several common soil constituents (Al^{3+} , K^+ , Na^+ , Mg^{2+} , Lu^{3+} , Fe^{3+} , Eu^{3+}). The deposit yield was affected in all cases by the addition of metal salts; Na^+ and K^+ had a lower impact on the deposit yield than Al^{3+} , Mg^{2+} , Lu^{3+} , Fe^{3+} , and Eu^{3+} . The alpha energy resolution of the deposits using silicon based detectors were reduced by the addition of lanthanide ions, and to a much lesser extent, by the addition of Mg^{2+} ions to the deposits. We have used these results to prepare better deposits and to design separation methods for microcalorimetry samples. LA-UR 11-06841

Log: 353. **DEVELOPMENT OF ALGORITHMS FOR FUSION OF NEUTRON AND X-RAY IMAGES USING MISSOURI S&T NEUTRON/X-RAY COMBINED CT FACILITY.** Sinha, V(1); Lee, HK (1); . (1) Missouri University of Science & Technology.

A novel imaging method combining traditional neutron and X-ray computed tomography has been developed at Missouri S&T. This new method holds much promise for non-destructive material detection and analysis where a range of z values and thermal cross sections may be present within an object. Where multiple materials having similar atomic number and differing thermal cross section or vice versa may be present, exclusive neutron or X-ray analysis may exhibit shortcomings in distinguishing interfaces. However, fusing neutron CT and X-ray CT offers the strengths of both and may provide a superior method of analysis. Presented in this paper is a first approach to developing an algorithm to fuse neutron and X-ray images. This new method will be compared to existing image fusion techniques such as linear superposition, nonlinear superposition, and wavelet transform-based methods. Moreover, specific material science applications will be presented and analyzed.

Log: 354. **DESIGN AND DEVELOPMENT OF MISSOURI S&T NEUTRON/X-RAY COMBINED COMPUTED TOMOGRAPHY SYSTEM.** Sinha, V (1); Lee, HK (1); . (1) Missouri University of Science & Technology.

Computed tomography (CT) using x-rays or neutrons can be used to visualize internal structures of an imaged object in 2D or 3D thus providing useful information for material science, biotechnology, homeland security, etc. As the characteristics of x-ray interactions are different from those of neutron interactions, information from x-ray CT images is different from that of neutron CT images. The new type of neutron and x-ray imaging facility has been designed and is operational at Missouri S&T research reactor. The experimental set-up has unique features to perform x-ray and neutron imaging simultaneously without obstructing the geometrical features for each imaging modality. The current system is optimized for small to medium sized object imaging of the magnitude order from 0.5 mm to 50 mm. The new facility opens a new horizon in the field of non-destructive testing specially for nuclear engineering, nuclear medical science, and material science research. In this paper, a novel type of combined x-ray and neutron imaging system will be introduced for superior analysis of certain imaging objects with examples of on-going research.

Log: 355. **DETERMINATION OF DEPTH OF A RADIONUCLIDE SOURCE IN A TISSUE EQUIVALENT PHANTOM.** H Aldousari¹, M Alkhorayef², DJ Highgate³, NM Spyrou^{1,2}
¹Department of Physics, University of Surrey, Guildford Surrey GU2 7XH UK ²Department of Radiological Sciences, College of Applied Medical Sciences, King Saud University, PO Box 10219 Riyadh 11433 Saudi Arabia. ³Department of Chemistry, University of Surrey, Guildford Surrey GU2 7XH UK.

Hydrophilic materials which refer to a group of cross-linked polymers originally developed in the sixties to produce soft contact lenses are characterised by an equilibrium water uptake in the range 15-95% by wet weight when hydrated in water or normal saline solution at 25°C. This ability to absorb controlled amounts of water makes them excellent human tissue substitutes. Four types of hydrophilic materials of different composition and thickness were placed between a Na²² point source and a single LaBr₃:Ce (5%) detector (Saint-Gobain) to measure the linear photon attenuation coefficient for each type separately and in combination. The depth of the source was determined by varying the width of the scattering window and calculating the ratio of the count in the full energy photopeak and varying the width of the scattering window below it. From the gamma ray spectra recorded a 'best' estimate of the depth of the source in the hydrophilic material was obtained. Theoretical models of the source-to-detector geometries are included and discussed.

Log: 356. **ACTINIDE SEPARATIONS USING IONIC EXCHANGE AND EXTRACTION CHROMATOGRAPHY RESINS.** Julie Champion,(a,b) Jennifer Shusterman,(a) Erin Gantz,(a) Eva Uribe, (a) Paul Ellison, (a) and Heino Nitsche(a). a: Department of Chemistry, University of California, Berkeley, Berkeley, CA 94720. b :Laboratoire SUBATECH, UMR CNRS 6457 IN2P3 Ecole des Mines de Nantes, 4 rue A. Kastler, 44307 Nantes Cedex, France.

The development of fast, effective procedures for the separation of each of the actinides from one another and fission products is necessary for advancements in actinide chemistry. Our group has investigated three procedures using ion exchange and extraction chromatography resins. The first procedure is for the isolation of americium from a large excess of plutonium and fission products. Steps of this separation use Dowex 1x8 anion exchange resin and others use the TEVA extraction chromatography resin. We determined that separation factors of greater than 106 could be obtained. The second separation is for the isolation of uranium from neptunium and fission products using UTEVA extraction chromatography resin. The last procedure explored uses the DGA extraction chromatography resin for trivalent actinide and lanthanide separations. We are working towards understanding the fundamental interactions of these resins with actinides and fission products to improve separation procedures to maximize separation factors.

Log: 357. **A NEW ROUTE OF PRODUCTION OF POLONIUM-210.** Ali Younes,(a) Cyrille Alliot,(b) Marcel Mokili, (a,b) Gilles Montavon,(a) and Julie Champion (a).(a) SUBATECH laboratory (UMR 6457), France, Nantes 44307.(b) ARRONAX laboratory, France, Nantes 44817.

Our work is focusing on the production of polonium-210 source which could be used for fundamental research of polonium chemistry. Po-210 is obtained after the decay of At-210 (8.1 h) produced by the nuclear reaction Bi-209 (α , 2n)At-210. A Bi-209 foil was irradiated for two hours by a 40 MeV alpha external beam delivered by the ARRONAX cyclotron in Nantes with an average beam current of 0.1 μ A . First tests were done with Po-210 activities ranging from 1 to 10 KBq. After 1 week (about 21 periods of At-210) , Po-210 and the byproducts (Bi-207 and Bi-206) were dissolved in 10 M nitric acid. The solution was then evaporated to dryness and the residue was dissolved in 7 M HCl. Po-210 was extracted in 10 % TBP/p-xylene mixture and finally selectively back-extracted in the presence of 9 M HCl. The recovery yield amounted to 90%.

Log: 358. **DECONTAMINATION OF RADIONUCLIDES ON CONSTRUCTION MATERIALS.** Samuleev, PV (1); Andrews, WS (1); Creber, KAM (1); Azmi, P (2); Velicogna, D (3); Kuang, W

(2); and Volchek, K (2). (1) Royal Military College of Canada, (2) Environment Canada, (3) SAIC Canada.

A wide variety of materials can become contaminated by radionuclides, either from a terrorist attack or an industrial or nuclear accident. The final disposition of these materials depends, in large part, on the effectiveness of decontamination measures. This study reports on investigations into the decontamination of a variety of building materials. The aim has been to find an effective, easy-to-use and inexpensive decontamination system for radionuclides of cesium and cobalt, considering both the chemical and physical nature of these potential contaminants. The basic method investigated was surface washing, due to its ease and simplicity. In the present study, a basic decontamination formulation was modified by adding isotope-specific sequestering agents, to enhance the removal of cesium(I) and cobalt(II) from such construction materials as concrete, marble, aluminum and painted steel. Spiking solutions contained Cs-134 or Co-60, which were prepared by neutron activation in the SLOWPOKE-2 nuclear reactor facility at the Royal Military College of Canada. Gamma spectroscopy was used to determine the decontamination efficiency. The results showed that the addition of sequestering agents generally improved the radiological decontamination. Although the washing of both cesium and cobalt from non-porous materials, such as glass and painted steel, achieved a 90-95% removal, the decontamination of concrete and marble was more challenging, due to the porous nature of the materials. Nevertheless, the removal efficiency from six-year-old concrete increased from 1 % to approximately 15 % for cobalt (II), and from 15 % to 35 % for cesium (I), with the use of isotope-binding agents, as opposed to a simple water wash.

Log: 359. **NON-VOLATILE ORGANIC ANALYSIS OF URANIUM ORE CONCENTRATES.**

Angel Wall (1), Debra Bostick (1), Cole Hexel (1), Rob Smith (1), Joe Giaquinto (1). (1) Oak Ridge National Laboratory.

In support of nuclear safeguards and non-proliferation efforts, Oak Ridge National Laboratory is responsible for characterizing uranium ore concentrate (UOC) samples obtained from two ore mining and milling sites. A sorptive extraction method has been developed for analysis of non-volatile organic compounds that might be used to identify characteristics of the purification process by which uranium was separated from these ores. This method utilizes Gerstel Twister™ stir bars coated with polydimethylsiloxane to extract organic components from aqueous media. A slurry of UOC is extracted with the Twister™ stir bar in 20% methanol/80% water containing deuterated internal hydrocarbon standards. Following extraction of non-volatile organics, the Twister™ stir bar is analyzed directly in the inlet of a gas chromatograph fitted with a quadrupole mass spectrometric detector. Results have been consistent and have shown excellent recoveries of internal standards, with the average recovery being 97.5%. Both qualitative and quantitative differences have been identified between the two sources of UOC utilizing this method. One source contained an increased concentration of amines which commonly are used in the recovery and purification of ores. Amines that were identified in this UOC source include dioctylamine, triisooctylamine, and Alamine® 336, a common industrial complexant. Also, when comparing both sources, the same UOC source contained various decanol and C20 compounds. Based on the results from this study, non-volatile organic analysis of UOC using sorptive extraction with Twister™ stir bars and GC-MS is a tool that can be used to facilitate sourcing unknown UOC.

Log: 360. **DIGITAL PULSE-SHAPE DISCRIMINATION APPLIED TO AN ULTRA-LOW-BACKGROUND GAS-PROPORTIONAL COUNTING SYSTEM - FIRST RESULTS.** AUTHORS: C.E. Aalseth, A.R. Day, E.S. Fuller, E.W. Hoppe, M.E. Keillor, E.K. Mace, C.T. Overman, M.E. Panisko, A. Seifert, G.A. Warren, R.M. Williams. ORGANIZATION: Pacific Northwest National Laboratory, Richland, WA, USA.

A new ultra-low-background proportional counter (ULBPC) design was recently developed at Pacific Northwest National Laboratory. This design, along with an ultra-low-background counting system (ULBCS) which provides passive and active shielding with radon exclusion, has been

developed to complement a new shallow underground laboratory (~30 meters water-equivalent) constructed at PNNL. After these steps to mitigate dominant backgrounds (cosmic rays, external gamma-rays, radioactivity in materials), remaining background events do not exclusively arise from ionization of the proportional counter gas. Digital pulse-shape discrimination (PSD) is thus employed to further improve measurement sensitivity. In this work, a template shape is generated for each individual sample measurement of interest, a "self-calibrating" template. Differences in event topology can also cause differences in pulse shape. In this work, the temporal region analyzed for each event is refined to maximize background discrimination while avoiding unwanted sensitivity to event topology. This digital PSD method is applied to sample and background data, and initial measurement results are presented in the context of low-background measurements currently being developed. These include tritium, carbon-14, argon-37, and argon-39. Applications such as nuclear treaty verification, elucidating the environmental carbon cycle, and the assay of low-background materials for next-generation nuclear physics experiments are discussed.

Log: 361. **MEASUREMENT OF LANTHANIDE ELEMENTS IN URANIUM MATRICES USING HPLC-ICPMS TECHNIQUES AND ISOTOPICALLY ENRICHED MIXED LANTHANIDE STANDARDS.** Giaquinto, J.M.; Aaron, W.S.; Ilgner, R.H.; Keever, T.J. Oak Ridge National Laboratory, Managed by UT-Battelle for the Department of Energy under Contract DE-AC05-00OR22725.

The measurement of the lanthanide elements in uranium matrices is a key component in the characterization of nuclear materials for numerous national and international organizations tasked with the management, tracking, and safeguarding of uranium used in all phases of the fuel cycle. To improve the analytical accuracy and precision of nuclear mass spectrometry measurements for this key elemental series, certified spiking mixtures of enriched lanthanides elements are needed. Some examples include: 1) Fission produced rare earth elements in irradiated fuels are key parameters for the evaluation of fuel performance and calculation of percent fissions. 2) The management of nuclear waste is enhanced with an accurate accounting of key lanthanide isotopes affecting burn-up credit for storage and disposal. 3) Geological variations of rare earth elements in uranium deposits provide forensic signatures for non-proliferation and safeguarding of worldwide stockpiles. Oak Ridge National Laboratory's Chemical Sciences Division (CSD) routinely performs measurement of the lanthanide series of elements using mass spectrometry in a variety of uranium matrices. To improve the analytical results for the most recent MOX spent fuel PIE campaign, "in-house" enriched spiking mixtures were created using stock materials obtained through ORNL's Isotope Business Office (IBO). Presented will be the success CSD has had using these enriched mixtures combined with HPLC-ICPMS and IDMS techniques. There will also be discussions concerning standards that are available from the IBO for use as starting materials to produce series of certified enriched lanthanide mixtures for use by the nuclear community.

Log: 362. **NEW CONCEPTS FOR RADIOMETRIC MEASUREMENTS OF ENVIRONMENTAL SAMPLES CONTAINING FISSION PRODUCTS AND ACTINIDES.** Glen A. Warren, Robert C. Runkle, Pacific Northwest National Laboratory, Richland, WA, USA.

The Radiation Detection and Nuclear Sciences Group at Pacific Northwest National Laboratory has a long history in conducting measurements of radioisotopes for various applications. This experience includes ultra-low background measurements, arrays of germanium detectors, automated sampling and measurement systems and coincidence measurement systems. A recent lab-supported effort has been studying how these capabilities, both in terms of hardware and experience, can be leveraged to enable environmental sampling measurements. One area of interest is the release of fission products and actinides into the environment from a reactor incident. While the initial survey of this area is still under way, one isotope of interest that surfaced early in the study is Pu-238. Existing techniques to assay this isotope suffer from measurement challenges. In alpha counting, there can be significant interference with Am-241, while in mass spectrometry, there can be interference with U-238. We are developing the

concept for a detector that through coincidence counting techniques can distinguish Pu-238 and Am-241. In addition, we will design the system to conduct radiometric measurements of other plutonium isotopes to enable a direct comparison of those isotopes. We will present our concept of the detector system for Pu-238, as well as discuss other radiometric measurements of fission products and actinides with which we intend to advance the state of the art for environmental measurements.

Log: 363. HISTORICAL AND CLIMATOLOGICAL RESEARCH IN THE HIMALAYA REGION BY 14C AMS DATING OF WOODEN DRILL CORES FROM HISTORIC BUILDINGS.

Kretschmer, W (1), Bräuning, A (2), Scharf, A (1), Leichmann, K (1), Wegner, B (1), Daragon, F (2). (1) Physics Institute, University of Erlangen, Germany, (2) Geographical Institute, University of Erlangen, Germany.

In recent years, the Geographical Institute of the University Erlangen could sample numerous wooden drill cores from historic buildings in four regions of High Asia and could evaluate them dendrochronologically. Part of the drill cores were collected from monasteries and temples in the Dolpo region of western Nepal, a barely studied region in the Inner Himalaya and situated in the rain shadow of the main Himalayan crest line. Another major part came from temples in Central Tibet. In many cases tree-ring dating of these drill cores was not possible, indicating that the sample woods exhibit a higher age than the present range of the existing tree-ring chronologies which only reach back to the 11th century. So these samples can be used to extend the tree-ring chronologies of this region, which could help to detect suggested monsoon variations during the Middle Ages. The historic tower buildings of Tibet and Sichuan are a special cultural heritage which has been rarely studied up to now. The knowledge of their exact age could help to better understand the cultural and historical context of their development and their function, and could support the effort to declare them a UNESCO World Heritage site. The Erlangen AMS laboratory has performed 14C - AMS datings on 200 samples of 74 of these drill cores. Using wiggle-matching these drill cores could be dated with enhanced precision, and in many cases important information about the time of construction of these important historic buildings could be obtained for the first time.

Log: 365. CHARACTERIZATION OF DGA RESIN FOR THE ANALYSIS OF FUEL RECYCLING PROCESS STREAMS. Audrey Roman, Elaine Go, Ralf Sudowe, University of Nevada, Las Vegas.

The analysis of the different process streams present in a reprocessing facility is essential to obtain material balance and accountancy. Current techniques for the analysis of grab samples are time and labor intensive and not well suited for situations in which a rapid sample analysis is required and/or a large number of samples need to be analyzed. To address this issue the utilization of rapid radiochemical separations based on commercial extraction chromatographic resins is being investigated. Use of DGA resin appears promising for the analysis of dissolved fuel and raffinate streams found in a reprocessing cycle. Even though it is known that DGA resin has a high affinity for Am, Cm, and Pu, the dissolved fuel and raffinate will have a very different composition and in particular a much higher mass loading than is found in environmental samples. To further elucidate the usefulness of DGA resin for the analysis of process streams, the interference and synergistic effects on Am, Cm, and Pu sorption caused by components likely to be present in different reprocessing streams is being investigated.

Log: 366. MEASUREMENT OF SR-90 AND STRONTIUM ISOTOPIC RATIO IN FUKUSHIMA SOIL AFTER THE NUCLEAR ACCIDENT. Fukutani, S(1); Fujii, T(1); Ueda, Y(2); Koyanaka, H(3); Tokuda, Y(4); Kubota, T(1); Koyama, A(1). (1)Research Reactor Institute, Kyoto University, (2)Research Institute for Sustainable Humanosphere, Kyoto University, (3)Institute for Integrated Cell-Material Sciences, Kyoto University, (4)Institute for Chemical Research, Kyoto University.

Due to the nuclear disaster of Fukushima dai-ichi Power Plant, many kinds of radionuclides including cesium, strontium and iodine were released and detected in land areas. Toward

radiation protection, radioactive decontamination, environmental restoration and so on, many measurements and evaluations about fallout radionuclides have been conducted, and many works still remain. We focused on measurement of Sr-90 in soils at Fukushima prefecture. Soil was collected after the accident, and we could obtain soil collected before the accident. Soils were nitrated and nuclides including strontium were allowed to leaching. Strontium was separated using Sr-Resin™ (Eichrom Industries), and Sr-90 (Y-90) was measured. And using the leachate with Sr-Resin™, we also measured isotopic ratio of strontium by Multi-Collector Thermal Ionization Mass Spectrometer (MC-TIMS), and we intend to discuss variation of strontium isotopic ratio before and after the nuclear disaster and its meaning.

Log: 367. **ANALYSIS OF CL-36 IN SRS HIGH ACTIVITY WASTE MATRICES.** White, T. L. (1); DiPrete, D. P. (1); Dobos, G. J. (1); DiPrete, C. C. (1). (1) Savannah River National Laboratory.

Radioactive waste cleanup and closure of waste storage tanks is currently underway at the Savannah River Site, prompting the need to characterize the residual contents of the tanks. Each tank heel has unique chemical constituents as well as unique radiological distributions compared to previously characterized heels. In addition to the challenges posed by variable matrix components and variable relative inventory distributions, target detection limits are often challenging to meet. Continuous programmatic evaluations of allowable closed tank radiological inventories result in steadily decreasing detection limit targets for isotopes required for characterization. Some target isotopes required for characterization need to be quantified to levels approaching 10 orders of magnitude below radioactive concentrations of interfering isotopes. One practically troublesome target analyte is ^{36}Cl . As a pure beta emitter, ^{36}Cl ($T_{1/2} = 3.01 \times 10^5 \text{ yr}$) must be isolated from interfering beta-emitting radionuclides such as ^{90}Sr , ^{90}Y , ^{137}Cs , ^{99}Tc , ^{14}C , as well as actinides and lanthanides prior to quantification. Due to its half-life, the target detection limit for ^{36}Cl is extremely low compared to the concentrations of other beta-emitting radionuclides via repeated resin-strikes, distillation, precipitation as AgCl , and analysis by gas flow proportional counting for the ^{36}Cl -containing precipitate. The separation is traced by neutron activation analysis of stable chloride imparted during the initial digestion. Recent campaigns have successfully achieved interference-free AgCl precipitates and, ultimately, detection limits of 22 dpm/g from matrices containing $\sim 1\text{E}^{10}$ dpm/g of other beta-emitting nuclides.

Log: 368. **THE SEPARATION OF AM-240 FROM PU-242 AND FISSION PRODUCTS.** Erin M. Gantz, Julie Champion, Paul Ellison, and Heino Nitsche University of California, Berkeley.

Recent work has yielded both a reaction to produce Am-240 and a comprehensive separation of Am-240 from Pu-242 and its fission products, allowing for the separation to generate a pure sample of Am-240 to measure the Am-240 neutron-induced fission cross section. This measurement is relevant to stockpile stewardship and has not been previously undertaken due to the 50.8 hour half life of Am-240. Because the production reaction and the separation have already been developed, current work is focused on how the fission products that are also generated during the proton-irradiation of Pu-242 will behave in the existing separation procedure. Longer-lived isotopes of Sb, Zr, Nb, and Eu have been used to model the fission products through a series of four separation columns: two anion exchange columns (DOWEX-1) and two extraction chromatography columns (TEVA). In addition, the separation procedure has been tested using hydrochloric acid and nitric acid as loading and rinsing reagents to reconcile elution differences, particularly with the behavior of Sb and Zr, discovered in previous experiments. Hydrochloric acid functioned as a more effective loading and rinsing reagent when separating the fission products from Am-240 and current research explores if hydrochloric acid will be compatible with the overall requirements of this work. Experiments are being undertaken to better quantify the degree of separation of Am-240 from Pu-242 and the fission products as well as to scale this procedure to simulate nuclide quantities encountered when preparing the actual Am-240 sample for a neutron-induced fission cross section measurement.

Log: 369. **A TECHNOLOGICAL OVERVIEW OF THE AERIAL MEASUREMENT SYSTEM RESPONSE TO THE FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT ACCIDENT.**

Alexander A. Plionis, Wendy Pemberton, Michael Reed, Lance McLean, and William Beal;
National Security Technologies Remote Sensing Laboratory.

In response to the Fukushima Dai-ichi nuclear power plant accident the U.S. National Nuclear Security Administration deployed a number of emergency response teams including the Aerial Measurement System (AMS). The AMS is comprised of an integrated array of NaI detectors that is configured to operate within an aerial platform. AMS scientists and technicians partnered with the U.S. Air Force to conduct broad area contamination surveys within 80 km of the plant to characterize ground level exposure rates for consequence management. An overview of tools and techniques for measurement and assessment is provided.

Log: 371. **FAST CONCENTRATION OF DISSOLVED FORMS OF CESIUM RADIOISOTOPES FROM LARGE SEAWATER SAMPLES.** Kameník, J (1); Dulaiova, H (1); Šebesta, F (2);

Šťastná, K (1,2). (1) School of Ocean and Earth Science and Technology, University of Hawaii at Manoa (2) Centre for Radiochemistry and Radiation Chemistry, Czech Technical University, Prague, Czech Republic.

Recent accidental and managed releases of radioactivity to the ocean from the Fukushima nuclear reactors prompted us to revisit procedures of cesium analysis in seawater. We tested and compared two methods that were developed for cesium analysis in freshwater samples and were adapted here for large volume seawater analysis. Concentration of dissolved forms of cesium in large seawater samples (≥ 100 L) was performed using composite absorbers AMP-PAN and KNiFC-PAN. These materials contained ammonium molybdophosphate (AMP) and potassium-nickel hexacyanoferrate(II) (KNiFC) as active components, respectively, and polyacrylonitrile (PAN) as a binding polymer. A specially designed chromatography column allowed fast flow rates (about 300 mL/min) through a bed of 25 mL of absorber. The flow-rate was regulated to pass a 100-L sample in about 4 to 6 hours. After cesium extraction the composite absorber was counted using gamma-spectrometry for determination of Cs-134 and Cs-137 activity in seawater in the central Pacific Ocean. The chemical recovery of cesium was monitored using a stable cesium tracer, which was analyzed using ICP-MS. Both absorbers provided recoveries high enough for low-level cesium analysis in seawater.

Log: 372. **AGE DATING OF MIXED SNM—PRELIMINARY INVESTIGATIONS.** Yuan D (1); Guss, PP (2); Yfantis, E (3); Klingensmith, A (4); Emer, D (4). (1) NSTec LAO, (2) NSTec Remote Sensing Laboratory, (3) University of Nevada, Las Vegas, Comp. Sci., (4) NSTec National Center for Nuclear Security.

Recently we investigated the nuclear forensics problem of age determination for mixed special nuclear material (SNM). Through limited computational mixing experiments and interactive age analysis, it was observed that age dating results are generally affected by the mixing of samples with different assays or even by small radioactive material contamination. The mixing and contamination can be detected through interactive age analysis, a function provided by the Decay Interaction, Visualization and Analysis (DIVA) software developed by NSTec. It is observed that for mixed SNM with two components, the age estimators typically fall into two distinct clusters on the time axis. This suggests that averaging or other simple statistical methods may not always be suitable for age dating SNM mixtures. Instead, an interactive age analysis would be more suitable for age determination of material components of such SNM mixtures. This work was supported by the National Center for Nuclear Security (NCNS).

Log: 373. **INTERPRETING ELEMENTAL AND ACTIVITY DISTRIBUTIONS WITHIN FALLOUT DEBRIS FROM HISTORICAL NUCLEAR WEAPONS TESTS.** K. Knight, R. Gostic, A. Lindquist, R. Ryerson, I. Hutcheon.

A series of soils from near-surface historical US nuclear weapons tests were collected for study. These soils contain a mixture of environmental materials, partially melted environmental materials, and anthropogenic glasses clearly formed in conjunction with the tests. While all components of the soils contain enhanced activity relative to environmental backgrounds, the wholly melted anthropogenic glasses significantly concentrate this activity. Activity measurements demonstrate that these materials retain remnant fuel from the test, as well as related fission products and activation products. While the average major element chemistry of these glasses is uniform and reflects the major element chemistry of the starting environmental materials, several scales of heterogeneities exist. The relative alpha, beta and gamma activities can vary dramatically between individual samples. In addition, within a single piece of anthropogenic glass, strong differences in activity distributions are sometimes observed, as well as variations in major and trace element concentrations. We present elemental mapping in combination with activity measurements and autoradiography to characterize and interpret heterogeneities in the distribution of elements within anthropogenic fallout glass. These data support the existence of multiple formation mechanisms for fallout debris and can be used to aid our understanding of the physical and chemical conditions governing fallout debris genesis, and thus contribute to its interpretation. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 374. **AIR MONITORING IN LAS VEGAS IN THE AFTERMATH OF THE REACTOR ACCIDENT AT FUKUSHIMA DAIICHI.** Sudowe, R; Turner, MF; Bene, BJ; Bene, KE; Despotopulos, JD; Faye, SA; Gharibyan N. University of Nevada, Las Vegas.

The strong earthquake and the following tsunami that hit Japan on March 11, 2011 resulted in a complete loss of electrical power as well as all cooling capabilities to reactors at Fukushima Daiichi. During the attempts to stabilize the reactors, radioactive fission products were released into the environment. It was expected that traces of the gaseous and volatile radionuclides released into the atmosphere would spread across the Pacific and eventually reach the U.S. In the aftermath of the accident, a group of students and researchers from the University of Nevada, Las Vegas collected air samples using a high-volume air sampler to measure radionuclides released from the Fukushima Daiichi accident in Japan as a training exercise for the students and to provide information to the general public. Grab sampler equipped with glass fiber filters and cartridges containing activated charcoal were used to sample the air from the roof of the Bigelow Health Sciences building in Las Vegas. Air samples were typically collected for a period of 24 hours and then measured for 24 hours using a HPGe detector. Detailed results from the gamma spectrometric analysis of the filter samples will be presented.

Log: 375. **PRODUCTION OF DOUBLE-SIDED TARGETS BY ELECTRODEPOSITION: INITIAL EVALUATION AND OPTIMIZATION OF PERFORMANCE LA-UR 11-06848.** W. Allen Moody, Evelyn M. Bond, Todd A. Bredeweg.

The detector for Advance Neutron Capture Experiments (DANCE) located at the Los Alamos Neutron Science Center (LANSCE) is used to perform neutron capture cross section measurements on radioactive and non-radioactive isotopes. Thin actinide targets for the DANCE detector are typically prepared by molecular deposition on thin titanium foils. For the preparation of double-sided deposits, a Teflon electroplating cell was constructed with two liquid chambers with a foil substrate in between, allowing electrodeposition on both sides of the foil. We have been studying the electrodeposition of uranium from isopropyl alcohol solutions using this cell. Effects of acid composition, uranium concentration, current, and deposition time will be described.

Log: 376. **A FAST, ACTIVE, NON-DESTRUCTIVE INTERROGATION TECHNIQUE FOR PRE/POST-DETONATION NUCLEAR FORENSICS USING SHORT-LIVED DELAYED GAMMA-RAY SIGNATURES FROM FISSILE AND FISSIONABLE MATERIALS.** Dr. Michael Hartman, University of Michigan; Bruce D. Pierson, University of Michigan.

Short-lived fission products generated from interrogation of high-purity thorium-oxide using the University of Michigan accelerator-based neutron generator were detected using a high-purity germanium detector. Rather than collecting the data as a discrete spectrum, each interaction in the detector was recorded with a unique time stamp accurate to 200 ns. The time-dependent spectra were used to focus the analysis more effectively on short-lived isotopes. Numerous short-lived fission products, including zirconium-99 (Zr-99) and arsenic-85 (As-85), both with half-lives near 2 seconds, were detected and quantified for comparison to natural uranium. The independent and cumulative fission yields of short-lived fission products are unique to isotopes of different actinides. Zr-99 and As-85 exhibit large differences in their accepted cumulative and independent fission yields between thorium-232 and uranium-238. Differences in fission yields of short-lived fission products provide unique information about unknown samples for expedient identification of nuclear material. Corrected peak ratios between gamma-ray lines of isotopes with large differences in fission yields generated during fission of thorium-232 provide a unique fingerprint for comparisons to different actinides and actinide mixtures. Data generated from these experiments also provides information about fission product half-lives along with independent and cumulative fission yields. Planned irradiations of natural uranium-oxide, "yellowcake," will be conducted and quantitatively compared to the thorium-oxide peak ratios to more thoroughly illustrate this method.

Log: 377. **EXTRACTION TRENDS AND EFFECTS OF SYNERGY FOR LANTHANIDES AND ACTINIDES IN COMBINING NEUTRAL AND ACIDIC PHOSPHOROUS CONTAINING REAGENTS.** A. Braatz(1), R. Ellis(2), M. Antonio(2), M. Nilsson(1). 1. Department of Chemical Engineering and Materials Science, University of California Irvine, Irvine, CA. 2. Chemical Sciences and Engineering Division, Argonne National Laboratory, Chicago, IL..

To improve efficiency of solvent extraction processes for various applications it is often useful to explore combinations of reagents that offer some synergy of extraction of metal ions. The synergistic extraction of metal cations related to used nuclear fuel has been observed in many different systems over the last few decades when combining acidic and neutral reagents. For example the neutral solvating reagent tributyl phosphate (TBP) in combination with an acidic reagent such as dibutyl phosphoric acid (DBP) has shown synergy for a range of cations including uranyl. This is relevant during treatment of spent nuclear fuel by the PUREX (Plutonium URanium EXtraction) process since TBP may degrade to DBP and cause disruptions in the process. The nature of the synergy and these disruptions has been studied previously but mostly based on extraction equilibrium data which may not give the complete picture. In this study we have investigated what effect combining TBP and DBP has on the extraction of trivalent actinides and lanthanides as well as water and acid. We have combined these studies with small angle X-ray scattering (SAXS) to study possible structural organization in the organic phase. Our results show that mixed TBP+DBP extractants affect not only the distribution ratios of metal ions but also effect water uptake and display tendencies for mixed aggregate formation in the organic phase.

Log: 378. **MORPHOLOGICAL COMPARISONS OF ACTINIDE-RICH CONSTITUENTS RESULTING FROM THE THULE, PALOMARES, AND BOMARC ACCIDENTS.** Bowen, JM; Glover, SE; Spitz, HB. Nuclear & Radiological Engineering Program, School of Dynamic Systems, University of Cincinnati.

The provenance of material released during a broken arrow incident is dependent upon a mixture of conditions such as the makeup of the original material and the circumstances surrounding its release. The material has the possibility for additional influence due to weathering if allowed to remain in the environment. This study compares the morphological characteristics of particles created during the 1960 BOMARC incident to previously published results from two other broken arrow incidents: Thule, Greenland (1968) and Palomares, Spain (1966). Each incident involves different circumstances that led to the creation of these actinide-rich constituents as well as different environmental conditions. After examining the particles by gamma spectroscopy, morphology and qualitative surface elemental analyses were performed by electron microscopy and complimentary energy dispersive X-ray spectroscopy. Unique characteristics observed among some of the BOMARC samples include portions with a smooth, almost crystalline texture as well as heterogeneously distributed uranium and plutonium on the surface of the particle.

Log: 379. **THE USE OF HPLC-ICPMS TECHNIQUES FOR THE RAPID SEPARATION AND DETAILED CHARACTERIZATIONS OF LANTHANIDES AND ACTINIDES IN NUCLEAR MATERIALS.** Keever, T.J.; Giaquinto, J.M.; Ilgner, R.H. Oak Ridge National Laboratory, Managed by UT-Battelle for the Department of Energy under Contract DE-AC05-00OR22725..

Nuclear forensics relies on many factors, including the determination of physical, isotopic and chemical analysis of radioactive materials, as well as characterization of ultra-trace quantities of materials, including impurities. The ability to determine the isotopic composition of nuclear materials with high precision and accuracy is necessary in order to distinguish a particular sample from other nuclear or radiological materials. A comprehensive analysis of such materials would include the determination of the lanthanides and actinides elements. The quantification of the radioactive isotopes for these elements can be achieved using radiochemistry counting techniques, but these techniques are burdened by time-consuming matrix separations, sample preparations and large total uncertainties. However, precise determinations of isotopic compositions using ICPMS techniques are advancing as detection capabilities are improving and rapid chemical methods for separations of isobaric interferences are being recognized. Presented will be the application of HPLC separation methods combined with isotope dilution mass spectrometry toward highly precise and accurate determinations of lanthanide and actinide isotopic ratios in irradiated nuclear fuels, and bulk uranium and plutonium materials.

Log: 380. **PERFORMANCE AND UTILIZATION OF THE POWDER NEUTRON DIFFRACTOMETER AT THE PULSTAR REACTOR.** Qinsheng Cai, Ayman I. Hawari, Nuclear Reactor Program, North Carolina State University, Raleigh, NC, 27695, USA.

The PLUSTAR is an open pool research reactor with a maximum power of 1-MWth. The powder neutron diffractometer is located at beam tube 4. The thermal neutron flux at the entrance to the beam tube is approximately 3×10^{12} n/cm²·s. A filtered collimation system is used to guide the incoming neutrons to a double focusing monochromator consisting of bent single crystal silicon blades. Using this arrangement, a monochromatic neutron beam with a wavelength of 1.478 Å is directed at the sample with a flux of approximately 0.5×10^5 n/cm²·s. The associated neutron detection system is based on a position sensitive detector array spanning 20° and is attached to a moveable cassette that can cover a scattering angle range of 5°-125°. The resolution (Dd/d) of the diffractometer is estimated to vary in the range of 0.1% to 0.3%. In order to limit background, a secondary collimator was placed in front of the monochromator to constrain the original 8-inch diameter beam into 4x5.25 inches, a beam stop was placed around the monochromator, a 0.75 inch thick B4C layer was added inside the detector shielding, and a radial oscillating collimator was mounted in front of the detector shielding. Currently, the diffractometer is used to study neutron radiation damage in nuclear graphite. Measurements on unirradiated and irradiated

samples were performed. The results indicate that the radiation damage can increase the probability of stacking faults, the d-spacing, and the fluctuation of the d-spacing. Alternatively, it will decrease the probabilities of AB and ABC stacking, the stacking number of the layers, the in-plane lattice parameter, and the lateral dimension of the plane.

Log: 381. **RAPID QUANTIFICATION OF TBP AND TBP DEGRADATION PRODUCT RATIOS BY FTIR-ATR.** Gillens, AR (1); Powell, BA (1). (1) Environmental Engineering and Earth Sciences, Clemson University, Clemson, SC.

Tributyl phosphate (TBP) is an organic solvent used to extract uranium and plutonium from used nuclear fuel. During nuclear reprocessing TBP degrades to dibutyl phosphate (DBP), monobutyl phosphate, butanol, and phosphoric acid. A Fourier Transform Infrared Spectrometry-Attenuated Total Reflectance (FTIR-ATR) technique was developed to determine approximate ratios of TBP and degradation products in an actual reprocessed TBP sample. The method was developed by combining variable concentrations of TBP and DBP to simulate degradation of TBP. This method is achieved by analyzing selected peak positions and intensity ratios of TBP and DBP at different stages of degradation. The results are validated using Gas Chromatography Mass Spectrometry (GC-MS). Additionally, degradation of TBP in acidic and basic solutions was monitored using GC-MS as well as the FTIR-ATR technique to determine degradation mechanisms of TBP. In mock degradation samples, the 1235 cm^{-1} peak position shifts to 1220 cm^{-1} as the concentration of TBP decreases and DBP increases. Peak intensity ratios of TBP positions at 1279 cm^{-1} and 1020 cm^{-1} relative to DBP positions at 1003 cm^{-1} and 909 cm^{-1} demonstrate an increasing trend as the concentration of TBP increases. The method developed from this study may be used as a tool to determine TBP use in nuclear reprocessing via a rapid FTIR-ATR measurement without GC-MS analysis. Understanding how TBP degrades when in contact with acidic and basic solvents will provide information on how the signature of TBP changes during nuclear reprocessing.

Log: 382. **RADIOACTIVE SOURCE FORENSICS - WHAT CAN WE MEASURE? .** Chamberlain, DB (1); Steeb, JL (1); Carney, KP (2); Finck, M (2); Brooks, G (3); Goldberg, MM (4). (1) Argonne National Laboratory, (2) Idaho National Laboratory, (3) Los Alamos National Laboratory, (4) Federal Bureau of Investigation.

Forensics methods are being applied to determine the origin of abandoned, interdicted, or dispersed radioactive materials. In addition to the traditional forensics examinations (e.g. fingerprints, fibers, tool marks) and evaluation of the capsule labels or engravings, the capsules are examined for their dimensions; use of single, double, or triple encapsulation; and composition. The analysis of the radioactive material within a capsule can also provide important clues to its origin. The major radioactive isotope is of course important, but likely just as important are the decay products or daughters, trace impurities, and the form of the material (e.g. metal, chloride, ceramic). Parent-daughter ratios provide data for determining the age of a radioactive material or the time since it was last separated from its decay daughters. Isotopic abundances and ratios are typically determined by several instrumental methods, including alpha, beta, gamma spectroscopy and inductively coupled plasma and thermal ionization mass spectrometry. Information obtained from all of these measurements can then be compared to information contained in signature libraries to help deduce the material's origins and history for nuclear forensics investigations. Signatures and issues related to analysis of these signatures are presented.

Log: 383. **LINUX-BASED AUTOMATION FOR SHORT LIFETIME NUCLEAR MEASUREMENTS.** Suarez, R (1); Hubbard, CW (1); Hayes, JC (1); Heimbigner, TR (1); Miley, HS (1); Mendez, JM (1). (1) Pacific Northwest National Laboratory.

Human operation of repetitive and time sensitive chemical processing needed for short half-lives and enduring monitoring missions is not a reasonable option. In addition, single-purpose automation software is relatively expensive to develop and maintain. A Linux-based control architecture was developed to facilitate construction of custom system automation and analysis applications. This stable control architecture provides the ability to design and automate complex chemical and physical processing tasks in the lab or field environment, implemented so far for the routine measurement of short lived xenon isotopes. This is achieved through a framework of clients and servers designed under the Linux operating system. Written under the C++ language, a client/server architecture was developed for automated systems enabling modularized processes to perform system control. In this scheme, narrowly focused clients and servers communicate with each other to perform tasks necessary for system execution. Each hardware interface, such as analog input/output modules, instrumentation, and detectors, have a server associated with it that can be accessed individually with various client interface programs. This architecture offers several advantages such as running subcomponents independently. This, for instance, would provide a simple way for testing and calibrating detectors without the need of an entire processing system. A state-machine based client/server program facilitates repetitive system execution. This allows a highly flexible mechanism to control systems and tailor them as needed.

Log: 384. **UNDERGROUND SOURCES OF RADIOACTIVE NOBLE GAS.** J.C. Hayes, T.W. Bowyer, K.B. Olsen, V.T. Woods, Pacific Northwest National Laboratory.

It is well known that radon is present in relatively high concentrations below the surface of the Earth due to spontaneous fission of uranium and thorium. However, the background levels of other isotopes such as Xe-133 and Xe-131m are important to understand in the sub surface because of their potential use to confirm violations of the Comprehensive nuclear Test Ban Treaty (CTBT) during an On Site Inspection (OSI). Recently PNNL carried out a number of measurements of xenon from the subsurface at the Nevada Nuclear Security Site (formerly known as the Nevada Test Site) to determine whether xenon isotope background levels could be detected from spontaneous fission or possibly from legacy 240Pu from historic nuclear testing. Results of those measurements will be presented. A review will also be presented of sources of xenon backgrounds that need to be accounted for during OSI noble gas measurements.

Log: 385. **MEASUREMENT OF FUKUSHIMA AEROSOL DEBRIS IN SEQUIM AND RICHLAND, WA AND KETCHIKAN, AK.** HS Miley, TW Bowyer, MD Engelmann, PW Eslinger, JA Friese, LR Greenwood, DA Haas, JC Hayes, ME Keillor, , RA Kiddy, RR Kirkham, JW Landen, EA Lepel, LS Lidey, KE Litke, SJ Morris, KB Olsen, RC Thompson, BR Valenzuela, VT Woods; Pacific Northwest National Laboratory, Richland, WA and SR Biegalski; The University of Texas at Austin, Austin, Texas.

Aerosol collections were initiated at several locations by PNNL shortly after the Great East Japan Earthquake of May 2011. Aerosol samples were transferred to laboratory high-resolution gamma spectrometers for analysis. Similar to treaty monitoring stations operating across the Northern hemisphere, iodine and other isotopes which could be volatilized at high temperature were detected. Though these locations are not far apart, they have significant variations with respect to water, mountain-range placement, and local topography. Variation in computed source terms will be shown to bound the variability of this approach to source estimation.

Log: 386. **COMPARISON OF PARALLEL RADIONUCLIDE COLLECTION AND ANALYSIS OF FUKUSHIMA DEBRIS IN RICHLAND, WA.** VT Woods, TW Bowyer, LR Greenwood, DA Haas, JC Hayes, ME Keillor, EA Lepel, HS Miley, SJ Morris; Pacific Northwest National Laboratory, Richland, WA and SR Biegalski; The University of Texas at Austin, Austin, Texas.

Two independent samplers were operated at PNNL in parallel during the acquisition of samples from the Fukushima reactor releases. One system is an automated aerosol collection and analysis unit, while the other was a modestly sized manual sampler of even higher daily air volume. The samples collected each day showed excellent similarity, although some variations were seen. These variations in a way define the reproducibility of the measurements made by the CTBT International Monitoring System, and show a simple way that useful parallel samples can be acquired for scientific purpose. In particular, a party wishing to have a 'copy' of a sample acquired by the Verification Regime of the Treaty could employ this method and have results similar to the IMS station at low cost and even higher sensitivity.

Log: 387. **CHROMATOGRAPHIC SEPARATION OF ERBIUM AND THULIUM FOR NEUTRON CAPTURE CROSS SECTION MEASUREMENTS.** Bene, BJ; Gharibyan, N; Sudowe, R. University of Nevada, Las Vegas.

The National Nuclear Science Administration's Stockpile Stewardship Academic Alliance program cites the "investigation leading to greater accuracy in the knowledge of low energy cross sections of stable and unstable nuclei and corresponding reaction rates for neutron-, γ - and ion-induced reactions" as one of the major research areas in low-energy nuclear science. One of these unstable nuclei of interest is Tm-171 which is part of a reaction network of elements that were used as radiochemical detectors to monitor the performance of nuclear devices. Tm-171 also plays an important role in astrophysics, as a branching point in the s-process of stellar nucleosynthesis. Measurement of its neutron capture cross section will help to gain better understanding of nuclear processes in stars as well as help Science-Based Stockpile Stewardship efforts. Tm-171 will be produced through neutron capture on stable Er-170 followed by beta decay. In order to perform neutron capture cross section measurements on Tm-171, milligram amounts need to be separated from an irradiated erbium target. A highly efficient and very selective separation based on ion chromatographic methods is currently under development to separate these neighboring lanthanides. Results of the optimization of an analytical scale cation exchange method with respect to various parameters such as temperature, flow rate, eluent and lanthanide concentrations will be presented.

Log: 388. **WHEN IS IN SITU GAMMA SPECTROMETRY MOTIVATED?** Boson, J(1); Ramebäck, H(1); Nylén, T(1). (1) Swedish Defence Research Agency (FOI).

For several decades, gamma-ray spectrometry measurements in situ have been a widely used method for identification and quantification of ground deposition of radionuclides. However, since radionuclides to some extent will penetrate into the ground, particularly in the case of wet depositions, because of the presence of vegetation or ground roughness, and due to the inherent measurement geometry, there will always be some attenuation affecting the emitted radiation, which will result in a decrease of the gamma rays incident on the detector. Therefore, the measurement efficiency is sensitive to the various source parameters such as soil density, ground roughness and activity depth distribution, and this often leads to large uncertainties in in situ measurements. This can of course be countered by extensive sampling to determine e.g. soil density and activity depth distribution. Paradoxically, however, if the activity content in a large number of soil samples is determined in order to estimate the activity depth distribution, one can get a good assessment of the total ground deposition activity directly from these samples, thus rendering the in situ measurement superfluous. It is therefore of interest to study the combined measurement uncertainty of in situ measurements as a function of the number of soil samples collected, i.e. as a function of the time and work load needed to characterize the source. When higher uncertainties can be tolerated, fewer samples will be needed for the source

characterization, but when lower uncertainties is needed for the measurement to be fit-for-purpose, soil sampling and laboratory measurement might be a better option.

Log: 389. **COMPARATIVE EVALUATION OF PRECONCENTRATION METHODS FOR THE RAPID ANALYSIS OF ACTINIDE ELEMENTS AND STRONTIUM.** Bene, KE; Bene, BJ; Sudowe, R. University of Nevada, Las Vegas.

Radiochemical separations are an essential part of nuclear forensic analysis used to identify the origin of nuclear material interdicted by law enforcement agencies, or to obtain signatures from radioactive samples collected after an explosion of a radiological dispersion device or improvised nuclear device. Especially in a post detonation situation it is vital to obtain results in as little time as possible. Therefore rapid methods for determining actinides and fission products are currently under development by a variety of laboratories. Optimization of these procedures mainly focuses on minimizing the time needed for dissolution or leaching of the sample and separation of the analytes. Another step, preconcentration of analytes, is however often overlooked even though it is an essential part of the analytical procedure especially in case of voluminous samples. In this study several preconcentration methods that are customarily used in the determination of actinide elements and strontium were investigated to provide a systematic comparison of their usefulness and ease. The recoveries were determined by liquid scintillation counting exploiting its high efficiency and the relative ease of sample preparation. Results in terms of recovery of strontium, thorium, uranium, plutonium and americium as well as the required time will be presented.

Log: 390. **A TECHNIQUE TO DISTINGUISH PLUTONIUM SOURCE TERMS IN ENVIRONMENTAL MIXTURES.** Oldham, WJ; Dry, DE; Bowen, SM; Goldstein, SJ; Murrell, MT. Nuclear and Radiochemistry (C-NR), Los Alamos National Laboratory.

A technical challenge in many environmental monitoring programs is distinguishing between local nuclear effluents and the ubiquitous actinide and fission product background derived from atmospheric nuclear testing. Efforts to uniquely identify multiple plutonium sources contained within environmental collections have previously achieved only mixed success. General techniques based on plutonium isotopic measurements and correlations between ^{239}Pu and fission products such as ^{137}Cs and ^{90}Sr require at least some prior independent knowledge of the non-fallout end-member, as well as consistent geochemical behavior between disparate chemical elements. Unfortunately, Cs, Sr and Pu each behave differently in the environment and attempted correlations between these elements have proven to be unreliable for detailed assessment. This paper will discuss a new method to identify atmospheric fallout in environmental collections based on radioanalytical measurements of the long-lived lanthanide fission product, ^{151}Sm ($t_{1/2} = 90$ years). This lanthanide fission product shows similar geochemical behavior compared to plutonium and americium isotopes and can in principle be used to "subtract" fallout-derived plutonium from sample mixtures that also contain a local plutonium source term.

Log: 391. **RUSTY NAILS ABSORB TRACE PLUTONIUM FROM THEIR LOCAL ENVIRONMENT.** Oldham, WJ; Miller, JL; Attrep, M. Nuclear and Radiochemistry (C-NR), Los Alamos National Laboratory.

Plutonium released into the environment displays a strong affinity for particulate and mineral surfaces such as iron oxides, manganese oxides, and aluminosilicates. Iron oxides are of particular interest because iron and iron-containing alloys are extensively used in contemporary society and may inadvertently function as high efficiency plutonium scavengers. Metals of this class, commonly covered with an oxide coating, could adsorb and sequester trace levels of plutonium encountered within their local environment. To test this intriguing idea we have collected discarded metal artifacts (rusty nails, bottle caps, etc) from remote environments around New Mexico and have analyzed their surfaces for adsorbed plutonium. The analytical method uses dilute hydrochloric acid to leach the oxide coating down to bare metal. The plutonium analyte contained within this solution is then purified using an anion-exchange procedure and then assayed using alpha spectrometry and ICP-MS. Plutonium present in these environments

due to fallout during the era of atmospheric nuclear testing is easily measured using this technique. Given the observed variation in absolute plutonium concentration as a function of sample size, time of exposure, etc we have also measured natural uranium contained in the same samples for use as a concentration proxy to facilitate sample inter-comparison.

Log: 392. **APPLICATION OF SYNCHROTRON RADIATION X-RAY MICROFLUORESCENCE IN BONE IMPLANTS.** Lima, I (1), Machado, C (2), Calansans-Maia, M (3), Granjeiro, J (4), Pereira, G (5), Lopes, R (1), (1) Nuclear Instrumentation Laboratory, PEN/COPPE/UFRJ, (2) Medical Science Program, UFF, Niterói/RJ, Brazil, (3) Oral Surgery Department, UFF, Niterói/RJ, Brazil, (4) Molecular and Cell Biology Department, UFF, Niterói/RJ, Brazil, (5) Department of Metallurgical and Materials Engineering, PEMM/COPPE/UFRJ, Rio de Janeiro/RJ, Brazil..

The bone is a specialized connective tissue that adapts to the shape and volume to functional demands of a complex and dynamic way, due its ability to regenerate completely, supplying the mineral needs and provides the mechanical support of the body. Its matrix is composed of organic and mainly inorganic elements, such as hydroxyapatite. Nevertheless, the process of bone repair can be affected by hereditary diseases, congenital or acquired at any age, infectious or otherwise, that culminate in dynamic bone reactions. This paper intends to show some applications in bone research, such as bone implant and osteoporosis drugs treatments in the health field, which provides a great increase in human's life expectancy. For that purpose, X-ray microfluorescence with synchrotron radiation imaging technique was used as a significant tool in order to investigate morphology and minerals contents parameters in micro order levels. The elemental mapping was carried out at Brazilian Synchrotron Light Laboratory, Campinas – São Paulo, Brazil working at D09-XRF beam line. A white beam (4 keV min – 23 keV max) was used for samples excitation. In order to focusing the x-ray beam it was used a capillary optics which is one of the best suits a micrometer XRF instruments. In this case, it was used a glass conical capillary of 20µm diameter. It was used a 45/45 degree geometry which means that the samples were placed at 45° to the incident beam and at this same angle of degree an Si(Li) solid state detector, with a resolution of 165 eV at 5.9 keV was used to collected the x-ray fluorescent radiation. The measurements were performed in a computed controlled XYZ table which allowed the choice of the interest region in each sample. It was possible to demonstrate that synchrotron radiation x-ray fluorescence microscopy reveals novel features of bone morphology and composition, which can lead the knowledge of several important parameters, such as porosity, connective and mineral content.

Log: 393. **APPLICATION OF SYNCHROTRON RADIATION IN 3D X-RAY MICROTOMOGRAPHY.** Pereira, G.R. (1), Rocha, H.S. (2), Anjos, M.J. (3), Lima I. (2), Lopes, R.T.(2), (1) Non-destructive Testing, Corrosion and Welding Laboratory(LNDC), Department of Metallurgical and Materials Engineering COPPE/UFRJ, Federal University of Rio de Janeiro, Rio de Janeiro/RJ - Brazil, (2) Nuclear Instrumentation Laboratory, Department of Nuclear Engineering COPPE/UFRJ, Federal University of Rio de Janeiro, Rio de Janeiro/RJ - Brazil, (3) Physics Institute –Stated University of Rio de Janeiro, Rio de Janeiro/RJ - Brazil

An X-ray Transmission Microtomography (CT) system combined with an X-ray Fluorescence Microtomography (XRFµCT) system was implemented in the Brazilian Synchrotron Light Laboratory (LNLS), Campinas, Brazil. The main objective of this work is to determine the elemental distribution in prostate, breast and lung samples in order to verify the concentration of some elements correlated with characteristics and pathology of each tissue observed by the transmission CT. The experiments were performed at the X-Ray Fluorescence beamline (D09B-XRF) of the Brazilian Synchrotron Light Laboratory, Campinas, Brazil. A quasi-monochromatic beam produced by a multilayer monochromator was used as an incident beam. The sample was placed on a high precision goniometer and translation stages that allow rotating as well as translating it perpendicularly to the beam. The fluorescence photons were collected with an energy dispersive HPGe detector placed at 90° to the incident beam, while transmitted photons were detected with a fast Na(Tl) scintillation counter placed behind the sample on the beam direction. The CT images were reconstructed using a filtered-back projection algorithm and the

XRF μ CT were reconstructed using a filtered-back projection algorithm with absorption corrections. The 3D images were reconstructed using the 3D-DOCTOR software. Analyzing the 3D visualization, it can be observed that the distribution of iron, copper and zinc are heterogeneous in those samples. XRF μ CT technique has enabled us to determine the elemental distribution inside of the sample without destroying it.

Log: 394. **A NEW (OLD) ROUTE TO THE ISOTOPIC PURIFICATION OF ACTINIDE TRACERS.** O.T. Farmer III (1), M. Liezers (1), M.L. Thomas (1), G. Eiden (1) and D. Duckworth (1), (1) Pacific Northwest National Laboratory.

One of the biggest limitations to precise low level actinide (U, Np, Pu) isotope ratio measurement is the isotopic purity of the tracers/standards available. Standards currently available always contain trace amounts of the analytical isotopes of interest that limits spiking levels which in turn impairs measurement precision. Advances in mass spectrometry now make it possible to enhance the isotopic purity of tracers like Pu²⁴⁴ on a small scale allowing the use of higher tracer spiking levels without degrading the low level detection of the lighter Pu isotopes. Work on this new research will be presented.

Log: 396. **XE-135 PRODUCTION FROM CF-252.** C.A. McGrath, Idaho National Laboratory, T. P. Houghton, Idaho National Laboratory, J. K. Pfeiffer, Idaho National Laboratory, R. K. Hague, Idaho National Laboratory.

¹³⁵Xe is a good indicator that fission has occurred and is a valuable isotope that helps enforce the Comprehensive Test Ban Treaty. Due to its rather short half life and minimal commercial interest, there are no known commercial suppliers. Readily available standards of this isotope for calibrating collection and analytical techniques would be very useful. ¹³⁵Xe can be produced in the fissioning of actinide isotopes, or by neutron capture on ¹³⁴Xe. Since the neutron capture cross section of ¹³⁴Xe is 3 mB, neutron capture is a low yield, though potentially useful, production route. ¹³⁵Xe is also produced by spontaneous fission of ²⁵²Cf. ²⁵²Cf has a spontaneous fission rate of about 6×10^{11} /s/g. The cumulative yield from the spontaneous fission of ²⁵²Cf is 4.19%; and the competing neutron capture reaction that depletes ¹³⁵Xe in thermal reactor systems is negligible because the neutron capture cross-section is low for fast fission neutrons. At the INL, scientists have previously transported fission products from an electroplated ²⁵²Cf thin source for the measurement of nuclear data of short-lived fission products using a technique called He-Jet collection. We have applied a similar system to the collection of gaseous ¹³⁵Xe, in order to produce valuable standards of this isotope.

Log: 397. **REMOTE EARLY WARNING FISSION AND ACTIVATION PRODUCT INDICATORS OF SOURCE IDENTIFICATION, AND MECHANISMS AND EXTENT OF RELEASE FROM THE FUKUSHIMA DAIICHI NUCLEAR FACILITY DISASTER.** Jon Schwantes & Christopher Orton, Pacific Northwest National Laboratory.

Measurements of several radionuclides within environmental samples taken from the Fukushima Daichi nuclear facility following the recent tsunami-initiated catastrophe were evaluated for the purpose of identifying the source term, reconstructing the release mechanisms, and estimating the extent of the release. Cs-136 to Cs-137 ratios conclusively identified Units 1-3 as the major source of radioactive contamination to the close-in environment surrounding the facility. A trend was observed between the fraction of the total core inventory released for a number of fission product isotopes and their corresponding Gibbs Free Energy of formation for the primary oxide form of the isotope, suggesting that release was dictated by chemical volatility driven by temperature and reduction potential within the primary containment of the vented reactors. Pu-238/Pu-239,240 ratios close-in and at 30 km from the facility indicated that the damaged reactors were the major contributor of Pu to surface soil at the source but that this contribution decreased rapidly with distance from the facility. The fraction of the total Pu inventory from units 1-3 released was estimated to be less than 0.1% based upon Pu/Cs isotope ratios relative to the within-reactor modeled inventory prior to venting and was consistent with an independent model evaluation that

considered chemical volatility based upon measured fission product release trends. Significant volatile radionuclides within the spent fuel at the time of venting but not as yet observed within environmental samples are suggested as potential analytes of concern for future environmental surveys around the site.

Log: 398. **ELECTROCHEMICALLY MODULATED SEPARATION OF IODINE FOR ANALYSIS OF I-129.** Pratt, SH; Engelman, MD; Hart, GL; Peper, SM; Sweet, L; Breshears, AT; Duckworth, DC (PNNL all).

An electrochemically modulated separations (EMS) method has been developed for the preconcentration and isolation of iodine for the purposes of determining I-129/I-127 ratios. The EMS cell was coupled to an inductively coupled plasma time of flight mass spectrometer (ICP-TOF-MS) for development of the method. Because I-129 is a fission product, and iodine is mobile, it is particularly useful for environmental monitoring for human dose evaluation. The goal of this work is to preconcentrate and isolate iodine with high recovery efficiency. It is envisioned that the method could be used on-line with multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) for rapid separation and analysis, with the MC-ICP-MS having a high dynamic range for the ratio measurement. This envisioned application, and challenges therein, will be discussed along with the separation method. The EMS method to be presented uses a bare gold electrode to which iodine adsorbs from a basic solution when the applied voltage is approximately 0 V. Deposition efficiencies for iodine adsorption are 80 – 90%. Adsorbed iodine can be release by applying a negative voltage and reducing it to iodide. None of the other halides adsorbed to gold in basic media, so iodide could be completely separated from bromide and chloride. Through this presentation, the benefits of using electrochemical redox reactions to effect separation and concentration of iodine in a rapid manner for mass spectrometric analysis will be demonstrated.

Log: 399. **TAKING A SECOND LOOK: RE-EXAMINING GROUND COLLECTED FALLOUT FROM LOW YIELD SURFACE NUCLEAR TESTS FOR FORENSIC SIGNATURES.** Gostic, RC (1); Knight KB (1); Spriggs G (1); Hutcheon I (1). (1) Lawrence Livermore National Laboratory.

Fallout samples from low yield surface and near surface nuclear tests are being re-examined for their forensic value using a combination of radiometric and mass spectrometry based techniques. Data from these studies indicate that soil samples collected along fallout plumes contain easily accessible information about key components of each test such as fuel isotopics and the elemental composition of structural component. Glassy material manually extracted from the soil samples constitute <5% of the bulk soil mass, yet contain >50% of the total activity. By gamma spectroscopy ²³⁵U concentrations are estimated to be >30 ug/g and ²³⁹Pu concentrations >20 ug/g in glasses recovered from two different tests. Analysis of the U bearing glasses by ICP-MS indicates a fuel with a minimum enrichment of 84% ²³⁵U. The stable isotope signal from ICP-MS measurements shows that Be is present in the glass at concentrations 4 times higher than the bulk soil, and that Cr, Co and Mo are present in the U bearing glasses at 3-20 times the bulk soil concentration. These results demonstrate that fallout from low yield surface nuclear tests collected as surface deposits long after the detonation contains valuable information about device characteristics. This information is relevant to characterizing historical activities from surface based nuclear test programs and is applicable to nuclear forensics research. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 400. **EXPERIMENTAL HALF-LIFE MEASUREMENTS OF SHORT-LIVED FISSION PRODUCTS & COMPARISON TO CURRENT ACCEPTED HALF-LIFE VALUES.** Bruce D Pierson, University of Michigan.

A sample of high-purity thorium-oxide powder was studied using cyclic irradiations with an accelerator-based neutron generator at the University of Michigan and a high-purity germanium detector. Each count cycle involved a 2 second irradiation at the face of the generator followed by

300 seconds of counting starting ~350 ms after irradiation. Precise control of the irradiation duration and transit times from the generator to the detector was provided by a set of optical sensors controlled by a pneumatic transfer system. The long count periods allowed for capture of the decay of numerous short-lived fission products like Arsenic-85 and Zirconium-99, but, more importantly, were used to minimize build-up of short-lived activity within the sample. Cyclic irradiations were performed to enhance the statistics of the half-life measurements. The data was also collected as a stream of time stamped events rather than as discrete spectra to ensure accurate time comparison between cycles. The half-lives of isotopes far from the line of stability along isobars with complex decay schemes are not well known. Half-life measurements from these experiments could provide more accurate results than the currently accepted half-life values. Higher accuracy values will enhance the fidelity of modeling delayed gamma-ray intensities at short times; thus, improving modeling predictions from active interrogation of fissile and fissionable materials.

Log: 401. **COMPARISON OF RADIONUCLIDE RATIOS IN ATMOSPHERIC NUCLEAR EXPLOSIONS AND NUCLEAR RELEASES FROM CHERNOBYL AND FUKUSHIMA SEEN IN GAMMA RAY SPECTROMETRY.** JA Friese; RF Kephart; DD Lucas. Pacific Northwest National Laboratory, Richland, WA.

The Comprehensive nuclear Test Ban Treaty (CTBT) is verified via remote radionuclide monitoring followed by an On Site Inspection (OSI) to clarify the nature of a suspect event. An important aspect of doing radionuclide measurements on site is the discrimination of other potential sources of similar radionuclides such as reactor accidents or medical isotope production. The Chernobyl and Fukushima nuclear reactor disasters offer two different reactor source term environmental inputs that can be compared against historical measurements of nuclear explosions. The comparison of whole-sample gamma spectroscopy measurements from these three events and the analysis of similarities and differences are presented. This analysis is a step toward confirming what is needed for measurements during an OSI under the auspices of the Comprehensive Test Ban Treaty.

Log: 402. **QUANTITATIVE ANALYSIS OF LONG-LIVED LANTHANIDE FISSION PRODUCTS: NAMELY 151SM AND 155EU.** Stosh A. Kozimor, Scott M. Bowen, Donald E. Dry, Warren J. Oldham, Stuart D. Ware.

A key technical requirement associated with the environmental monitoring of trace nuclear materials involves a demonstrated ability to detect and characterize plutonium isotopic ratios. Inter-element ratios involving long-lived fission products and plutonium can also provide important signatures of source terms and age. This, however, requires the behavior of these elements in the environment to be well understood. Unfortunately for many long-lived fission products, i.e. Cs and Sr, these chemical processes are complicated and render it difficult draw meaningful conclusions from the relative elemental abundances. Described here is a new research effort that focuses on developing methods to accurately measure low levels of long-lived lanthanide fission products that are anticipated to track plutonium in the environment, namely 151Sm and 155Eu. Specifically we will present a robust method for isolating these elements from large environmental collections and describe our efforts to advance current methods for detecting these isotopes and age dating the analyzed samples.

Log: 403. **MEASUREMENTS OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN FOOD SAMPLES AND WATER FOR HUMAN CONSUMPTION IN AUSTRIA FOR THE CALCULATION OF THE INGESTION DOSE.** Landstetter, C; Katzlberger, C. Austrian Agency for Health and Food Safety.

A new report on food habits of the Austrian population in the year 2006/07 was released in 2008. According to the Austrian radiation protection law, food law and the Commission Recommendation 2000/473/Euratom on the application of article 36 of the Euratom Treaty concerning the monitoring of the levels of radioactivity in the environment for the purpose of

assessing the exposure of the population as a whole, mixed diet and foodstuffs are measured within a monitoring program. In addition to this drinking and mineral water samples are measured for natural and artificial radionuclides. Based on the results of these measurements, literature data and the data of the new report on food habits the ingestion dose for the Austrian population is recalculated. In general the major part of the ingestion dose is caused by natural radionuclides especially K-40. The influences of different diet habits are discussed and exemplarily quantified.

Log: 404. **COMPARISON OF IN VITRO DISSOLUTION OF URANYL ACETATE USING THE TRADITIONAL FILTER SANDWICH METHOD AND THE SLIDE-A-LYZER MINI DIALYSIS DEVICE.** Murry, MM (1); LaMont, S (2); Glover, SE (1); Spitz, HB (1). (1) University of Cincinnati, (2) Los Alamos National Laboratory.

The solubility of a material is an important parameter in evaluating the risk to humans following accidental inhalation of a substance. Conventional methods of measuring in vitro solubility involve exposing the substance to a solvent such as water, saline, or solutions which simulate lung fluid, and measuring the fraction of material that dissolves with time. The method for determining solubility of airborne contamination typically involves sealing an air filter, on which a known particle mass or activity has been collected, between two hydrophilic membranes and immersing the assembly in the desired solvent for pre-determined periods of time. This method is impractical if the contaminant is a single, micron-sized particle. A new method of isolating particles has been investigated that replaces the membrane filter "sandwich" with the Slide-A-Lyzer MINI Dialysis Device which is a low-binding polypropylene and integrated regenerated cellulose membrane attached to the bottom of a plastic insert cup that conveniently fits into a conical tube for dialysis. The evaluation of the filter sandwich method verse the dialysis device method for in vitro dissolution testing will identify whether the dialysis device method has similar kinetic diffusion properties as the filter sandwich method. Uranyl acetate was used to evaluate its rate of dissolution using the filter sandwich method and the Slide-A-Lyzer MINI Dialysis Devices. The filter sandwich method confined the uranyl acetate between two filters and exposed solvent to both sides of the filter. The Slide-A-Lyzer MINI Dialysis Devices confined the uranyl acetate in an insert cup and exposed only the bottom of the insert cup to the solvent. Historical data using the filter sandwich method found that the majority of uranyl acetate dissolved into solution within the first 30 minutes.

Log: 405. **HIGH PERFORMANCE ION CHROMATOGRAPHY FOR SEPARATION OF FISSION AND ACTIVATION PRODUCTS.** Erin Finn, Leah Arrigo, Kevin Swearingen, Lori Metz, Matt Douglas. Pacific Northwest National Laboratory.

Analysis of activation and fission products is a complex and time-consuming endeavor requiring separation of inorganic species spanning the entire periodic table. Separation and subsequent quantification of activated isotopes of environmental and structural materials following detonation may provide useful information about the possible dose to nearby individuals. Instrumental techniques such as High Performance Ion Chromatography (HPIC) have the potential to provide more efficient and more rapid separations than are possible using traditional bench chemistry techniques. In this work, we examine HPIC methods for separation of various activation product ions from each other to remove interfering species and improve the limits of detection. Separated fractions are collected and quantification is achieved by radiometric or analytical instrumental methods. Ionic species of beryllium, strontium, cesium, barium, sodium, calcium, magnesium, iron, chromium, tungsten, scandium, manganese, cobalt, cadmium, nickel, zinc, and lanthanides are considered. Separation of the alkali and alkaline earth cations can be readily accomplished on a cation exchange column, while separation of most transition metals can be accomplished by use of a bifunctional ion exchange column. However, ions retained on the cation exchange column are not necessarily retained on the bifunctional column, and vice versa, so that sequential and parallel injections are necessary in order to accommodate the diverse chemistries of the elements of interest. This work details the methods developed to accommodate the full suite of elements listed.

Log: 406. **COMMERCIALLY AVAILABLE SYSTEM OFFERS HIGH-SENSITIVITY ANALYSIS OF RADON IN 2.5L WATER SAMPLES.** Derek Lane-Smith, DURRIDGE Company.

Radon concentrations in open water are often just a few dpm/L. While systems that bring a closed loop of air into equilibrium with the water, such as the RAD AQUA, are easily able to measure such low concentrations, till now there has been nothing available commercially that can reliably analyze a discrete sample with such a low concentration. The DURRIDGE Big Bottle RAD H2O provides a standard, commercially available system capable of measuring radon concentrations in water down to 1 pCi/L (2.2 dpm/L) and, with care, below. The system is described, the algorithm for calculating the radon in the water is derived, some precautions are highlighted and examples of data are presented with an analysis of their uncertainty. (If the session chairman is agreeable, a short video demonstrating the method can be shown.).

Log: 407. **THE DESIGN, CONSTRUCTION, AND INITIAL CHARACTERIZATION OF AN ULTRA-LOW-BACKGROUND GAS-PROPORTIONAL COUNTING SYSTEM.** A. Seifert, C.E. Aalseth, A.R. Day, E.S. Fuller, E.W. Hoppe, M.E. Keillor, E.K. Mace, C.T. Overman, G.A. Warren. Pacific Northwest National Laboratory, Richland, WA, USA.

Over the past several years, the Department of Energy has funded the development of an ultra-low background proportional counter (ULBPC) technology at the Pacific Northwest National Laboratory (PNNL). The resulting detector is the product of an effort to produce a low-background, physically robust gas proportional counter for applications like radon emanation measurements, groundwater tritium, and ^{37}Ar . In order to fully take advantage of the inherent low-background properties designed into the ULBPC, a comparably low-background dedicated counting system is required. An ultra-low-background counting system (ULBCS) was recently built in the new shallow underground laboratory at PNNL. With a design depth of 30 meters water-equivalent, the shallow underground laboratory provides approximately 100x fewer fast neutrons and 6x fewer muons than a surface location. The ULBCS itself provides additional shielding in the form of active anti-cosmic veto (via 2-in. thick plastic scintillator paddles) and passive borated poly (1 in.), lead (6 in.), and copper (3 in.) shielding. This work will provide details on PNNL's new shallow underground laboratory, examine the motivation for the design of the counting system, and provide results from the characterization of the ULBCS, including initial detector background.

Log: 408. **CHARACTERIZATION METHODOLOGY FOR THE INDIANA UNIVERSITY NREP FACILITY.** M. Halstead (1), J. Petrosky (1), A. Bickley (1) P. Sokol(2) S. Clark (3) (1) AFIT/ENP (2) Indiana University (3) NSWC Crane .

The neutron energy spectrum of the NREP target at the Indiana University Center for Exploration of Energy and Matter is being characterized using a combination of Monte Carlo simulations along with spectrum unfolding using SAND-II. The unfolding algorithm uses neutron activation analysis data in conjunction with an initial guess to deduce the spectrum. To properly understand the resultant spectrum and ensure minimal error, an analysis of systemic and calculation uncertainty is accomplished. Error is tracked through the analysis process from the start of experiment to the final unfolding results. We find that experimental error is currently dominant but can be mitigated with several procedural steps. In unfolding, we find that cross-section data tables are the limiting factor. Being written in the 1960s, SAND-II includes cross-section tables that are coarse and limited. A Matlab script has been written that utilizes the ENDF/B-VII tables, which are far more detailed with lower error estimations. A comparison of the results from these two versions demonstrates extensive error reduction. Additionally, using a modified Monte Carlo technique for minimizing the least-squares difference at each iteration step, solution convergence time is minimized and total error propagation is reduced. The final result is a tool that can be used for analyses of spectra with confidence in the outcome and a comprehensive understanding of the uncertainty.

Log: 409. **RESOLUTION OF URANIUM ISOTOPES WITH KINETIC PHOSPHORESCENCE ANALYSIS.** S.M. Miley (Pacific Northwest National Laboratory), A.T. Hylden (Pacific Northwest National Laboratory and University of Pennsylvania), J.I. Friese (Pacific Northwest National Laboratory).

This study was conducted to test the ability of the Chemchek Kinetic Phosphorescence Analyzer Model KPA-11 with an auto-sampler to resolve the difference in phosphorescent decay rates of several different uranium isotopes, and therefore identify the uranium isotope ratios present in a sample. Kinetic phosphorescence analysis (KPA) is a technique that provides rapid, accurate, and precise determination of uranium concentration in aqueous solutions. Utilizing a pulsed-laser source to excite an aqueous solution of uranium, this technique measures the phosphorescent emission intensity over time to determine the phosphorescence decay profile. The phosphorescence intensity at the onset of decay is proportional to the uranium concentration in the sample. Calibration with uranium standards results in the accurate determination of actual concentration of the sample. Different isotopes of uranium, however, have unique properties which should result in different phosphorescence decay rates seen via KPA. Results will show that a KPA instrument is capable of resolving uranium isotopes.

Log: 410. **CHARGED-PARTICLE ACTIVATION ANALYSIS: A NOVEL APPROACH TO NUCLEAR METROLOGY.** M.Anwar Chaudhri (1,2) , M. Nasir Chaudhri (2). 1. University of Erlangen-Nuernberg. 2. Pakistan Council of Scientific & Industrial Research.

A novel approach has been developed to study the effect of French Atomic Tests series of 1974 on the Australian atmosphere. This is to investigate the changes in the elemental/isotopic concentrations of the atmospheric particulates collected in Australia just before and just after the onset of the atomic tests in the Pacific. The atmospheric particulates were collected on Polystyrene filters in high volume air samplers placed all along the Australian East Coast at locations in Port Moresby (New Guinea), Townsville, Brisbane, Sydney, Melbourne and Hobart. The elemental concentrations in the filters were measured by using the technique of charged particle activation analysis. A number of elements, such as S, Ca, Ti, Cr, Fe, Ni, Cu, Zn, Se and Hg ranging in concentrations from 0.001- 3.27 micrograms / cubic metre, were detected. The changes observed in the concentrations of these elements in the two sets of samples, taken just before and just after the Atomic Tests, are attributed to synoptic rather than Nuclear Fall-Out effects. With the availability of more versatile analytical methods, such as the ICP-MS, it is suggested that this new approach to nuclear metrology should be more commonly used.

Log: 411. **INDUCTIVELY- COUPLED-PLASMA-MASS-SPECTROMETRY (ICP-MS) AND IST APPLICATIONS IN BIOMEDICAL FIELDS.** M. A. Chaudhri (The University of Erlangen-Nuernberg, Germany & Pakistan Council of Scientific and Industrial Research, Lahore, Pakistan) AND J. Watling (Forensic Sciences, University of Western Australia, Perth, Australia).

The ICP-MS is very sensitive technique for trace elements determination in a variety of samples, including bio-medical. Most of the elements and isotopes from Li up to U can be investigated simultaneously with this technique with detection sensitivity of as low as a fraction of one ppb, in favourable circumstances. The samples have to be in liquid form, as is the case of most of the body fluids, or brought into liquid form. However, for studying solid samples, a Laser Ablation Facility has also been installed, After a brief introduction of this technique its applications in the following biomedical fields are presented:1. The role and significance of trace elements in osteopenia and osteoporosis 2.Determination of trace elements in eye-chambers fluid – a unique approach to study possible causes of Glaucoma and age-related Macula Degeneration (AMD).. 3. Micro distribution of trace elements in bladder and kidney stones..4.Elemental composition of some proven herbal medicines. 5. Hyper-tension and trace elements in Pakistani population. 6.Uranium in Bavarian (German) females The convenience and versatility of this powerful physical sciences analytical technique, as a better alternative to nuclear analytical methods, would be discussed.

Log: 412. **THEORIES OF CHARGED-PARTICLE ACTIVATION ANALYSIS.** M: Anwar Chaudhri(1,2) and M. Nasir Chaudhri(1) : 1. Pakistan Council of Scientific & Industrial Research 2. The University of Erlangen-Nuernberg, Germany.

The accuracy of Chaudhri's method for charged-particle activation analysis 1, has been further demonstrated by extensive calculations. The nuclear reactions $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$, $^{63}\text{Cu}(\text{He},\text{p})^{65}\text{Zn}$, $^{107}\text{Ag}(\alpha,\text{n})^{110}\text{In}$ and $^{208}\text{Pb}(\text{d},\text{p})^{209}\text{Pb}$, whose cross sections were easily available have been examined for the detection of ^{12}C , ^{63}Cu , ^{107}Ag and ^{208}Pb respectively in matrices of Cu, Zr and Pb, at bombarding energies of 4 – 22 MeV. The "standard" is assumed to be in a carbon matrix. It has been clearly demonstrated that Chaudhri's method, which makes the charged particle activation analysis as simple as neutron activation analysis, provides results which are almost identical to, or only about 1-2 % different, from the results obtained by using the full "Activity Equation" involving solving complex integrals. It is valid even when the difference in the average atomic weights of matrices of the standard and the sample is large.

Log: 413. **TRACE ELEMENTS IN OSTEOPENIA/OSTEOPOROSIS: A POTENTIAL, RADIATION FREE AND CHEAP METHOD FOR BONE-MINERAL-ASSESSMENT.** M.A. Chaudhri (Uni. Of Erlangen-Nuernberg & Pakistan Council of Scientific & Industrial Research), W. Kemmler (University of Erlangen-Nuernberg), I. A. Harsch (Uni. Of Erlangen-Nuernberg), J. Watling (Uni. Of Western Australia).

In order to examine the possible role of trace elements in bone metabolism and osteoporosis we have measured the concentrations of more than 30 elements from Li up to U (with special attention to Mg, Fe, Cu, Zn, Sr & Se) in plasmas of 25 osteopenic females, who have been attending the Osteoporosis Research Centre of the University of Erlangen-Nuernberg. The bone mineral densities (BMD), both of the hip as well as of the Lumbar Spine L1-L4, were determined using the commonly used technique of Dual Beam X-Ray Absorptiometry. About 3-5 ml blood sample was obtained from each subject, separated into plasma and red cells and freeze dried. The elemental analyses were carried out in our laboratories in Perth using the technique of Inductively-Coupled-Plasma Mass Spectrometry (ICP-MS), which has the capability of measuring many elements and isotopes from Li upto U down to sub ppb levels in favourable circumstances. The results would be presented and discussed. Furthermore, it is shown that how trace element measurement can give a cheap, quick and radiation free method for bone mineral assessment

Log: 414. **A COMPTON-VETOED GERMANIUM DETECTOR WITH INCREASED SENSITIVITY AT LOW ENERGIES.** Stephan Friedrich, Cameron R. Bates, Morgan T. Burks, Owen B. Drury, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA; David P. DiPrete, Savannah River National Laboratory, Aiken, SC 29808, USA.

The difficulty to directly detect plutonium in spent nuclear fuel due to the high Compton background of the fission products motivates the design of a Gamma detector with improved sensitivity at low energies. We have built such a detector by operating a thin high-purity Ge detector with a large scintillator Compton veto directly behind it. The Ge detector is thin to absorb just the low-energy Pu radiation of interest while minimizing Compton scattering of high-energy radiation from the fission products. The subsequent scintillator is large so that forward-scattered photons from the Ge detector interact in it at least once to provide an anti-coincidence veto for the Ge detector. For highest sensitivity, additional material in the line-of-sight is minimized, the radioactive sample is kept thin, and its radiation is collimated. We will discuss the instrument design, and show the performance of a prototype that employs two large CsI scintillator vetoes. Spectra of a thin Cs-137 reference source show a background suppression of a factor of ~2 around ~100 keV, limited by scattering in the collimator and a dead layer in the Ge detector. This is not yet sufficient to see weak Pu signals in spectra of fission product sources we have fabricated. However, Monte-Carlo simulations show that a background suppression by a factor of

~20 is possible with future instrument upgrades, as needed to directly detect Pu emissions for realistic spent nuclear fuel compositions.

Log: 415. **ACCELERATION AND AUTOMATION OF RADIOCHEMICAL METHODS FOR ACTINIDE ANALYSIS.** Grate, JW; O'Hara, MJ; Farawila, AF; Douglas, M; Maiti, TC; Petersen, SL. Pacific Northwest National Laboratory.

The isotopic analysis of actinides from complex matrixes such as environmental samples entails bringing the actinides into solution, separating them from the sample matrix, separating actinide elements from one another, and concentrating them for presentation as a source to a detector. Separations are required regardless of the detection method (radiometric or mass spectrometric) since the matrix and other actinides can interfere with detection. Conventional manual methods for these procedures can be both tedious and time-consuming to perform. We have developed a sequence of automated methods to perform at workstations in order to both accelerate the processes of radiochemical sample preparation, and to reduce the overall time to completion. Dissolution may be advantageously performed using microwave assisted methods with automated control of heat application and duration. We have developed and validated a procedure for total sediment dissolution. Matrix separations may be performed using column-based extraction chromatography methods rather than precipitations. Actinides are separated by anion exchange, also a column-based method. We have developed an automated workstation for column separations to process several samples in parallel, profoundly reducing labor and operator attention compared to gravity feed or vacuum box approaches. It has been designed with rigorous prevention of cross contamination. Finally, we have developed methods using acoustic streaming to accelerate single bead separations and preparation of beads as mass spectrometry sources. Examples and performance data will be presented.

Log: 416. **RE-DETERMINATION OF (K0,Q0)-VALUES FOR 10 ISOTOPES OF ANALYTICAL INTEREST.** F. Farina Arboccò^{1,2}, P. Vermaercke², K. Strijckmans¹, K. Smits²
1 Department of Analytical Chemistry - Ghent University; Krijgslaan 281 S12, BE-9000 Gent, Belgium; 2 k0-INAA Laboratory - SCK•CEN, Belgian Nuclear Research Centre; Boeretang 200, BE-2400 Mol, Belgium.

Most of the k0-NAA database has been reviewed and/or updated every decade since its launch in 1980. However, some of its values are nearly 31 years old. Sometimes these values were adopted from the nuclear literature at that time, or were experimentally determined by only 1 laboratory. This is the case, for instance, of: ⁴⁶Sc, ¹⁴⁰La, ¹⁴²Pr, ⁷⁶As and ⁶⁴Cu. Other isotopes are listed with accurate pairs of (k0,Q0)-values but were quoted as having (or possibly having) a Cadmium Transmission Factor (FCd) different from unity, such as: ¹⁸⁶Re, ¹⁸⁸Re, ¹⁸⁷W, ¹⁵³Sm and ¹⁹⁴Ir. An independent re-determination of these cases among others has been suggested by the k0-NAA community. In this work we aim at the experimental re-determination of the k0 and Q0 values for the aforementioned isotopes, through bare and Cd-covered irradiations of diluted Al-alloy standards. Self-shielding effects on these samples have been determined to be negligible or were otherwise accounted for when differing from unity. In order to satisfy the metrological level required for (k0,Q0)-determination, between 2 to 3 repeats were done on 2 irradiation channels (BR1 reactor; Mol - Belgium) with different neutron spectra parameters: $f = (38.6, 98.5)$; $\alpha = (0.06, 0.10)$. A software was developed comprising the latest official k0-NAA literature and formulae, allowing us to analyze the huge amounts of data-points arising from the activity ratios of up to 20 measurements per standard. The architecture of the software was oriented on reducing the chances of systematic errors from human input and data redundancy; with help of SQL constraints and Data-Relations. Our relative percentile differences to the literature values were $\leq 10\%$ for Q0's and $\leq 5\%$ for k0-values. The effect of new adopted k0-related nuclear data (Er, half-life) will be described.

Log: 417. **NEW MEASUREMENT OF THE $^{237}\text{U}(\text{n},\text{f})$ CROSS SECTION AT THE LEAD SLOWING-DOWN SPECTROMETER.** T.A. Bredeweg (LANL), M. Jandel (LANL), E.M. Bond (LANL), R.C. Haight (LANL), J.M. O'Donnell (LANL), D.J. Vieira (LANL), J.B. Wilhelmy (LANL), C.W. Alexander (ORNL), G. Belier (CEA-DAM).

Accurate neutron-induced reaction data are important to many issues in stockpile stewardship, nuclear reactor design and re-certification, nuclear non-proliferation and nuclear forensics. Of particular interest are the production and destruction reactions for all of the major and most of the minor actinides, including both neutron-induced capture and fission. Recently, we have performed a re-measurement of the neutron-induced fission cross section of ^{237}U ($t_{1/2} = 6.75$ days). This challenging experiment included irradiation of a pure ^{236}U sample in the HFIR reactor at ORNL followed by chemical separation to remove fission products and the ^{237}Np decay product. A ^{237}U target was prepared and installed in a LANL designed compensated ionization chamber which was used in a four day measurement at the Lead Slowing-Down Spectrometer (LSDS) located at the LANSCE WNR facility. Preliminary results from the measurement will be presented.

Log: 418. **"CLEAN ENERGY FROM SUGAR CANE BAGASSE: QUALITY EVALUATION BY NEUTRON ACTIVATION ANALYSIS".** C. Sbrana,¹ E.A.N. Fernandes,¹ J.C. Marson,¹ M.A. Bacchi,¹ G.A. Sarriés,² A.E.L. Reyes,² 1. Centro de Energia Nuclear na Agricultura, Universidade de São Paulo.

Ethanol is a major component of the Brazilian energy matrix, as important as gasoline, with the distinction of being an environmentally friendly fuel from renewable sources. The electricity from burning bagasse, a by-product of the extraction of juice from sugar cane, is the second most consumed in Brazil. Compared to the first, which comes from hydroelectric plants, it has advantages of not flooding large areas and therefore not producing methane from the huge amount of flooded organic matter. The burning of bagasse is cleaner than fossil fuels, due to reduced emissions of sulfur oxides, slow burning and low flame temperature which reduce formation of nitrous oxide. The process of cogeneration of electricity and steam production is strongly dependent on the conditions of the raw material, being the mineral impurities one of the main variables, which is basically dirty on the surface of sugar cane. These characteristics directly influence the calorific power of the bagasse, which is the energy released during complete combustion that will be burned to produce steam and electricity. This work used chemical tracers of soil in sugar cane such as Sc, Fe, Hf and Th, determined by instrument neutron activation analysis to identify the amount of mineral impurities present in sugar cane, correlating with the calorific power determined on the bagasse. The calorific power showed to be inversely proportional to the amount of impurities present in sugar cane, influencing directly the industrial efficiency.

Log: 419. **COMPARISON OF ACTIVE AND PASSIVE ENVIRONMENTAL SAMPLING FOR SAFEGUARDS APPLICATIONS.** Paula Cable-Dunlap (1), Lee Trowbridge (1), Debra Bostick (1), Denise Lee (1), Brian Anderson (1), Andrew Harter (1), Roger Kapsimalis (1), Lindsay Sexton (2), Jeff DeGange (2), Daniel Radford (2) ; 1) Oak Ridge National Laboratory 2) Savannah River National Laboratory.

Environmental sampling (ES) of surfaces using cotton swipes at nuclear facilities is one of the standard sampling protocols used by International Atomic Energy Agency (IAEA) inspectors to verify treaty compliance. Described herein is a series of studies undertaken to 1) survey a variety of low latent uranium content sampling media for efficacy of collecting nuclear materials and 2) compare active collection of particulates to swipe sampling. Findings from the evaluation of 40 natural and synthetic woven swipes will be presented. Results from a joint Oak Ridge (ORNL) and Savannah River National Laboratory (SRNL) project wherein a variety of passive deposition surfaces and active collections were examined for efficacy of sampling uranium processing effluents will also be detailed. For the swipe material evaluation work, the collection efficiency was evaluated on woven materials as-received and coated with complexants/solvents. The relative merits of each sampling matrix were reviewed on the basis of: 1) latent U background, 2)

ease of separating the material from the nuclear components, 3) presence of isobaric interferences from the swipe, and 4) efficiency in collecting uranium from surfaces. The second series of experiments evaluated the collection characteristics of active versus swipe sampling. ORNL and SRNL are collaborating on the development of the next generation of ES equipment for air grab and constant samples that could become an important addition to the international nuclear safeguards inspector's toolkit. Described herein are findings from the evaluation of collection efficiency of swipe sampling and active collection using an Aerosol Contaminant Extractor (ACE).

Log: 420. **ADVANCES IN MC-TIMS AND MC-ICP-MS FOR PRECISE AND ACCURATE U ISOTOPE MEASUREMENT**. Trinquier, A; Bouman, C; Lloyd, N.; Schwieters, J. Thermo Fisher Scientific, Bremen, Germany.

Accurate and precise uranium isotopic analysis in nuclear safeguards, nuclear fuel cycle industries and environmental monitoring is challenging because of the extreme range of relative U isotopic abundances, limitations in analyte quantities, applicable instrumental mass fractionation methods, and requirements for certified isotopic reference materials. Both the Thermo Scientific TRITON Plus MC-TIMS and NEPTUNE Plus MC-ICPMS offer new solutions to address these analytical challenges for nuclear industry applications, in particular: (1) A versatile detector array provides simultaneous measurement of minor isotopes on high-sensitivity and high-performance SEMs, and major isotopes on Faraday collectors (combined with in-run SEM-Faraday cross-calibration); (2) Different gain current-amplifiers (10e10, 10e11 and 10e12 Ohm) can be combined; (3) a new dual RPQ provides excellent abundance sensitivity (0.5 ppm at 1 amu); (4) The NEPTUNE Plus with Jet Interface combines a large capacity dry interface pump with a special set of sample and skimmer cones giving unparalleled sensitivity, with U ion yields > 3 % [1], yielding precisions and accuracies at the permil range for $^{234}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$, and the sub-permil range for $^{235}\text{U}/^{238}\text{U}$; (5) The TRITON Plus provides precise routine acquisition schemes for total evaporation (TE) and modified TE (MTE)2. The MTE2 allows for better instrumental mass fractionation corrections, in-run peak tailing correction (with or without dual RPQ), and precision and accuracy in the sub-permil and permil-range for $^{234}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$ (2se). [1] Bouman, C. et al., *Geochim. Cosmochim. Acta*. 73, 2009, 13, Suppl. 1. [2] Richter, S. et al., *J.Anal.At.Spectrom.*, 2011, 26, 550.

Log: 421. **STUDYING OF NOBLE GAS DIFFUSION THROUGH A POROUS MEDIA USING PROMPT GAMMA ACTIVATION ANALYSIS**. Carlos Rios Perez, The University of Texas at Austin; MR Deinert, The University of Texas at Austin.

Detection of noble gas isotopes in the atmosphere is the only measurement capable to confirm the nuclear nature of a well-contained underground nuclear explosion. A critical parameter in transport of these gases to the surface after a below ground test is the diffusion coefficient of the gas through the geological media. Prompt gamma activation analysis has been shown to be a technique capable of measuring the concentration of noble gasses in a sample. Here we demonstrate the suitability of this nuclear technique to measure the effective diffusivity of high purity argon in nitrogen within a uniform porous media. The experimental set-up used for this study consists of a 1 meter long by 10 cm diameter steel tube packed with 20/30 sieve SiO_2 sand. Prompt gamma activation analysis was used to measure the concentration of both argon and nitrogen within the experimental system. Experimental data were compared to the predictions of a numerical model to compute an effective diffusion coefficient of ?? argon.

Log: 422. **MECHANIZATION OF SUGARCANE HARVESTING: BETTER QUALITY OF RAW MATERIAL?**. Marson, JC (1); Fernandes, EAN (1); Sbrana, C (1); Bacchi, MA (1); Sarriés, GA (2); Reyes, AEL (2).(1) Centro de Energia Nuclear na Agricultura, Universidade de São Paulo (2) Escola Superior de Agricultura Luiz de Queiroz, Universidade de São Paulo

The efficiency of the processes involving the sugar-ethanol productive chain is key to ensuring competitiveness and profitability for the industry. In this context, the quality control of raw

materials delivered to the industry plays a fundamental role and has demanded incorporation of a set of improvements in harvesting operations. Today the Brazilian sugarcane harvesting is steadily changing from manual to mechanical procedure. In São Paulo, state, with 53% of the sugarcane cultivated area in Brazil, the mechanization increased from 49% in 2007/2008 crop to 65% in 2011/2012 crop. This change may influence the amount of impurities carried together with the stalks of sugarcane implying negative impacts like losses of sugar, low industrial efficiency and higher costs of transport and maintenance of equipments. Therefore, quantification of such impurities is an important tool in harvesting management. This work aimed at monitoring of the mineral impurities in sugarcane mechanically harvested, raw and burned while standing in the field, in different soil types and weather conditions. Samples were collected from 33 truckloads coming from two sugarcane fields with clay soil and sandy soil. Instrumental neutron activation analysis was used to determine soil tracer elements, mainly Fe, Hf, Th and Sc. The statistical test ANOVA showed significant differences ($p < 0.0001$) between clay soil and sandy soil, and raw and burned sugarcane. Nevertheless, the soil content of the truckloads varied from 0.9% to 19.9%, amounts similarly obtained in the manual harvesting.

Log: 423. **RADIOANALYTICAL METHODS FOR MONITORING AND RESEARCH INTO THE IMPACTS OF URANIUM MINING IN THE ALLIGATORS RIVERS REGION.** Peter Medley (Environmental Research Institute of the Supervising Scientist), Andreas Bollhoefer (Environmental Research Institute of the Supervising Scientist).

The Supervising Scientist Division (SSD) is a division of the Australian Federal Government Department of Sustainability, Environment, Water, Population and Communities (DSEWPAC). SSD undertakes a range of monitoring, inspection and research activities relating to impacts of uranium mining in the Alligator Rivers Region (ARR) of Northern Australia and provides high-level technical advice to projects relating to uranium mining elsewhere in Australia. Our research and monitoring programs assess the potential and actual effects on ecosystems and human health from the impacts of uranium mining. The ARR is located in the Wet-Dry tropics and covers an area of almost 30,000 km², with approximately two thirds of this area occupied by Kakadu National Park. The significance of Kakadu's heritage values has been recognised by the inclusion of the park as a World Heritage site, one of only 28 such sites across the world that is listed for both its natural and cultural values. The ARR is also home to significant mineral reserves, including uranium ore. In 1975, the Ranger Uranium Environmental Inquiry concluded that the hazards of mining and milling uranium, if properly regulated and controlled, were not sufficient to justify a decision not to develop Australian uranium mines. The report also recommended the establishment of a comprehensive system of environmental monitoring and research in the ARR, overseen by a Coordinating Committee representing all the agencies involved and chaired by a Supervising Scientist. This led to the establishment of SSD in 1978 under the Environment Protection (Alligator Rivers Regions) Act 1978. Within SSD the Environmental Radioactivity (EnRad) section operates a radioanalytical laboratory to support the monitoring and research activities of the Division. EnRad also provides commercial services in radioanalytical measurement to external clients. The radioanalytical laboratory specialises in low-level measurement of environmental radioactivity using alpha and gamma spectrometry, and more recently, liquid scintillation counting. In-house procedures have been developed for the measurement of uranium and thorium series isotopes ²¹⁰Pb, ²¹⁰Po, ²²⁶Ra, ²²⁸Th, ²³⁰Th, ²³²Th, ²³⁴U and ²³⁸U via alpha spectrometry and uranium and thorium series and ¹³⁷Cs, ⁷Be and ⁴⁰K via gamma spectrometry. The LSC instrument has been calibrated for gross alpha/beta determination in water samples and a method for ²¹⁰Pb is currently being developed. The laboratory handles a wide variety of samples, including surface and ground water, fish, mussels and other animal and plant material, and soils and sediments. The activity concentration of samples varies widely – over 5 orders of magnitude. Examples of projects we are involved in include (i) radionuclide uptake in bush foods for assessment and modelling of ingestion doses to the local Aboriginal population (ii) routine monitoring of radium in creeks and waterbodies downstream of the Ranger Uranium Mine (iii) assessment of areas impacted by past and present mining activities. This poster provides an overview of radioanalytical techniques and some environmental applications at SSD.

Log: 424. **NANOPARTICLES AND THIN FILMS FOR THE RAPID EXTRACTION AND ASSAY OF ALPHA-EMITTING RADIONUCLIDES AND RADIOIODINE FROM URINE.** Matthew J. O'Hara, Jennifer C. Carter, Wilaiwan Chouyyok, Jay A. MacLellan, Cynthia L. Warner, Marvin G. Warner, R. Shane Addleman.

In the event of an accidental or intentional release of radionuclides into a populated area, hundreds or possibly thousands of people would need to be evaluated for the intake (inhalation or ingestion) of radionuclides. Alpha-emitting radionuclides cannot typically be detected with the use of in vivo measurement techniques. Therefore, in vitro methods, likely urine radiobioassays, would need to be performed. However, radiological laboratories have little surge capacity to handle processing the number of samples that such an event would likely generate. At present, the quantitative determination of alpha-emitting radionuclides in bioassay samples (e.g., urine) is labor intensive, and requires significant sample preparation and analysis time, and consumes specialized expendables (e.g., chromatographic separation media). Researchers at the Pacific Northwest National Laboratory are investigating new approaches to radiobioassay measurements by developing and evaluating novel materials that can potentially streamline the radiobioassay process. Surface chemistries applied to nanoparticles and thin films can enable rapid chemical extraction of targeted radionuclides from bulk urine samples with little or no sample pretreatment, while simultaneously providing a means to create alpha counting sources quickly and with minimal labor. Results from the extraction of alpha-emitting radionuclides and radioiodine from water and urine samples will be presented, along with an estimation of the time requirements and analytical limits of detection of these methods.

Log: 425. **MONTE CARLO SIMULATIONS OF BACKGROUND CHARACTERISTICS OF HPGE DETECTORS IN THE OGOYA UNDERGROUND LABORATORY.** R. Breier (1), Y. Hamajima (2), P.P. Povinec (1). (1) Comenius University of Bratislava, Slovakia; (2) University of Kanazawa, Japan.

An increasing number of experiments have been devoted to the detection of very rare events, e.g. in double beta-decay, dark matter and neutrino experiments, as well as in environmental physics. Their common feature is a utilization of high sensitive low-level counting spectrometers, operating very often underground. For the background optimisation of a counting system it is useful to carry out Monte Carlo simulations, so the background characteristics can be estimated before constructing a counting system. The aim of our work has been a developing of a computing code that would allow to compute background components of low-level HPGe gamma-spectrometers operating in the Ogoya underground laboratory at the depth of 270 m w.e. The simulation code is based on the CERN's GEANT 4 package, with the Hangman's cosmic-ray shower library as a generator of cosmic-ray particle fluxes. Background characteristics of various HPGe (planar, coaxial and well) detectors will be presented and compared with simulations.

Log: 428. **INPUT OF I-129 INTO THE WESTERN PACIFIC OCEAN RESULTING FROM THE FUKUSHIMA NUCLEAR EVENT.** Tumey, SJ(1); Guilderson, TP(1,2); Broek, T(2); Brown, TA(1). (1) Center for Accelerator Mass Spectrometry - Lawrence Livermore National Laboratory, (2) Department of Ocean Sciences - University of California - Santa Cruz.

Iodine-129 is a long-lived radioisotope (15.7 Ma half-life) that is produced naturally by cosmic ray interactions with xenon in the atmosphere and as a fission product of uranium. A major fission fragment, I-129 is also produced by anthropogenic nuclear activities. While atmospheric nuclear weapons testing caused a significant increase in global I-129 levels, localized releases associated with nuclear fuel reprocessing have been observed in the environment, mainly concentrated in the oceans. The long radioactive half-life of I-129 coupled with the long marine residence time of iodine (340 kA) have enabled localized I-129 releases to be exploited as a conservative tracer of the movement and mixing of oceanic water bodies. Such studies have historically been focused on the Atlantic Ocean due to releases from the Sellafield and La Hague reprocessing plants. The Fukushima nuclear event in 2011 resulted in a significant release of I-

129 into the Pacific Ocean. This release presents an opportunity to use I-129 as an oceanographic tracer in the Pacific Ocean. Preliminary results of I-129 in the Pacific Ocean resulting from the Fukushima nuclear event will be presented. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 429. **MEASURING ADSORPTION COEFFICIENTS OF PLUTONIUM AT ENVIRONMENTALLY RELEVANT CONCENTRATIONS.** Tumey, SJ(1); Brown, TA(1); Martinelli, RE(1); Zhao, P(2); Begg, JDC(2); Kersting, AB(2); Zavarin, M(2); (1) Center for Accelerator Mass Spectrometry - Lawrence Livermore National Laboratory; (2) Glenn T. Seaborg Institute - Lawrence Livermore National Laboratory..

Plutonium (Pu) is a major contaminant of concern at several US Department of Energy sites. Remediation strategies require accurate models for the transport of Pu in the subsurface environment, but historically such models have underpredicted Pu migration rates when compared to field measurements. The deficiencies in transport models can primarily be attributed to an incomplete understanding of the complex geochemical behavior of Pu and its attachment to mobile colloids (< 1 μm particles) in groundwater. Models rely on sorption/desorption rates that are experimentally determined at high concentrations. The assumption that these rates are constant over such a wide range of environmental concentrations may not be valid. To test this hypothesis, we have conducted a suite of experiments to determine the adsorption coefficient of Pu at very-low, environmentally relevant concentrations (i.e., femtomolar to attomolar). Such low concentrations are now accessible using state-of-the-art mass spectrometric tools such as accelerator mass spectrometry. Here we will present initial results of experiments involving Pu sorption to goethite and montmorillonite. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 430. **INVESTIGATING THE NATURAL ABUNDANCE OF U-236 IN URANIUM ORE CONCENTRATES.** Tumey, SJ(1); Steier, P(2); Brown, TA(1); Hutcheon, ID(1); Martinelli, RE(1). (1) Lawrence Livermore National Laboratory; (2) University of Vienna.

In the nuclear fuel cycle, uranium ore is mined and milled to produce uranium ore concentrate (UOC) prior to conversion to uranium hexafluoride for enrichment. Its high uranium concentration and ease of transport has made UOC a fungible commodity traded freely on the worldwide market. Consequently, UOC is an attractive material for diversion from civilian power purposes to weapons production, and can be a leading indicator of proliferation. Nuclear forensic analysis can help identify the specific production location (mine, milling site) of UOC by examining chemical and physical signatures that characterize the geologic setting and depositional environment of the parent uranium ore, as well as the type of mine or milling process used in extracting uranium from the parent ore. Recently, U-236, which is produced naturally by neutron capture on U-235, has been measured in UOC samples from a number of different mines and shows potential utility for geolocation of parent uranium ore bodies. Here, we present further measurements of U-236 in UOC samples that will help assess the feasibility of U-236 as a nuclear forensic signature. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log: 431. **NEUTRON AND GAMMA RAY DOSES FROM A ^{252}Cf BRACHYTHERAPY SOURCE IN A WATER PHANTOM.** Ibrahim Al-Saihati (1), Naqvi A. A.(2). (1) Department of Electrical Engineering, (2) Department of Physics, King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia..

Monte Carlo study has been carried out to calculate neutron and gamma ray dose from an Isotron ^{252}Cf Brachytherapy sources in a Water Phantom. For dose calculation, the prompt fission gamma ray energy spectrum data of Cf-252 source was taken from Maienshein's measurements while the Cf-252 source neutron energy spectrum was modeled using isotropic Maxwellian

neutron energy spectrum. Neutrons and gamma ray axial and radial doses were calculated in the phantom body. Primary and secondary gamma rays due to source gamma rays and capture gamma rays was in the phantom body. As expected, within first 1.5 cm distance in the transverse direction, the neutron dose decreases rapidly. Then it further decreases gradually. Neutron dose due to the source casing thickness was also calculated for casing thickness varying from 0.1 mm to 0.25 mm but no significant increase in total dose due to casing dose was observed. The secondary gamma ray dose rate, calculated in the presented study, is very small compared to the primary gamma dose rate. Neutron and secondary gamma ray dose calculated in this study agree with the published data. Results of these study will be presented here.

Log: 432. **OPTIMIZATION AND ESTIMATION OF COLD NEUTRON PROMPT ACTIVATION ANALYSIS SYSTEM BY USING MONTE CARLO CODE.** S.M.T HOANG (1,2); J.H MOON (1); Y.S CHUNG (1); B.G PARK (3); G.M SUN (1). (1) KAERI, (2) KOREA University of Science and Technology, (3) Seoul National University.

Prompt gamma activation analysis (PGAA) from neutron capture is a nuclear analytical technique for non-destructive determination of elemental and isotopic compositions. By employing cold neutron beam guide, furthermore, greatly reduces the gamma-ray background at the PGAA sample while maintaining a neutron capture rate comparable to that of standard thermal neutron PGAA. A new Cold Neutron Induced Prompt Gamma Activation Analysis (CN-PGAA) system has been under constructed since April, 2009 at HANARO Cold Neutron Building (KAERI, South of KOREA). A Compton Suppression Factor of CN-PGAA system has been estimated about 8 with Co-60 radioactive source by using MCNPX simulation in this study. Several parameters of CN-PGAA system were studied to estimate and optimize a performance and arrangement of CN-PGAA system such as a scintillator material of annulus and back catcher guarded detector of Compton suppression spectrometers in CN-PGAA system, a distance from a target sample to n-type HPGe detector window, a relative position between n-type HPGe and guarded detector of Compton suppression spectrometers, etc. A detection limit of CN-PGAA system was also estimated with several configuration of this system. Additional, another Monte Carlo code, McStas, was adopted to predict an intensity and neutron flux of cold neutron beam guide that of a cold neutron source in CN-PGAA.

Keyword: PGAA, Compton suppression factor, MCNPX, McStas, Cold neutron.

Log: 433. **FISSION PRODUCT SEPARATION FROM SEAWATER BY ELECTROCOAGULATION METHOD.** T. Kitagaki(1)(2); T. Hoshino(2); Y. Sambommatsu(2); K. Yano(2); M. Takeuchi(2); T. Suzuki(1). (1) Tokyo Institute of Technology, (2) Japan Atomic Energy Agency.

In Fukushima daiichi nuclear power plant, seawater is urgently used to cool the reactor core. Therefore a large amount of seawater containing high levels of fission products (FP) is accumulated and its treatment is serious problem. In general, FP such as cesium, strontium and iodine in water are treated by passing the column filled with adsorbent such as zeolite or using chemical precipitation. However, because the adsorptions are dominated at the surface of adsorbent, the column using larger diameter adsorbent increases the amount of waste. Furthermore, chemical precipitation has complicated process and unmanageable wastes to use various chemical reagents. On the other hand, Electrocoagulation method which separates efficiently fine particles without addition of chemical reagent is expected to construct a separation system that can reduce the amount of waste and decrease adsorption time using fine particles of adsorbent. In this study, the powdered adsorbent such as zeolite; Mordenite, type A, silver zeolite, and ferrocyanide; potassium ferrocyanide, potassium nickel ferrocyanide, added to seawater containing simulated FP; cesium, strontium and iodine. After that Electrocoagulation with Al-Mg alloy electrode was done and aggregation promoter made by wood ash is added. As a result, various adsorbents are rapidly separated from liquid phase and separation efficiency of simulated FP is more than 90%. In addition, because this method doesn't use organics, treatment of the waste storage is easy. Thus, the applicability of the electrocoagulation method for rapid separation of FP from the seawater and ease waste treatment is confirmed.

Log: 434. **REMOVAL OF RADIOACTIVE CESIUM, STRONTIUM AND IODINE FROM NATURAL WATERS WITH BENTONITE, ZEOLITE AND ACTIVATED CARBON.** Kubota, T(1); Fukutani, S(1); Ohta, T(2); Mahara, Y(1). (1) Kyoto university, (2) Hokkaido university.

A large amount of radioactive material has been released by the accident of nuclear power plants at Fukushima, Japan. Concerns about additional release into the environment still remain according to the status of the power plants. We investigated the removal performance with bentonite, zeolite, and activated carbon from seawater and river water spiked with Cs-134, Sr-85, and I-131 produced at Kyoto University Research Reactor Institute. Radioactive strontium accidentally released from nuclear power plants is substantially Sr-89 and Sr-90; however, we used Sr-85 because of ease of isotope production and radioactivity measurement. Seawater was collected at Onahama coast, 56 km south from Fukushima Daiichi Nuclear Power Station, and river water was collected at Tone-Ohzeki, which is the diversion weir of a water treatment plant for Tokyo metropolitan district. The removal performance from seawater was inferior to that from river water. Results for bentonite showed more than 90% removal of Cs-134 while negligible performance to I-131. Although the removal performance of Sr-85 with bentonite was lower than that of Cs-134, precipitation of strontium with carbonate provided up to 90% removal of Sr-85. Results for zeolite showed the similar removal tendency to those for bentonite; however, the removal performance of these radioactive nuclides with zeolite was inferior to that with bentonite. Results for activated carbon showed the negligible removal performance of all these radionuclides. Redox reaction of iodide to iodine with NaClO slightly promoted adsorption of I-131 on activated carbon to attain 30% removal of I-131.

Log: 435. **REAL-TIME IMAGING OF NUTRIENT UPTAKE MANNER IN PLANTS USING CONVENTIONAL RADIOISOTOPES.** Nakanishi, TM; Knno, S; Kobayashi, NI; Hirose, A; Ishibashi, H; Tanoi, K Univ. of Tokyo.

We present real-time imaging systems developed for plant nutrient uptake. One is for macroscopic imaging targeting whole the whole plant, from root to up-ground part and the other is the microscopic imaging system to image mainly the root. In the case of the macroscopic imaging system, a fiber optic plate (5cm x 5cm) where CsI scintillator was deposited on one side of the surface was placed as close as possible to the plant. The radioactive tracer was supplied from the root and the radioactivity from the plant was monitored by a highly sensitive CCD camera. In the case of the microscopic imaging, a fluorescent microscope was modified so that three images, light, fluorescent and radiation images were taken at the same time. Using these systems, not only beta-ray emitters, such as P-32, Ca-45, some of the gamma-ray emitters was able to apply for the system. In the case of a rice plant, phosphate uptake manner was compared between soil culture and water culture. After several days of imaging, it was found that the phosphate was taken up much faster when cultured in water. In the case of soil culture, only the phosphate around the root was taken up by the plant. In the case of microscopic imaging system, phosphate uptake manner from the roots of Arabidopsis was also monitored and analyzed.

Log: 437. **RADIOACTIVE CESIUM UPTAKE BY AGRICULTURAL PRODUCTS AFTER NUCLEAR ACCIDENT IN FUKUSHIMA.** Nakanishi, TM Univ. of Tokyo.

After the nuclear power plant accident at Fukushima, several research groups were formed in Graduate School of Agricultural and Life Sciences, The University of Tokyo. Each group is consisted of several researchers with different specialties. For example, rice, soil, agricultural engineering researchers are discussing together to solve the radioactive uptake manner from soil to plants. The radioactive cesium was strongly adsorbed to the soil surface and the binding was found to be stronger with time. The radioactive cesium is very difficult to elute from the soil. Even with some chemicals, only 10 to 20% of the cesium was washed out from the soil with first treatment. However, from the second treatment, hardly any cesium was dissolved to the solution.

After two months from the accident, radioactivity was found in wheat leaves as spots, suggesting that radioactive cesium was firmly adsorbed on the leaves and only small amount was transferred to the other tissue. The only leaves, which were expanded at the time of the accident was highly radioactive but the radioactivity was drastically decreased in the leaves which were emerged after the accident. The fall out in the soil was not uniform but found as spots. When Cs uptake manner of a rice plant from soil and water was compared, it was found that the Cs in water solution was much easier to be taken up by the plant. The radioactivity found in the wide range of agricultural products will be presented.

Log: 438. **TIME-RESOLVED PROMPT GAMMA-RAY SPECTROMETRY FOR COINCIDENCE/ANTICOINCIDENCE EXPERIMENTS.** Bryan Tomlin and Rolf Zeisler, NIST, Analytical Chemistry Division.

Coincidence and anti-coincidence measurements in prompt gamma neutron activation analysis with pulsed neutron beams have been discussed as options for the improvement of detection sensitivities. Unfortunately the mechanical creation of neutron pulses with beam shutters leads to a significant decrease of beam intensity on the target; reduction ratios of more than 200 can be found in rotating slit shutters. To overcome this disadvantage a 1 to 1 or better ratio in neutron beam pulse length to beam off status would be beneficial. To achieve this efficiently the discrimination of coincidence (i.e., prompt) versus anti-coincidence (i.e., decay) gamma rays should be accomplished with time-resolved gamma-ray spectrometry. In contrast to gated spectra, the continuously acquired gamma ray spectrum allows to identify decay lines through the change of count rate during the beam-off intervals and thus the discrimination between prompt and decay emissions. Experiments for the demonstration of this principle were carried out at the NIST NCNR thermal PGAA facility. Element-specific spectra were collected with a time-resolved spectrometer while the change from beam-on to beam-off mode was simulated with a fast stationary shutter. The results show that this approach can improve PGAA yields in coincidence mode.

Log: 439. **GENERATION OF META IMAGES FOR VISUALIZATION OF THE DATA OBTAINED BY ANALYSIS OF THE TIME SERIES AUTORADIOGRAMS.** V. P. Kolotov (1), D. S. Grozdov (1), N. N. Dogadkin (1) and V.I.Korobkov (2). (1) Vernadsky Inst.of Geochem. & Analyt.Chem., (2) Moscow State University, Russia.

It has been shown that pixel-by pixel computer processing of series of autoradiography images allows to estimate the decay dynamics over the analyzed thin section surface. The results obtained while processing (apparent half-life data for each pixel) are stored in few internal two-dimensional arrays (raw data). The work undertaken deals with development of different ways how to represent these data in the form of meta images. Two approaches have been developed. The first one consists in generation of a set of cross-sections over the data arrays containing apparent half-life values. The pixels having half-life within the set limits are pointed by a color. The obtained meta images are useful for spatial analysis, but raw data sometimes are not enough selective. Additional treatment of the data has allowed detecting a useful feature to be used to increase the data selectivity. It has been found that for zones of the samples containing uniform distribution of radionuclides (for example, like in an inclusion) the frequency analysis of number of pixels in the zone on half-life value has normal distribution. This statement is not valid for background areas. This feature has been used for development of the second approach. A two dimensional filter for raw data processing has been developed. The application of the filter allows increasing significantly the contrast of the raw data while generation of the correspondent meta images. These meta images allow to detecting platinum group elements inclusions in the course of gamma-activation analysis of large size (up to 30 cm²) geological samples thin sections.

Log: 440. **SOFTWARE PACKAGE FOR RADIONUCLIDES MAPPING BY MEANS OF PROCESSING OF AUTORADIOGRAPHY IMAGES.** D. S. Grozdov, V. P. Kolotov, N. N. Dogadkin. Vernadsky Inst.of Geochem.&Analyt.Chem., Moscow, Russia.

A comprehensive software package has been developed for analysis of large size thin-sections by means of gamma-activation autoradiography. The task of the software is to support gamma-activation screening detection of micro inclusions of platinum group elements. The software consists in few internally connected parts: quantitative densitometry with using of slide scanners to digitize images obtained by nuclear photo detectors (conception of virtual scanner having linear response, linearization of detector response on fluens of beta-particles, consideration of efficiency registration on energy of beta particles); processing of series of autoradiogram images obtained during the sample cooling for estimation of the apparent half-life for each pixel; generation of meta images representing spatial distribution of the obtained half-life's (few options, including novel two dimensional filter for contrasting of image areas containing inclusions). The software package uses a lot of input parameters (list of scanners, their calibrations data, information on samples irradiation and images obtained for different time of sample cooling, etc.). To simplify the data control and it's transmission to the programs, the information is stored in multi table's relational database (MS Access) accessible by the software. Different examples of the obtained results are given in the presentation.

Log: 443. **GEOECOLOGICAL INVESTIGATIONS OF ARTIFICIAL RADIONUCLIDES IN THE AQUATIC SYSTEM OF THE OB RIVER AND KARA SEA.** Travkina, AV(1); Stoyanov, VV (2). (1) GEOKHI, (2) SEATECHIRM.

Data of the resent investigations on studying the radioecological situation in Ob river-Kara Sea aquatic system and adjacent part of the Kara Sea are presented. It has been obtained the levels of the main man-made radionuclide in water samples and sediments. The features of it's distribution have been shown. The results on the radionuclides concentration in water samples allowed us to propose that the suspended matter (nominally defined as $>0.45 \mu\text{m}$) can retain more than 90% ^{137}Cs whereas a smaller fraction ($\leq 31 \%$ in all cases) of ^{90}Sr is associated with suspended material. It means that the large fraction of radionuclides is tranported by river flow and enters in mixing zone in association with suspended matter. Features of horizontal distribution of radionuclides in different parts of Kara sea are closely connected with the presence of local sources of radioactive pollution in adjacent areas such as global fallout, local dumping of radioactive wastes from nuclear fuel reprocessing plants in Selafield and La Hague, transferring of radionuclides from Russian reprocessing plants in Siberia and so on. Radionuclides getting to Kara sea, are involved in cyclonic stream system and partially deposits in suitable places with natural geomorphologic or geochemical (an oxidizing condition in silts) traps.

Log: 444. **ELABORATION OF RADIOMETRIC ENZYME ASSAYS FOR IODOTHYRONINE DEIODINASES.** Pavelka, S (1,2). (1) Institute of Physiology, Czech Acad. Sci., Prague, (2) Central European Institute of Technology, Brno University of Technology, Czech Republic.

In the present studies, we developed novel, reliable methods for extremely sensitive radiometric determination of enzyme activities of iodothyronine deiodinases (IDs) of types 1, 2 and 3. These enzymes catalyze selective mono-deiodinations of iodothyronines and play crucial roles in the biotransformations of thyroid hormones. The newly developed radiometric assays for IDs were based on the use of appropriate high-specific-radioactivity ^{125}I -labeled iodothyronines as substrates; high-performance TLC separation of radioactive products from the unconsumed substrates; film-less autoradiography of radiochromatograms using storage phosphor screens; and quantification of the separated compounds with a BAS-5000 (Fujifilm Life Science Co.) laser scanner. For the proper measurement of the individual IDs, we found out first the optimum assay conditions, including the concentrations of the respective labeled substrates, appropriate concentrations of thiol cofactor, the amount of total protein and enzyme concentration in the incubation mixtures, and suitable incubation times. Further, we demonstrated the applicability of our sophisticated methods by following the alterations of IDs activities induced in cultured rat astroglial cells by a series of purinergic agonists, retinoic acid, and their combination. To characterize in more details the induction of each type of deiodinase by purines, we determined also time-course and dose-response curves using ATP as a representative of purinergic agonists. The developed methodology enabled us to determine IDs enzyme activities in microsomal

fractions of different rat and human tissues, as well as in homogenates of cultured mammalian cells, in the range as low as 10×10^{-18} katal. Financial support from Central European Institute of Technology and from the Czech Science Foundation (GA CR grant No. 304/08/0256) is acknowledged.

Log: 445. **AMS AND ICP-MS FOR DETERMINATION OF LONG-LIVED ENVIRONMENTAL RADIONUCLIDES.** Xiaolin Hou, Center for Nuclear Technology, Risø DTU, Technical University of Denmark.

Radionuclides are conventionally measured by detecting their characteristic radiation using alpha spectrometry, beta counting including liquid scintillation counting and gamma spectrometry depending on their decay modes. Accordingly these methods are high sensitive for short half-lived radionuclides. Mass spectrometry, typically used to measure isotopes of elements, can be also used for measurement of radionuclides. In these methods, the atoms of the radionuclide of interest are directly measured. Therefore, mass spectrometry methods are normally sensitive for the measurement of long-lived radionuclides. Among various inorganic mass spectrometric methods, inductively coupled plasma mass spectrometry (ICP-MS) and accelerator mass spectrometry (AMS) are two most popular used mass spectrometry techniques for the measurement of radionuclides, especially long-lived radionuclides. With the improvement of ICP-MS technique and more instruments to be installed, the application of this technique is becoming more popular tool for measurement of radionuclides. By hyphenation with automated separation system, ICP-MS will play a critical role in rapid determination of radionuclide for emergency analysis. AMS is the most sensitive analytical technique for many long-lived radionuclides, the new development of this techniques, especially the miniaturization of AMS system significantly reduce the cost of instrument as well as maintenance and operation, this stimulated and enhanced the application of this technique in the environmental researches. This work aims to discuss the application of ICP-MS and AMS in the measurement of some most important radionuclides, such as ^{99}Tc , ^{129}I , ^{236}U , ^{237}Np , ^{239}U , and ^{240}Pu , especially the new progress in the analytical methods of these radionuclides for environmental researches.

Log: 447. **EXPLORING BACKGROUND VARIABILITY OF A LOW-LEVEL (QUARTZ) GAS PROPORTIONAL COUNTER USING PULSE SHAPE AND DISTRIBUTION ANALYSIS.** George A. Klouda, National Institute of Standards and Technology.

Low-level gas proportional counting presents special challenges to the measurement process. Generally, measurement systems for this purpose have unique signal-processing capabilities through specialized electronics and software for maintaining a stable low background, thus achieving extremely low detection and quantification limits for the nuclide of interest. As a result, they are also more sensitive to small signals not germane to the sample which may affect the measurement if not identified. However, a common thread through these systems is often their ability to digitally capture and timestamp each event for retrospective analysis. The NIST low-level counting (llc) system serves as an example which was originally designed to measure atmospheric ^{37}Ar . In response to an event trigger, the system acquires its pulse timestamp, digital waveform, energy, and rise time. A monitor logs each event as either anti-coincidence or coincidence in nearly real-time, displays its digital waveform, and presents a 2-dimensional spectroscopic representation of rise time and energy of events as they occur. In post analysis software, other special capabilities include channel-channel interactions, low-level energy discrimination, pulse arrival time and inter-arrival time distribution analysis and the ability to browse waveforms and sort the event record. Thus, through individual pulse acquisition and analysis spurious events can easily be identified. Some examples of these unwanted events are related to ground loops, electrostatic charge, micro-discharges, counting gas purity and laboratory effects such as electromagnetic and mechanical influences. The discussion will briefly describe the capabilities of the NIST llc system, present some observations of events of known and unknown origin, and conclude with an assessment of uncertainty for measurements at the mBq level.

Log: 448. **RADIOPHARMACOLOGY: RELATIONS BETWEEN LEPTIN AND THYROID HORMONES METABOLISM IN MURINE WHITE ADIPOSE TISSUE.** Pavelka, S (1,2); Macek Jilkova, Z (3); Kopecky, J (3). (1) Department of Radiometry, Institute of Physiology, Czech Acad. Sci., (2) Central European Institute of Technology, Brno Technical University, (3) Department of Adipose Tissue Biology, Institute of Physiology, Czech Acad. Sci., Prague, Czech Republic.

The aims of the present studies were: to measure possible changes in activities of the key enzymes of thyroid hormones (TH) metabolism in murine white adipose tissue (WAT); and to describe the role of metabolic conversions of TH in WAT during obesogenic treatment of the mice and in response to caloric restriction or leptin administration. Adult male C57BL/6J mice were subjected to these three different treatment protocols. Subcutaneous and epididymal WAT and interscapular brown fat (BAT) depots were dissected and used for morphometric and enzymatic analyses. In addition, blood and samples of liver were also collected (and the latter used as controls for enzymatic determinations). Plasma levels of leptin, as well as total and free thyroxine (T4) and triiodothyronine (T3) concentrations were determined using RIA kits. Enzyme activities of iodothyronine deiodinases of the types 1 (D1), 2 (D2) and 3 (D3) in the liver, WAT and BAT were measured with the aid of our newly developed radiometric enzyme assays. We found that D1 enzyme activity in WAT was stimulated by a high-fat-diet feeding, which also increased plasma levels of leptin. However, D1 or D2 activities in BAT did not change. Caloric restriction decreased D1 activity in WAT, but not in the liver, and reduced leptin levels. In return, leptin injections increased D1 activity in WAT. In summary, our results demonstrate, for the first time, changes in D1 activity in WAT under the conditions of changing adiposity, and a stimulatory effect of leptin on D1 activity in WAT. Financial support from Central European Institute of Technology and from the Czech Science Foundation (GA CR grant No. 304/08/0256) is acknowledged.

Log: 449. **NATIONAL CENTER FOR NUCLEAR SECURITY (NCNS) - THE NUCLEAR FORENSIC PROJECT.** Amanda Klingensmith (NSTec), Ping Lee (NSTec), Dennis Barker (NSTec).

The National Center for Nuclear Security (NCNS) was established by Congress in response to the administration's "Écommitment to the pursuit of several treaties related to nuclear weapons, including the Comprehensive Test Ban Treaty and the START follow-on treaty." Integral to the success of these negotiations is "...the ability to verify complianceÉdemonstrating new technologies to detect the proliferation of weapons of mass destruction and to deal with them if they should cross our bordersÉ[and to] send an important signal to the international communityÉ [that the NNSS] will have a new focus on reducing the threat of weapons of mass destruction. (ref, Secretary of Energy letter to Congress, Sept, 21, 2009). In this new role, the NCNS, located at the NNSS, has three urgent and immediate tasks: 1.)Treaty verification technology development, test, and evaluation, 2.) Non-proliferation technology development, test and evaluation, 3.)Technical nuclear forensics in support of on-site inspection. NCNS is a diverse organization that reaches across disciplines in a collaborative effort to provide the most realistic environment available for the development and testing of proposed treaty verification as well as nonproliferation technologies. Under these guidelines exists the Nuclear Forensics Program, a multi-lab collaborative research effort to address the gaps in the field of nuclear forensics. This poster or presentation will address what NCNS is and what the Nuclear Forensics Program hopes to bring to the scientific arena.

Log: 450. **NATIONAL TECHNICAL NUCLEAR FORENSICS: AN OVERVIEW OF THE U.S. GOVERNMENT APPROACH.** Kentis Connelly, S (1); Dion, H (2); Kersting, AB (3). (1) U.S. Department of Homeland Security, (2) National Nuclear Security Administration, (3) Lawrence Livermore National Laboratory.

Technical nuclear forensics is the collection, analysis, and evaluation of pre-detonation (intact) and post-detonation (exploded) radiological or nuclear materials, devices, and debris, as well as the immediate effects created by a nuclear detonation. After 9/11, the U.S. Government (USG) recognized that the efforts to develop a unified nuclear forensics capability needed to be

enhanced and consolidated to ensure a more rapid advancement of capabilities. Over the past 10 years, the National Technical Nuclear Forensics (NTNF) program has become a top U.S. national security mission executed by the Departments of Defense, Energy, Homeland Security, Justice, State, and the Office of the Director of National Intelligence. Three key components of the USG program include: 1) Research & development to develop capabilities to interpret nuclear signatures to distinguish the origin of interdicted nuclear or other radioactive materials ; 2) International collaboration to develop nuclear forensics core capabilities and share best practices, lessons learned, and other information to help improve the ability of governments to identify smuggling networks and those responsible for illicit trafficking in nuclear or other radioactive materials and for planned and actual attacks using these materials or weapons; and 3) Expertise development to restore and sustain an enduring nuclear forensics workforce and ensure the USG is able to accomplish the NTNF mission from a human capital standpoint.

Log: 451. **MINIMUM DETECTABLE ACTIVITY, SYSTEMATIC UNCERTAINTIES, AND THE ISO 11929 STANDARD.** J.M. Kirkpatrick; R. Venkataraman; B.M. Young. Canberra Industries, Inc., 800 Research Parkway, Meriden CT 06450, USA.

The Currie formulation for Minimum Detectable Activity (MDA) has served for decades as the standard method for estimating radiological detection limits – it is simple and statistically defensible. It does, however, lack a means to account for the effects of systematic uncertainties. In recent years we have seen various efforts to incorporate systematic uncertainties into an MDA framework. Perhaps most notable of these is the recent ISO standard 11929 for the Determination of Characteristic Limits in Ionizing Radiation Measurements. This standard brings a Bayesian perspective to the problem of characteristic limits in radiation measurements that is in many ways both welcome and long overdue. In this paper, however, we note some apparent drawbacks to the ISO11929 approach. Namely, for reasonable values of the systematic uncertainty the correction it makes to the Currie MDA is negligible; while for large systematic uncertainties, calculated MDA values can become infinite. In between these two extremes, the user has little basis for evaluating the reliability of the result. To address these issues, we consider the problem from a new approach, developing a straightforward phenomenological statistical model of the MDA that treats systematic uncertainties explicitly. We compare its predictions with results of the ISO11929 formulation as well as the traditional Currie approach. Finally, some recommendations for alternative handling of the MDA in the face of significant statistical uncertainties are presented.

Log: 453. **DEVELOPMENT OF A RADIOCHEMICAL PROCEDURE DEDICATED TO ^{36}Cl MEASUREMENT IN STEEL NUCLEAR WASTE SAMPLE.** C. Hamon¹, P. Perret¹, F. Goutelard¹, R. Brennetot¹, I. Laszak¹, C. Andrieu² Commissariat à l'Energie Atomique et aux énergies alternatives, CEA Saclay, bâtiment 459, PC171, DEN/DANS/DPC/SEARS/LASE, 91191 Gif-sur-Yvette Cedex, France, ² Electricité de France, EDF – CIDEN / Département Etudes - Division Déconstruction/Groupe Inventaire et Agréments, 154 Avenue Thiers, CS 60018, 69458 LYON CEDEX 06 .

Within the French UNGG nuclear power plant dismantling framework, metallic radioactive wastes will be generated. The closer to the nuclear core, the higher the steel parts are activated and their content in ^{36}Cl becomes important. Because of its high mobility and long half-life, ^{36}Cl could have a major impact on ultimate disposal. In various steels, ^{36}Cl concentrations between 0.15 and 12.9 ppm have been reported [ref]. Depending on the neutron flux intensity, the ^{36}Cl activity calculated for a 1ppm concentration of stable chlorine varies from 0.03 to 91Bq/g, whereas the upper acceptance limit is 5Bq/g for medium activity wastes. Currently, 200 steels samples have already been collected prior to dismantling. As no data were available on the stable chlorine content in these materials, a tailored radiochemical procedure was developed to measure the ^{36}Cl . It allows for the analysis of a great number of samples together with a detection limit well below 5Bq/g. A nitric / hydrochloric acid mixture was optimized for a complete dissolution of stainless steel samples while minimizing the amount of chlorine added. Since 25mmoles of chlorine are still required to dissolve 0.2g of stainless steel, the following separation steps were adapted.

Ultimately, the achieved radiochemical protocol allows, within its first step, to sort the samples according to their activity in less than one day. For ^{36}Cl activity higher than 3Bq/g, the whole protocol is carried out in only three days providing a pure ^{36}Cl fraction. Results on activated stainless steels will be presented.

Log: 454. **ARE RADIOCHEMISTS READY TO USE LAB ON CHIP?** C. Mariet, F. Goutelard
Commissariat à l'Energie Atomique et aux énergies alternatives, CEA Saclay, bâtiment 459,
PC171, DEN/DANS/DPC/SEARS 91191 Gif-sur-Yvette Cedex, France.

Microfluidics deal with the behavior and manipulation of fluids that are geometrically constrained into small, typically sub-millimeter scale. It also includes the technology to develop and manufacture lab-on-a-chip (LOC) defined as a device that integrates one or several laboratory functions on a single chip of few square centimeters in size. Over the past few years, microfluidics devices have enjoyed success in certain niche applications, notably in pharmaceuticals, biotechnology and life sciences. However, recent advances and innovations could make the technology ubiquitous. The advantages of miniaturization include lower sample/reagent consumption, a reduced risk of contamination, a lower per unit cost, lower power consumption, and higher functionality due to automation and portability. Reduction of the dose to the manipulator can be added into this list in radiochemical analysis. Nevertheless, only few applications in the analysis of nuclear material have been reported. The aim of this paper is to explore the reason why, despite all the advantages, it takes so long to apply LOC in the nuclear analysis and to underline the difficulties and identify the key solutions. First, the scaling effect on fluid, mass, heat, electric charge transfers and on chemical reactions will be described. The microfabrication techniques and existing functional microfluidic devices will be presented. Several separation techniques (Precipitation, Solid phase extraction, Chromatography ...) and chemical reactions have been developed for radionuclide measurements in lab scale. Finally, an evaluation of the work that needs to be carried out to implement the micro device in nuclear applications will be presented.

Log: 455. **INTEREST OF METHACRYLATE ANION-EXCHANGE MONOLITHS FOR RADIOCHEMICAL MINIATURIZED ANALYSIS.** Anthony Bruchet(1,2), Velan Taniga(3), Clarisse Mariet(1), Vincent Dugas(2), Florence Goutelard(1), Jérôme Randon(2), Laurent Malaquin(3). (1.)Commissariat à l'Energie Atomique, Saclay, DEN/DPC/SEARS/LANIE, Gif-sur-Yvette, France, (2.)Laboratoire des Sciences Analytiques (UMR CNRS 5180), Université de Lyon, Villeurbanne, France, (3.)Laboratoire des Macromolécules et Microsystèmes en Biologie et Médecine (MMBM) (Institut Curie, Paris, France).

Since their introduction in the early 1990s polymethacrylate monoliths have emerged as a powerful alternative for microscale separations or sample treatment. Their relatively simple implementation in columns with small internal diameters makes them particularly attractive for the new chromatographic challenges of complex matrices analysis and on-chip separations. Despite their relatively poor ion-exchange capacity due to their highly porous structure, their use as anion exchangers is of large interest for nuclear analyses as numerous separations are based on this process. A photopolymerized poly(glycidyl methacrylate-co-ethylene dimethacrylate) monolith has been optimized to improve its chromatographic performances especially regarding IEC[1,2], reaching a value of 300 $\mu\text{mol/g}$. The procedure initially developed in fused silica capillaries has been successfully implemented in cyclic olefin copolymer (COC) micro-devices. Synthesis and chromatographic results obtained with this monolith in COC micro-devices will be presented and discussed in terms of reproducibility and robustness. Application to analysis of nuclear material will be presented.[1] Synthesis and Characterization of Ammonium Functionalized Porous Poly(glycidyl methacrylate-co-ethylene dimethacrylate) Monoliths for Microscale Analysis and Its Application to DNA Purification A. Bruchet et al., Journal of Biomedical Nanotechnology Vol. 7, 1–11, 2011 [2] Improving chromatographic performances of glycidyl methacrylate anion-exchange monolith for fast nano-Ion Exchange Chromatography. A. Bruchet et al. Journal of Separation Science. Vol 34 (16-17) 2079-2089, 2011.

Log: 456. **ON THE NATURE OF RADIOIODINE RELEASED FROM MEDICAL ISOTOPE PRODUCTION.** Engelmann, MD (1); Wahl, JH (1), Doll, C (1), Hoffman, E (2). (1) Pacific Northwest National Lab, (2) Australian Nuclear Science and Technology Organisation..

Observed radioiodine (I-131) releases from medical isotope production facilities occur both as a relatively continuous low-level bleed of I-131 activity, and occasional spikes of considerably higher radioactivity that typically correspond to specific process events from within the facility. Studies show that much of this activity is not efficiently trapped on activated charcoal. It has been suggested frequently that the chemical species of released radioiodine are highly volatile organic forms, but there has been little done to back this assertion with specific data. Recently, we have conducted solvent extraction experiments to target the organic fraction of I-131 released from a medical isotope production facility. These studies indicate that roughly 10% of the released I-131 activity appears to be organic. Understanding the chemical forms of iodine released will help to improve abatement system technology.

Log: 457. **THERMAL NEUTRON BEAM FACILITY AT GEORGIA TECH.** Eric A. Burgett* and Nolan Hertel, Georgia Institute of Technology (* Presently at Idaho State University).

A thermal neutron beam line has been created at the Georgia Tech using a graphite pile. The pile is constructed of nuclear grade graphite and is 1.524 m by 1.524 m by 2.438 m. To cut down on thermal neutron background, the pile is wrapped on all sides by a layer of boral sheeting with an opening to allow out thermal neutrons in one sheet. This boral wrap effectively cuts off most of the thermal neutrons diffusing out from other locations on the pile surface. The neutrons can be provided by a number of sources, but the principal source is a 1.998 TBq $^{241}\text{Am-Be}$ (α, n) source. The movable stringers of which the pile is constructed allow for the $^{241}\text{Am-Be}$ to be located in different positions in the pile, thereby allowing for the generation of beams with different proportions of fast and thermal neutrons if desired. The beam exit is essentially a hole cut in the boral shielding to allow testing apparatuses to be coupled to the pile. The beam was originally designed to test boral samples, but has more recently been used to characterize thermal neutron detector performance. With the source in its "reference" location, simple cadmium cutoff measurements have yielded a cadmium ratio of approximately 22 for the beam. Monte Carlo simulations have been performed to assist in the more complete characterization of the system.

Log: 458. **ASSESSING INTERNAL CONTAMINATION OF INHALED FISSION PRODUCTS.** Emily Freibert, Georgia Institute of Technology; Nolan E. Hertel, Georgia Institute of Technology; Armin Ansari, Radiation Studies Branch, The Centers for Disease Control and Prevention.

In the event of a nuclear power plant accident, fission products can be released into the atmosphere and inhaled by people. In order to rapidly triage the large number of people potentially impacted, a detection system is needed that can acquire such data quickly to approximate internal contamination levels and determine if an individual needs further screening. A commercially available portal monitor using plastic scintillators was investigated as a tool to assess internal contamination levels for fission products deemed to be principally of concern. The portal monitor count rates for 1Bq loading of each fission product in each organ of the body were computed using MCNP, a previously validated model of the portal monitor, and two MIRD-based anthropomorphic phantoms, a child and an adipose male. Biokinetic data from the Dose and Risk Calculation software (DCAL) were used to determine radionuclide concentrations in each organ as a function of time. For each phantom type, these data were folded together to determine the count rates associated with inhalation of each individual gamma-emitting fission product that would lead to a committed effective dose of 10 mSv.

Log: 459. **RADIOMETRIC DETECTOR OPTIONS TO AID IN DOE HIGH ACTIVITY WASTE TANK IN-SITU CHARACTERIZATION EFFORTS.** DiPrete, DP (1); DiPrete CC (1); McMahon KJ (1); Malek MA (1); Couture AH (1). (1) Savannah River National Laboratory.

Waste cleanup efforts currently underway at the Savannah River Site have created a need to characterize the inventories of the various waste tank heels prior to tank closure. Occasionally, results from laboratory analyses indicate alternative sub-sampling strategies are needed, resulting in repetitive efforts to sample and analyze tank bottoms. Ideally, in-situ tank analyses using a radiometric probe capable of being lowered into a waste tank could aid in identifying waste structures on tank bottoms requiring further sampling and characterization. Ideally, the probe would provide information determining which structures were higher in concentrations of actinides and fission products characteristic of DOE high level waste (HLW) heels. Although options are limited for what isotopes can be measured directly without extensive radiochemical speciations, the low energy photon spectra of HLW does offer some intriguing possibilities for what could be characterized from a radiometric probe designed to look at that energy spectrum. One possibility would be to design a probe centered on some recently developed technology from AMPTEK Inc. Such a detector would be relatively insensitive to the high photon background that would paralyze conventional gamma probes (i.e. NaI) that have typically been used to measure radioactive emissions in the tanks in the past. The detector is capable of making high resolution measurements at very high count rates (in excess of 500,000 cps). An overview of measurements made on various HLW samples using planar and semi-planar HPGe detectors, detectors based on AMPTEK pin diode technologies, as well as some additional detector technologies will be discussed.

Log: 460. **A TECHNICAL NUCLEAR FORENSICS GRADUATE FOCUS: WHAT IT REQUIRES.** J. Petrosky(1), J. McClory (1), A. Bickley(1) (1) AFIT Engineering Physics Department.

The need for technical nuclear forensics experts is needed as part of the nation's holistic approach to nuclear deterrence. The rapid advance of potential portable and sensitive forensics tools, and the concern over an aging nuclear expert population, provide both an opportunity and urgency to the development of a focused nuclear forensics curriculum. Owing to the substantial focus on research and technical science and engineering applications required for pre and post detonation analysis, a graduate level of education with a broad physics, nuclear/radiological engineering, and radiochemistry content is required. This presentation will focus on the authors' present efforts to develop a curriculum within a unique nuclear engineering program that supports the development of graduate level technical nuclear forensics experts and prepares them to fill technical positions as researchers and program managers in key government organizations and laboratories. Primary components of such a program would include an understanding of nuclear weapons effects, measurement techniques, radiochemistry, and statistical analysis; methods for ensuring graduates have an understanding of the nuclear forensics and attribution organizational structures; and validity and recommendation of internships and research assistantships is explored. Current texts will be reviewed with an emphasis on the current technical nuclear forensics information available. In resident, distance learning, and mixed modality methods are presented and analyzed.

Log: 462. **DETERMINING UNCERTAINTY FOR NUCLEAR FORENSICS AND OTHER DATING MEASUREMENTS.** Simon Jerome, National Physical Laboratory, Teddington, United Kingdom.

Assessment of uncertainties is an essential part of any measurement, especially when the use of such data may have wide ranging outcomes, and this especially so for measurements made in support of nuclear forensics measurements. However, the assessment of uncertainties for age dating is complex due to the convoluted nature of the calculations that lead from an isotope ratio measurement to an assessment of the age of the material so measured. Such isotope ratios are based on the ingrowth of a daughter nuclide, be it stable or radioactive, arising from the decay of a radioactive parent radionuclide. In some cases, notably the decay series ^{237}Np - ^{233}Pa - ^{233}U and ^{243}Am - ^{239}Np - ^{239}Pu , the ratio between the parent radionuclide and the granddaughter radionuclide is the measurement of interest. Despite this, it is possible to solve the expression explicitly, although the expressions involved are quite complex and the relationship between the component

uncertainties is not a simple linear one, and the derivation sensitivity coefficients is not necessarily straightforward nor helpful. In this paper the relevant component terms of the uncertainty budget of a parent-daughter pair are derived and an examination of their relative contribution to the overall uncertainty budget are assessed. Finally, this analysis is used to examine some of the more important parent-daughter pairs in age dating.

Log: 463. **IMPROVED ACTINIDE TIMS ION SOURCE.** Matthew G. Watrous, Idaho National Laboratory.

A procedure has been developed to improve the ionization efficiency for uranium to be measured by thermal ionization mass spectrometry (TIMS). A new type of porous refractory material has been developed as a thermal ionization emitter that is an improvement over both direct filament and resin bead loading. Application of the porous ion emitter to uranium is demonstrated to provide a utilization efficiency ranging between 1% to 2% across a sample size range of 0.2 - 10 pg, better than that achieved from resin beads and much better than that achieved with direct loading onto a filament. It has been shown to produce more ions detected from the same amount of starting material in the source than the resin bead method for uranium. It has shown similar performance to the resin bead method for the production of ions of plutonium.

Log: 464. **PROGRESS TOWARDS THE PRODUCTION OF NEPTUNIUM-236G.** Simon Jerome and Cyrus Larijani.

The radiochemical analysis of ^{237}Np is important in a number of fields of activity, such as nuclear forensics, environmental analysis and measurements throughout the nuclear fuel cycle. However analysis is complicated by the lack of a stable isotope of neptunium. Although various tracers have been used, including ^{235}Np , ^{239}Np and even ^{236}Pu , none are entirely satisfactory. However, ^{236}gNp would be a better candidate for a neptunium yield tracer, as its long half-life – 1.55×10^5 years – means that it is useable as both a radiometric and mass spectrometric measurements. This radionuclide is notoriously difficult to prepare, and limited in scope. In this paper, we examine the options for the production of ^{236}gNp , based on work carried out at NPL since the last MARC conference. However, this work was primarily aimed at the production of ^{236}Pu , and not ^{236}gNp and therefore the rate of production are based on the levels of ^{236}Pu generated in the irradiation of (i) ^{238}U with protons, (ii) ^{235}U with deuterons, (iii) ^{236}U with protons and (iv) ^{236}U with deuterons. The derivation of a well-defined cross section is complicated by the relevant paucity of information on the variation of the ^{236}mNp : ^{236}gNp production ratio with incident particle energy. Furthermore, information on the purity of ^{236}gNp so produced is similarly sparse. Accordingly, the existing data is assessed and a plan for future work is presented.

Log: 465. **A METHOD FOR ANALYSIS OF NEUTRON DEPTH PROFILING SPECTRA OF HANARO CN-NDP SYSTEM.** B.G. Park (1); H.D. Choi (1); G.M. Sun (2). (1) Seoul National University (2) Korea Atomic Energy Research Institute.

A Cold Neutron Depth Profiling (CN-NDP) system is being developed in Cold Neutron Activation Station (CONAS) which is being constructed at the HANARO research reactor of Korea Atomic Energy Research Institute (KAERI). For the NDP applications, the process of conversion energy spectrum of charged particle to depth profile of interested isotopes is required. In this study, the principle of the relation between the energy spectrum and the depth profile is presented for the case of energy broadening and the case without any energy broadening. Estimating the typical spectral unfolding or inversion algorithms for CN-NDP system, NIST SRM-2137 test spectrum has been calculated by using the TRIM code and numerical method. And the analysis software for charged particle energy spectrum has been developed for the simple case. The main routine of the analysis code was developed in MATLAB language to provide a graphical user interface and to run in the MS-Windows system.

Log: 467. **RADIOMETRIC ENZYME ASSAYS UTILIZING [125I]-IODOTHYRONINES AS SUBSTRATES.** Pavelka, S (1,2). (1) Institute of Physiology, Czech Acad. Sci., Prague, (2) Central European Institute of Technology, Brno University of Technology, Czech Republic.

Employing our newly developed radiometric assays for iodothyronine deiodinases (IDs) of types 1, 2 and 3 (D1, D2 and D3), as well as adapted radiometric enzyme assays for iodothyronine sulfotransferases (ST) and uridine 5'-diphospho-glucuronyltransferase (UDP-GT), we studied the interaction of an antidepressant drug fluoxetine (Fluox) with the metabolism of thyroid hormones (TH) in the rat. All of the mentioned enzymes play crucial roles in the biotransformation of TH in peripheral tissues. For example, inadequate activities of brain IDs that could lead to local insufficient concentration of 3,3',5-triiodo-L-thyronine (T3), might be one of the pathogenic factors of depression. Appropriate 125I-labeled iodothyronines of high specific radioactivity were used as specific substrates in all of the described enzyme assays. Applying these radiometric assays, we found profound changes in IDs activities, especially in the pituitary and cerebellum. The treatment of rats with Fluox alone caused a moderate increase in D2 and, in turn, a slight decrease in D3 activities in cerebellum and some other regions of the CNS. No significant changes in D1 activity were detected. On the other hand, the administration of T3 alone caused, in accordance with our expectation, a substantial decrease in pituitary D2 activity and a simultaneous increase in D1 and D3 activities practically in all tissues studied. The measurements of ST enzyme activities in liver and kidney cytosolic fractions did not demonstrate any significant effects of Fluox administration on the induction of these enzymes. In contrast, in samples of liver microsomes of rats treated with fluoxetine, we found about two-fold higher UDP-GT activities in comparison with control rats. Financial support from Central European Institute of Technology and from the Czech Science Foundation (GA CR grant No. 304/08/0256) is acknowledged.

Log: 468. **RADIOANALYTICAL METHODS FOR FOLLOWING DEVELOPMENT OF DIET-INDUCED OBESITY.** Pavelka, S (1,2). (1) Institute of Physiology, Czech Acad. Sci., Prague, (2) Central European Institute of Technology, Brno University of Technology, Czech Republic.

With the use of advanced radioanalytical methods, we measured changes in the enzyme activity of the type 3 iodothyronine deiodinase (D3) in murine white adipose tissue during development of dietary obesity. Previous studies have established that thyroid hormones (TH) play important roles in the development and function of both brown and white adipose tissue (WAT). However, data about local transformations of TH in WAT are still scarce. High-fat-feeding induction of obesity was followed in male C57BL/6J mice maintained on a special high-fat (HF) diet for 2 weeks before analysis, in comparison with the same animals maintained on a standard low-fat (LF) diet. D3 activity in WAT was measured with the aid of our newly developed radiometric enzyme assay. Serum total thyroxine (T4) and triiodo-thyronine (T3) concentrations were determined using RIA kits. HF-diet feeding resulted in an increased size of adipocytes and in a significantly higher weight of both epididymal-visceral and dorsolumbar-subcutaneous fat depots. Total T4 and total T3 plasma levels were significantly elevated in mice fed HF-diet, in comparison with mice maintained on LF-diet. Development of HF-diet-induced obesity in the mice was associated with an enhancement of D3 activity in WAT, especially in subcutaneous fat depots. In conclusion, HF-diet-induced obesity in mice caused increased turnover of TH in WAT. Financial support from Central European Institute of Technology and from the Czech Science Foundation (GA CR grant No. 304/08/0256) is acknowledged.

Log: 469. **EVALUATION OF $\text{LaCl}_3\text{:Ce}$ AND $\text{LaBr}_3\text{:Ce}$ PROBES FOR GAMMA-RAY SPECTROSCOPY AND DOSIMETRY MEASUREMENTS USING MONTE CARLO SIMULATIONS.** K.S. Alzimami^{1, 3}, K. G. Alsafi², A. A. Alfuraih^{1,3}, M. Alkhorayef¹, A. A. Alghamdi⁴, A. Ma⁴, and N.M. Spyrou^{1, 3}: ¹Department of Radiological Sciences, King Saud University, Kingdom of Saudi Arabia, ²Department of Physics, King Abdulaziz University, Kingdom of Saudi Arabia, ³Department of Physics, University of Surrey, Guildford, Surrey, UK, ⁴Department of Radiological Sciences, Dammam University, Kingdom of Saudi Arabia.

Cerium-doped lanthanum crystals, particularly $\text{LaBr}_3\text{:Ce}$, have lately drawn significant interest due to their high scintillation yield, superior energy resolution and fast decay time which make them attractive for gamma-ray spectroscopy compared to NaI(Tl) . The main objective of this work is the investigation of performance of relatively new commercial $\text{LaBr}_3\text{:Ce}$ and $\text{LaCl}_3\text{:Ce}$ probes for gamma-ray spectroscopy and dosimetry measurements. The crystals were irradiated by a wide range of energies (^{57}Co , ^{22}Na , ^{18}F , ^{137}Cs and ^{60}Co). The study involved recording of detected spectra and measurement of energy resolution, photopeak efficiency, internal radioactivity measurements as well as dose rate. The Monte Carlo package, Geant4 Application for Tomographic Emission (GATE) was used to validate the experiments. Overall results showed very good agreement between the measurements and the simulations. The $\text{LaBr}_3\text{:Ce}$ crystal has excellent energy resolution, energy resolutions of $(3.05 \pm 0.03)\%$, $(3.25 \pm 0.03)\%$ and $(6.25 \pm 0.03)\%$ were achieved for $\text{LaBr}_3\text{:Ce}$, $\text{LaCl}_3\text{:Ce}$ and NaI(Tl) respectively. The disadvantage of the Lanthanum halide scintillators is their internal radioactivity. The internal radioactivity is due to naturally occurring radioisotopes ^{138}La and ^{227}Ac . Nevertheless, the internal radioactivity level is acceptable (2.15 to 6.45 Bq of ^{138}La) and this drawback is likely to be serious only for the very long time of counting applications. Our results and experience with GATE demonstrate the accuracy and flexibility of GATE for dose calculations. Gamma probe with $\text{LaBr}_3\text{:Ce}$ scintillator has shown an accurate and quick dose measurements at PET Units which allows accurate assessment of the radiation dose received by staff members compared to the use of EPD.

Log: 470. **OPTIMIZATION OF ZR-89 PRODUCTION USING MONTE CARLO SIMULATIONS.** Alfuraih, A(1); Alzimami, K(1); Ma, A(2); Alghamdi, A(2). (1) King Saud University (2) Dammam University.

Positron emitter Zr-98 is gaining more momentum as an effective radionuclide for developing new immuno-PET agents for in-vivo oncology imaging and monoclonal Antibody (mAb) quantification. Zr-98 disintegrates through positron decay (22.7%) and through electron capture (73.3%) with a half-life of 78.4 h. This half-life and the sufficient high abundance of positrons makes Zr-98 well suited for antibody labeling. In the production of Zr-89, a proton beam is focused to bombard an Yttrium-89 foil target. Neutrons are produced as well as radioactive Zr-89. Optimization of primary beam energy enables maximum production of Zr-89 and minimizes other competing reactions such as the formation of Zr-88 and Y-88. With new developments of hospital based medical cyclotrons the production of Zr-89 can be obtained at medical cyclotron proton energy range by employing Yttrium-89 target. Accurate modeling of the proton beam and the target is an essential step assuring optimal beam energy and target specifications and for further radiopharmaceutical labeling process. In this study MCNPX Monte Carlo code is utilized to model proton interactions by simulating different proton energies to reach the optimal production specification. The simulation results provide a base for the experimental work assessment.

Log: 471. **URANIUM IN NIST SRM GLASS BY FEMTOSECOND LASER ABLATION MC-ICP/MS.** Andrew M. Duffin, Garret L. Hart, and Gregory C. Eiden, Pacific Northwest National Laboratory.

We employed femtosecond Laser Ablation Multicollector Inductively Coupled Mass Spectrometry for the determination of uranium isotope ratios in a series of standard reference material glasses (NIST 610, 612, 614, and 616). This uranium concentration in this series of SRM glasses is a combination of isotopically natural uranium in the materials used to make the glass matrix and isotopically depleted uranium added to increase the uranium elemental concentration across the

series. Results for NIST 610 are in excellent agreement with literature values. However, other than atom percent ^{235}U , little information is available for the remaining glasses. We present atom percent and isotope ratios for ^{234}U , ^{235}U , ^{236}U , and ^{238}U for all four glasses. Our results show deviations from the certificate values for the atom percent ^{235}U , indicating the need for further examination of the uranium isotopes in NIST 610-616. Our results are fully consistent with a two isotopic component mixing between the depleted uranium spike and natural uranium in the bulk glass.

Log: 472. ELECTROCHEMICALLY MODULATED SEPARATIONS FOR RAPID AND SENSITIVE ISOTOPIC ANALYSES. Liezers M(1); Hart, GL(2); Duckworth, DC(3).

The desire for more rapid and sensitive analytical determinations of actinides in the environment is being addressed in our laboratory through the development of Electrochemically Modulated Separations (EMS) coupled to Multiple Ion Counting Inductively Coupled Plasma Mass Spectrometry (MIC-ICP-MS). This work is focused on the characterization of EMS separations of U and Pu from swipe materials. EMS provides electrochemically controlled surface complexation of either U(IV) or Pu(IV), depending on the potential applied to an anodized glassy carbon electrode. The accumulation and subsequent rinse results in >90% recovery efficiency, matrix elimination, and concentration of the selected actinide. All this is performed solely under the control of applied potential. Once isolated, the concentrated actinide is released into a ~1 microliter volume of dilute nitric acid, resulting in a volumetrically concentrated transient signal that is ideal for multiple ion counting detection. Coupled with ThermoScientific's Neptune Plus MIC-ICP-MS, atom utilization efficiencies are nominally 1%, comparable to the gold standard thermal ionization mass spectrometry, but requiring only 20 minutes for separation and analysis. Methods, analytical figures of merit, and advances in multiple electrode (i.e., actinide) separations will be highlighted.

Log: 473. EVALUATION OF SOLUTION CONDITIONS ON EXTRACTION EFFICIENCY FOR POLYMER LIGAND FILMS. Dominic S. Peterson, Edward R. Gonzales, Crystal L. Tulley-Cordova, Claudine E. Armenta, Hakim Boukhalfa - Los Alamos National Laboratory.

In recent years, we have described the use of a method to combine radiochemical sample preparation and analyte separation to provide for a rapid method of sample preparation. In this method, extractive ligands are dispersed in a polymer matrix and placed on a surface. These polymer ligand films can be produced by immobilizing the polymer on a surface, filter, or as a thin membrane; alternatively, a polymer can also be synthesized around the extractive ligand to produce a cross-linked polymer structure. The polymer ligand film surface is then amenable to direct analysis by alpha spectrometry. These approaches offer fast uptake kinetics, high material loading, and good selectivity. We have demonstrated these systems for the rapid uptake of plutonium and americium from solution and their subsequent analysis by alpha spectrometry, with no additional sample preparation required. In this presentation, we will discuss a variety of sample parameters that affect the extraction of radionuclides, including pH, temperature, concentration, and Oxidation state of the radionuclide. We will also discuss how the structure of the polymer ligand film can affect the data quality obtained. (LAUR-11-6773)

Log: 474. MATHEMATICAL EFFICIENCY CALIBRATION METHODS FOR HIGH QUALITY LABORATORY BASED GAMMA SPECTROMETRY SYSTEMS. P.J. LeBlanc (1), A. Adekola (1), F. Bronson (1), K. Morris (1), W.F. Mueller (1), R. Venkataraman (1); (1) - Canberra Industries Inc., 800 Research Parkway, Meriden, CT, 06450, USA.

The efficiency calibration of laboratory based gamma spectrometry systems typically involves the purchase or construction of calibration samples that are supposed to represent the geometries of the unknown samples to be measured. For complete and correct calibrations, these sample containers must span the operational range of the system, which at times can include difficult configurations of size, density, and source distribution. The efficiency calibration of a system is dependent not only on the detector, but on the detector-source configuration, and therefore is

invalid from sample to sample unless successive samples are identical. An alternative to source-based calibrations is to mathematically model the efficiency response of a given detector-sample configuration. In this approach, the measurement system is calibrated using physically accurate models whose parameters can generally be easily measured. The system operation can then be validated with a representative set of measurements, typically using radionuclide sources that can be traced to national standards organization. Using modeled efficiencies, systems can be quickly adapted to changing sample containers and detector configurations. This paper explores the advantages of using mathematically computed efficiencies in place of traditional source-based measured efficiencies for laboratory samples, focusing specifically on the possibility of sample optimization for a given detector, uncertainty estimation, and cascade summing corrections.

Log: 475. **THE COMMERCIAL FUTURE OF FOOD IRRADIATION.** JOSEPH BUTTERWECK, AEROSPACE & ENVIRONMENTAL MEDICINE GROUP.

The United States is the only nation that has advanced food irradiation to a sustainable commercial business. However the full potential has not been achieved. Rejection by the consumer is not a major obstacle. The nuclear industry, Government regulatory agencies, and the food industry all have to share some blame for the very slow progress of putting treated products on the retail grocery shelves. Pasteurization of hazardous foods of animal origin has been accepted since the 1930's. Cold pasteurization of beef and poultry is still the first technical HACCP intervention option. However, under the current environment, the profitability is marginal.

Log: 476. **PLUTONIUM DETERMINATION ON LARGE SEAWATER SAMPLES USING MnO_2 CARTRIDGES.** M. Eriksson¹, K. Ämmälä², I. Levy¹, J. Gastaud¹, J. Scholten³, J. Lehto², 1) IAEA-EL, Monaco, 2) University of Helsinki, Finland, 3) Universität Kiel, Germany.

Plutonium concentrations in the southern oceans are very low. Large volume samples combined with pre-concentration techniques and low detection limits are usually required for accurate Pu activity determination. This paper will describe a method where MnO_2 cartridges have been used for Pu determination in seawater. The advantages with this technique are that large volume of seawater can be passed through the cartridge system, no radiochemical yield determinant is required and that limited radiochemical manipulation is necessary onboard the ship when sampling. We have tested this technique on a southern ocean transect cruise and compared the result with the MnO_2 -precipitation method. It is expected that Pu is present in multiple oxidation states in seawater, which might have different affinity to absorb to the MnO_2 cartridge. However we found a very good agreement between the MnO_2 -precipitation and the MnO_2 cartridge techniques, showing that the cartridge system can be used in the southern Atlantic Ocean for the determination of Pu.

Log: 477. **SOURCE PREPARATION OF ACTINIDES AND POLONIUM USING COINS.** E. Holm⁽¹⁾, M. Eriksson⁽²⁾, B. Lind⁽¹⁾, I. Levy⁽²⁾, G. Kinn⁽¹⁾, (1)Norwegian Radiation Protection Authority, Norway (2) IAEA-EL, Monaco.

Electrodeposition of actinides such as Pu, Am, U, Th is normally performed on stainless steel discs. Also platinum and copper has been used. Polonium sources are prepared by spontaneous deposition on silver, nickel and copper. World wide coins are generally manufactured by bronze, steel, copper plated steel, cupronickel, brass, alloys of Cu-Ni-Zn sometimes with Al. The diameter varies between 16 and 25 mm and many of them fit into regular deposition cells such as liquid scintillation vials. The price of coins that can be used can be found for between 0.0012- 0.2 Euro i.e. cheaper than electro polished stainless steel discs (1 Euro). Usable coins are found in most countries. We have electro deposited actinides on Norwegian (50 Øre), Swedish (50 Öre) UK (1 Penny), Bulgarian (5 and 20 Stotinka), Thai coins (1 Bath) and European coins (0.01, 0.02, 0.05 Euro). Deposition was done from an ammonium sulphate solution at pH2 with a current of 1 A during 2 hours. Polonium was successfully spontaneously deposited from a weakly acid solution onto a 1 bath Thai coin (cupronickel). The coins were measured with alpha spectrometry using

ion implanted silicon detectors. The result is generally very good, indicating that the coins can be used in e.g. emergency situation if the stainless steel discs not available. In addition the coins tested in this study are considerable cheaper than the stainless steel disk. With high recoveries (60-100 %) depending on the elemental composition of the coins and energy resolution between 25-40 keV (FWHM). A pulse height distribution of ^{242}Pu and ^{243}Am is shown in Fig.1. (5 Bulgarian stotinka, brass).

Log: 478. **ENGINEERING UPGRADES TO THE RADIONUCLIDE AEROSOL SAMPLER/ANALYZER FOR THE INTERNATIONAL MONITORING SYSTEM.** Forrester, JB (1); Carty, F (2); Comes, L (2); Miley, HS (1); Morris, SJ (1); Ripplinger, M (1); Slaugh, RW (1); Van Davelaar, P (2). (1) Pacific Northwest National Laboratory, (2) General Dynamics-Advanced Information Systems.

The Radionuclide Aerosol Sampler/Analyzer (RASA) is an automated aerosol collection and analysis system designed by Pacific Northwest National Laboratory in the 1990's and is deployed in several locations around the world as part of the International Monitoring System (IMS) required under the Comprehensive Nuclear-Test-Ban Treaty (CTBT). The utility of such an automated system is the reduction of human intervention and the production of perfectly uniform results. However, maintainability and down-time issues threaten this utility, even for systems with over 90% data availability. Engineering upgrades to the RASA are currently being pursued to address these issues, as well as Fukushima lessons learned. Current work includes a new automation control unit, and other potential improvements such as alternative detector cooling and sampling options are under review. This paper presents the current state of upgrades and improvements under investigation.

Log: 479. **LOW-BACKGROUND UPGRADES FOR A CTBT RADIONUCLIDE LABORATORY.** Forrester, JB (1); Greenwood, L (1); Miley, HS (1); Myers, A (1); Overman, C (1). (1) Pacific Northwest National Laboratory.

The International Monitoring System (IMS) is a verification component of the Comprehensive nuclear Test Ban Treaty (CTBT), and in addition to a series of radionuclide monitoring stations, contains sixteen radionuclide laboratories capable of verification of radionuclide station measurements. This paper presents an overview of a new commercially obtained low-background detector system for radionuclide aerosol measurements recently installed in a shallow (>30 meters water equivalent) underground clean-room facility at Pacific Northwest National Laboratory. Specifics such as low-background shielding materials, active shielding methods, and improvements in sensitivity to IMS isotopes will be covered.

Log: 480. **ASSESSMENT OF UNCERTAINTY IN NUCLEAR FORENSICS ANALYSIS.** Gattiker, JR; Vander Wiel, S; Dale, DE; Los Alamos National Laboratory.

Nuclear forensics can be viewed as a complex inverse analysis. Models used to describe the progression of a nuclear event involve expertise and techniques from basic chemistry and physics to empirically calibrated data models. Combining these domains relies on expert knowledge to piece the various analytic components together to form a complete description of the unfolding nuclear event. The nuclear forensics problem starts with measurements at the end of this chain and seeks to find the initial conditions of the device and site. This inverse analysis includes many sources of uncertainty, including: uncertainty in the environment initial conditions, sampling and measurement error, radiochemistry analytic error, uncertainties in computational models and associated model-based analysis, and the uncertainties in expert judgment used to guide and constrain forensics analysis. Each of these contributions to uncertainty is challenging to characterize; joining them together into a set of conclusions presents deeper challenges. We discuss some insights from ongoing projects focusing on expressing uncertainty in the elements and overall process of forensics. We review the statistical analysis and uncertainty quantification (UQ) methods that are applicable to stages of the problem, and how these elements can be

drawn together, with expert judgment, to move toward rigorous statements of uncertainty in forensics conclusions. We also discuss communication of uncertainty, including frameworks of uncertainty representation, and approaches to standardized language for communication.

Log: 481. **BIOGEOCHEMICAL CONTROLS ON PLUTONIUM TRANSPORT IN THE SUBSURFACE.** Annie B. Kersting(1), Mavrik Zavarin(1), Brian A. Powell(2), Pihong Zhao(1), James Begg(1), Ruth Tinnacher(1), Ruth Kips(1), Harris Mason(1), Scott Tumey(1), Jen Fisher(3) and Patrick Huang(1). (1.)Glenn T. Seaborg Institute, Lawrence Livermore National Laboratory. Physical & Life Sciences, PO Box 808, Livermore CA 94550. (2.)Clemson University, and (3.)Desert Research Institute.

Radiocarbon analysis was performed to assess whether the content of biologically-derived components in diesel fuels could be measured by liquid scintillation counting (LSC) without any sample preconversion to eliminate chemical quench effects. Different fuel mixtures were prepared containing biodiesel and fossil diesel fuels and analyzed using a Quantulus TM ultra-low-level liquid scintillation counter. The specific ^{14}C activity of the mixtures was obtained from LSC measurements and directly related to the concentration of carbon originating from the biodiesel fuel. A linear relationship exists between the fuel mixture ^{14}C activity and the concentration of biodiesel in the mixture, indicating that direct determination of biodiesel content in fuels is possible via LSC measurements.

Log: 487. **IRRADIATION IN COMBINED TREATMENTS AND FOOD SAFETY.** Monique Lacroix, INRS-Institut Armand-Frappier, Canadian Irradiation Centre.

Irradiation combined with others processes can contribute to insuring food safety to consumers and controlling severe losses during transportation and commercialisation. We have demonstrated that using in synergy with other treatments; a lower dose could be used to eliminate pathogenic bacteria and permit a better protection of the sensorial quality and to prolong the shelf life of foods. Results indicated that some bacteria are more sensitive to irradiation under modified atmosphere (MAP) and the presence of active compound can increase the bacterial radiosensitivity by more than 4 times under air and by more than 10 times under MAP. Mild heat treatment or addition of natural antimicrobial compounds before irradiation treatment has also permitted an increase of *Bacillus cereus* radiosensitization. An increase of the bacterial radiosensitization of 1.5 and 1.56 was respectively observed. The effectiveness of the use of edible coating containing natural antimicrobial compounds, modified atmosphere packaging (MAP) or mild treatment before irradiation treatment was demonstrated in order to inactivate *Listeria monocytogenes*, *Salmonella Typhimurium*, *Escherichia coli* and *Bacillus cereus* growth or *Bacillus cereus* spore germination, to increase the bacterial sensitivity to irradiation, to reduce the water loss and to extend the shelf life of the food when stored at 4°C. Also, the use of edible coating previously crosslinked by irradiation have permitted a better control of the active compounds release. Studies of combined treatments were used in ready to eat vegetables, fruits and meat products.

Log: 488. **AN/LN SEPARATION BY USE OF BTP AND BTBP LIGANDS. NEW INSIGHTS USING ESI MS AND VIBRATIONAL SIDE BAND SPECTROSCOPY.** Clemens Walther(*†), Michael Steppert,† Ivana Císařová,# Thomas Fanghänel,‡,§ Andreas Geist,† Patric Lindqvist-Reis,† Petra Panak,†,§ Petr Štěpnička # Sascha Trumm, †,§ * Institute for Radioecology and Radiation Protection, Leibniz University Hanover, Herrenhäuser Str.2, D30419 Hanover, Germany.† Institute for Nuclear Waste Disposal, Karlsruhe Institute of Technology, P.O. Box 3640, 76021 Karlsruhe, Germany.# Department of Inorganic Chemistry, Faculty of Science, Charles University in Prague, Hlavova 2030, 128 40 Praha 2, Czech Republic.‡ Institute for Transuranium Elements, European Commission, Joint Research Centre, 76125 Karlsruhe, Germany.§ Ruprecht-Karls Universität Heidelberg, Physikalisch-Chemisches Institut, 69047 Heidelberg, Germany.

The present work focuses on highly selective ligands for An(III)/Ln(III) separation: bis-triazinyl-bipyridines (BTBP). By combining TRLFS, nano-ESI MS, vibronic sideband spectroscopy and X-ray diffraction we obtain a detailed picture of the structure and stoichiometry of the first coordination sphere of Eu(III)-BTBP complexes in octanolic solution, with the main focus on the 1:2 complexes, as extraction studies revealed, that those are the species extracted into the organic phase. The investigations on Eu(III) complexes of BTBP with different triazin alkylation revealed differences in the formed complexes due to the bulkiness of the ligands. Due to the vibronic sidebands in the fluorescence spectra, we were able to detect whether or not nitrate ligands are coordinated in the first coordination sphere of the Eu-complexes. In solution, less sterically demanding BTBP offer enough space for additional coordination of anions and/or solvent molecules to form 9-coordinated Eu-BTBP 1:2 complexes, while bulkier ligands tend to form 8-fold coordinated structures. We also report the first crystal structure of a Ln-BTBP 1:2 complex and that of its 1:1 complex, both of which 10-coordinated.

Log: 489. **PROSPECTS FOR THE INTRODUCTION OF WIDE AREA MONITORING USING ENVIRONMENTAL SAMPLING FOR PROLIFERATION DETECTION.** Dr. Ned Wogman, Pacific Northwest National Laboratory.

The International Atomic Energy Agency (IAEA) is committed to strengthening and streamlining the overall effectiveness of the IAEA safeguards system. The IAEA has investigated the use of environmental monitoring techniques and a variety of techniques were studied as part of extensive field trials. The efficacy of long-range monitoring depends on the availability of mobile signature isotopes or compounds and on the ability to distinguish the nuclear signatures from background signals and attribute them to a source. This paper serves as a scientific basis to start discussions of environmental sampling techniques that could be considered for wide-area monitoring for the detection of undeclared nuclear activities.

Log: 490. **DEFENSE THREAT REDUCTION AGENCY BASIC RESEARCH PROGRAM.** Petersen, DC. Defense Threat Reduction Agency.

The Defense Threat Reduction Agency (DTRA) safeguards the United States and her allies from weapons of mass destruction (WMD) by providing capabilities to reduce, eliminate, and counter the threat and mitigate its effects. The DTRA accomplishes its mission by investing in basic research efforts at universities, national labs and DoD service labs to better counter threats posed by WMD. We also facilitate productive relationships with other scientific organizations and seek to identify promising research efforts overseas. Through the Basic Research program, the DTRA recruits and trains scientists and engineers to develop a talented workforce for the future. Basic research is directed towards greater knowledge or understanding phenomena without regard to specific applications. The DTRA Basic Research Program pursues efforts within and across disciplines such as chemical science, computer/network science, materials science, mathematics, nuclear science, and physics. Research efforts are needed in the science of WMD sensing and recognition to advance capabilities to detect, identify, and characterize WMD materials.

Log: 491. **RADIATION AND HEAT INDUCED CHEMISTRY ISSUES IN PROCESSING LARGE AMOUNT OF ACTINIDES, LANTHANIDES AND FISSION PRODUCTS.** Miting Du and Chuck W. Alexander, Nuclear Material Processing Group, Fuel Cycle and Isotopes Division, Oak Ridge National Laboratory.

In separations of actinides, lanthanides and FP elements, R&D experiments with radioactive tracer or small amount of radioactive material, which gave satisfactory separation results, may not show the same when processing a large amount of same nuclear material. Due to the effects of high radiation and heat from the nuclear materials, repeat of the experiments on an enlarged scale may lead to unexpected chemical behavior of the organic extractants/resins/media in use, which gives separation results differ from that at low radioactive level. This presentation will give some of such examples in separations of actinides, lanthanides and fission product elements. Alternatives of processing pathways/methods will be proposed and discussed.

Log: 492. **FUKUSHIMA RADIO-MICROANALYTICAL PARTICLE MEASUREMENTS.** CJ Zeissler (NIST, Gaithersburg MD 20899); LPG Forsley (JWK Int. Corp., Annandale VA 22003); RM Lindstrom (NIST); J Davis (NIST); S Newsome (SeaBotix Inc., San Diego CA 92106); A Kirk (JK Int. Corp.).

Many hundreds of particles collected by air filters operated April, 2011 through May, 2011 (spanning about 6 weeks) from a harbor 40 km north of the Daiichi Nuclear Power Plant, Fukushima Prefecture, Japan are undergoing microanalysis. Nondestructive nuclear counting, autoradiography, particle isolation and chemical analysis by x-ray fluorescence techniques were applied to produce information on the radionuclides present, particle population information such as activity distributions, and the chemistry of particles found to host the radionuclides.

$^{134}\text{Cs}/^{137}\text{Cs}$ ratios are reported and compared to other literature on Fukushima debris detected elsewhere. Particles that were individually measured range in activity from about 700 mBq down to about 1 mBq, with the lower detection limit dependent on counting time and background conditions. The activities of individual particles between 2 mBq to 65 mBq are represented by a log normal distribution. Particles are described in terms of activity distributions, which are then related to size distributions and the chemistries of the particles to which the radionuclides are attached. The measurements are expected to assist with separate efforts to understand the nature of the accident, the structures or material affected, radionuclide transport and potential environmental or biological uptake.

Log: 493. **CHALLENGING STUDIES IN RADIANALYTICAL AND NUCLEAR CHEMISTRY BY USING ACCELERATORS AT TOHOKU UNIVERSITY.** T. Ohtsuki, Research Center for Electron Photon Science.

We describe the status of the 300 MeV electron linear accelerator(LINAC) and the 110 MeV cyclotron used for radioanalytical and nuclear chemistry at Tohoku University. In our accelerator facility, several scientific programs in production of radioactive sources have been performed for nuclear chemistry, activation analysis, life-science, inorganic chemistry, hot-atom chemistry, and production of radioactive tracers for materials science, etc. First we show the instrumental developments of the irradiation system and the measurement system of several radio-activities in the LINAC and Cyclotron facilities. Further, typical topics can be shown as following at the conference. The formation of atom-doped C60 and C70 etc. has been investigated by using several types of radionuclides produced by nuclear reactions. From the trace of the radioactivities after high performance liquid chromatography (HPLC), it was found that formation of endohedral fullerenes (or heterofullerene) with small atoms (Be, Li), noble-gas atoms (Kr, Xe) and ^4B - ^6B elements (Ge, As, Se, Sb, Te etc.) is possible by a recoil process following the nuclear reaction. The decay rate of ^7Be electron capture (EC) was measured in C60 and Be metal with a reference method. The half-lives of ^7Be endohedral C60 ($^7\text{Be}@C60$) and ^7Be in Be metal (Be metal(^7Be)) were found to be 52.65 ± 0.04 and 53.25 ± 0.04 days, respectively. This amounts to a 1.13% difference in the EC-decay half-life between $^7\text{Be}@C60$ and Be metal(^7Be). The result is a reflection of the different electron wave-functions for ^7Be inside C60 compared to when ^7Be is in a Be metal.

Log: 494. **THE EFFICACY OF FACIAL SWIPES AND SWABS FOR DETERMIANTION OF LUNG DEPOSITION IN SIMULATED AEROSOL EXPOSURE..** Ko, R(1); Li, C(1); Shew, C(2); Waller, E(2); Perera, S(2). (1)RPB (2)UOIT.

The use of facial swipes and nasal swabs is a procedural step in screening individuals for contamination in the event of nuclear emergency or occupational exposures. However, differences in environmental conditions, physiological conditions, collection methods, and analysis methods limit the certainty for differentiating between low, medium, and high exposure groups. The present study investigates swipe and swab sample collection under various conditions in an aerosol deposition chamber containing an artificial breathing apparatus.

Lanthanum oxide in the 5 micron AMAD range was used to simulate dispersal of alpha emitters. The distribution of the particles between the face, respiratory tract, and lungs were measured using both ICP-AES and NAA.

Log: 495. ALPHA SPECTROMETRIC EVALUATION OF SRM-995 AS A POTENTIAL URANIUM/THORIUM DOUBLE TRACER SYSTEM FOR AGE-DATING URANIUM

MATERIALS. F.E. Stanley (1); S.E. Glover (1); A.M. Stalcup (1); H.B. Spitz (1); D.M. Beals (2) : 1) University of Cincinnati, 2) Savannah River National Laboratory.

Uranium-233 ($t_{1/2} \sim 1.59 \times 10^5$ years) is an artificial, fissile isotope of uranium that has significant importance in nuclear forensics. The isotope provides a unique signature in determining the origin and provenance of uranium-bearing materials and is valuable as a mass spectrometric tracer. Alpha spectrometry was employed in the critical evaluation of a ^{233}U standard reference material (SRM-995) as a dual tracer system based on the in-growth of ^{229}Th ($t_{1/2} \sim 7.34 \times 10^3$ years) for ~ 35 years following radiochemical purification. Preliminary investigations focused on the isotopic analysis of standards and unmodified fractions of SRM-995; all samples were separated and purified using a multi-column anion-exchange scheme. The $^{229}\text{Th}/^{233}\text{U}$ atom ratio for SRM-995 was found to be 1.598×10^{-4} ($\pm 4.50\%$) using recovery-corrected radiochemical methods. Using the Bateman equations and relevant half-lives, this ratio reflects a material that was purified ~ 36.8 years prior to this analysis. The calculated age is discussed in contrast with both the date of certification and the recorded date of last purification.

Log: 496. THE RAPID DETERMINATION OF STRONTIUM 89 AND STRONTIUM 90 IN ENVIRONMENTAL SAMPLES. O'Brien, TC (1); Jassin, LE (1); Horwitz, EP (2); McAlister, DR (2). (1) Eichrom Technologies, (2) PG Research Foundation, Inc.

The analysis of strontium-89,90 following a nuclear incident is very important for determining the potential for dose due to beta emissions in the potentially affected area. There is a need to rapidly determine strontium-89,90 with low detection limits in a variety of environment samples. A new rapid, simple and effective chromatographic method for the separation of strontium and yttrium from environmental samples will be described. Currently in many laboratories only the strontium is retained from the initial sample separation requiring the subsequent ingrowth of the yttrium-90 and then a second separation for the determination of yttrium-90. The use of stacked cartridges allows for the independent retention of yttrium from the parent strontium radioisotopes from the original sample during initial chromatographic separation. This independent retention and subsequent elution for analytical measurement of yttrium and strontium parent allows for the rapid measurement of the target beta emitting radionuclides. With both rapid total radio-strontium and yttrium-90 measurements it is possible to calculate the strontium-89,90 present in the original sample within hours of sample delivery to the laboratory. The sample preparation method, separation procedure and beta measurement and analytical data analysis for a number of sample matrices will be presented. The use of stacked cartridges with vacuum box technology leading to rapid sample separation flow rates will be covered.

Log: 497. FUKUSHIMA EVENT RECONSTRUCTION USING MODELING AND ISOTOPE RELATIONSHIPS. Hoffman, I., Korpach, E., Mekarski, P., Ungar, K., Yi, J., Zhang, W.

Using particulate and noble gas data from the Comprehensive Nuclear Test-Ban-Treaty International Monitoring System, Canadian National Surveillance networks and atmospheric transport modelling, It is possible to associate some of the main events in the Fukushima accident timeline with observations, and perturbations in isotopic ratios in Japan, through the Pacific and into North America. Additional sources identified during the accident period will be commented upon in the context of CTBT treaty verification.

Log: 498. **AN AUTOMATIC REAL-TIME FIXED POINT SURVEILLANCE NETWORK AND ITS AMBIENT GAMMA DOSE-RATE MONITORING AT THE CANADIAN EMBASSY IN TOKYO DURING THE FUKUSHIMA NUCLEAR ACCIDENT.** Kurt Ungar, Weihua Zhang, Ed Korpach, Mike Benotto, Rodney Berg, Ryan Lawrie, Christopher Tulk, Brian White and Jing Yi.

To provide the federal preparedness and response framework for all radiological and nuclear (RN) emergencies affecting Canadians, Health Canada (HC) has been working with federal nuclear emergency plan (FNEP) partners to integrate fixed point surveillance (FPS) and laboratory sample analysis data. The FPS network consists of numbers of spectroscopic NaI(Tl) detectors measuring, in real-time, ambient gamma dose-rate around Canadian nuclear facilities, population centers, ports that handle nuclear vessels and additional quick deployable detectors for special events and RN emergencies. In response to the nuclear emergency in Fukushima, Health Canada deployed several detectors to the western part of Canada and the Canadian embassy in Tokyo. The ambient gamma dose-rate levels were monitored on these systems to assist decision-making during the emergency, and ensure the health and safety of Canadians at home and abroad. This paper first provides a brief description of the design, installation and operation of the FPS network. The results presented in this paper are mainly based on the ambient dose-rate monitoring by the detectors installed at the Canadian embassy in Tokyo during Fukushima nuclear accident.

Log: 499. **DEVELOPMENT OF A NEW AEROSOL MONITORING SYSTEM AND ITS APPLICATION IN FUKUSHIMA NUCLEAR ACCIDENT RELATED AEROSOL RADIOACTIVITY MEASUREMENT AT THE CTBT RADIONUCLIDE STATION IN SIDNEY OF CANADA .** Kurt Ungar, Weihua Zhang, Marc Bean, Mike Benotto, Jeff Cheung, and Brian Ahier.

A high volume aerosol sampler ("Grey Owl") has been designed and developed at the Radiation Protection Bureau, Health Canada. Its design guidance is based on the need for a low operational cost and reliable sampler to provide daily aerosol monitoring samples that can be used as reference samples for radiological studies. It has been developed to provide a constant air flow rate at low pressure drops (about 3 kPa for a day sampling) with variations of less than 1% of the full scale flow rate. Its energy consumption is only about 1.5 kW for a filter sampling over 22,000 standard cubic meter of air. It has been demonstrated in this Fukushima nuclear accident related aerosol radioactivity monitoring study at Sidney station, B.C. that the sampler is robust and reliable. The results provided by the new monitoring system have been used to support decision-making in Canada during an emergency response.

Log: 500. **DETERMINATION OF U CONCENTRATION IN GINSENG SAMPLES USING BY DELAYED NEUTRON ACTIVATION ANALYSIS.** J.M. Lim(1), J.H. Moon(2), S.H. Kim(2), M.J. Kang(1), J.H. Jeong(3), J.H. Lee(3). (1.) Environmental Radioactivity Assessment Team, Korea Atomic Energy Research Institute. (2.) Department of Neutron Basic Science, Korea Atomic Energy Research Institute. (3.) Department of Environmental Engineering, ChungNam National University.

Uranium and its salts are both toxic and radioactive. And it is well known that the soluble uranium compounds are most toxic to the kidneys. However, some high sensitivity methods including thermal ionization mass spectrometry (TIMS), inductively coupled plasma mass spectrometry ICP-MS, and isotope dilution mass spectrometry (ID-MS) are widely applied for U analysis; there are many limitations such as acid digestion or dilution to apply for solid samples (e.g., soil, food, particulate, and rock). Delayed Neutron Activation Analysis (DNAA) is the analytical method for measuring U-235 by neutron activation followed by delayed neutron counting. U analysis by DNAA has big advantages in terms of lower detection limits and shorter analytical time than that by instrumental neutron activation analysis (INAA). In this study, we present the analytical results of U for ginseng collected in the five farm areas in Korea. The ginseng samples were classified with cultivation areas and its cultivation periods (e.g., 1, 3, and 5 years). The samples, which are pretreated by microwave washing and drying, were also divided with head, body, epidermal, and root, respectively. The average U concentrations in 5-year ginseng samples were 41.7 ± 12.2 ng/g.

Among the body, head, epidermal, and root, the U concentrations in body showed the lowest level. ANOVA test indicated that the geographical and durational difference of U concentration in ginseng samples ($P < 0.05$) was significant.

Log: 501. QUALITY ASSURANCE OF RADIOACTIVITY OF TRITIUM DETERMINED BY HIGH TEMPERATURE FURNACE AND LIQUID SCINTILLATION COUNTER. J.M. Lim(1), M.J.

Kang(1), G.S. Choi(1), J.H. Lee(2). (1.) Environmental Radioactivity Assessment Team, Korea Atomic Energy Research Institute. (2.) Department of Environmental Engineering, ChungNam National University.

Because of the potential impact on the human body and on the ecosystem, the wastes from nuclear sites are particularly important as radioactive by-products caused environmentally detrimental effects. The wastes (e.g., activated carbon, oils, resins, etc.) were consistently generated with the actions to prevent the emission of the radioactive pollutants from nuclear sites. The radioactivity of the wastes has been determined by specific methods. Tritium (^3H), which is a pure beta emitter (18.6 Kev), is a mainly produced by the neutron capture reaction of Li. For the decision of disposal methods of the wastes, the analytical methods could determine the radioactivity of ^3H which existing in the very low levels. The ^3H of activated carbon samples were trapped in HNO_3 by combusting the samples in a purpose-designed tubular high temperature furnace. Measuring of radioactivity was carried out in the liquid scintillation counter. In this study, various performance tests of this method were conducted with standard solutions. In order to evaluate the dynamic range of the method, the real samples with spiking ^3H standard solution were also analyzed. As the duplicated samples ($n=3$) were analyzed to determine the repeatability of the method, the relative standard deviation of the results showed about 4.4%. The accuracy and precision of procedure also examined with NIST SRM solutions. Because of importance for the analytical results in the low level radioactivity, the method detection limit and the uncertainty of the method was evaluated in detail. The major factors of standard uncertainty were grouped into the sample preparation, combusting, trapping, and scintillation counting, respectively. Finally, the major factors of standard uncertainties were determined by sensitivity tests.

Log: 502. RADIONUCLIDES PRODUCTION BY DEUTERON BEAMS IRRADIATIONS.

Bonardi, ML (1), Manenti, S (1), Gini, L (1), Groppi, F (1), (1) L.A.S.A. - Radiochemistry Laboratory, Università degli Studi di Milano and INFN Sez. di Milano, via F.lli Cervi 201, I-20090 Segrate (MI), Italy.

Theranostic medicine is a new integrated therapeutic system which can diagnose, deliver targeted therapy and monitor the response to therapy. This multifunctional platform is an emerging highlight in medicine that combines both diagnostic/imaging and therapeutic aspects: the nuclear physician can follow the real biodistribution of the radiopharmaceutical inside the patient after the injection and the follow-up during the repeated treatments. The radioisotopes used for metabolic radiotherapy are α , β and Auger electron emitters. Many of them are also γ emitters and can be detected by gamma-camera, SPECT or PET. Many of these "neutron reach" radionuclides are produced by nuclear reactor with a very low specific activity - AS. In selected cases they can be produced by bombardment of targets by charged particle beams, in No Carrier Added Form with very high AS. If the irradiations are made with deuteron beams some more advantages are obtained: 1) the higher stopping power in respect to the protons allows to employ targets with smaller thickness: the volume of reagents, the synthesis systems and the discharge of radioactive material for radioprotection purpose are smaller, the AS and chemical purity of the final product are higher. 2) deuterons usually present higher cross sections in compound nucleus region. We present some results obtained at our Laboratory for the production optimization of some theragnostic radionuclides produced by irradiation at JRC – Ispra Cyclotron with deuteron beams accelerated up to 19 MeV and the Arronax Cyclotron, that allows to extend the energy interval up to 35 MeV.

Log: 503. **NUCLEAR AND RADIOCHEMICAL TECHNIQUES: A POWERFUL TOOL FOR NANOTOXICOLOGICAL STUDIES.** Sabbioni, E (1,2), Manenti, S (2), Bonardi, ML (2), Bellini, CM (2), Groppi, F(2), (1) ECSIN – European Center for the Sustainable Impact of Nanotechnology, Veneto Nanotech –Rovigo, Italy,(2) L.A.S.A. - Radiochemistry Laboratory, Università degli Studi di Milano and INFN Sez. di Milano, via F.lli Cervi 201, I-20090 Segrate (MI), Italy.

Nanotechnology, perceived as one of the key technologies of this century. Despite the term “nanotechnology” is synonymous with things that are innovative and highly promising, little attention has been dedicated to the other side of the coin, i.e. the research on toxicological effects and on the relations with factors that can affect the nanotoxicity on human health and on the environment. Research on the impact of engineered nanoparticles (NPs) is strongly hampered by a lack of reliable tools to detect, visualize and quantitatively trace particles movement and transfer in complex environmental and biological systems. A few methods, such as labelling with fluorescent probes, may overcome some of the detection problems; however, leading a significant modification of the particles to be traced, the engineered NPs behavior is modified. Conversely radiolabelling or radioactivate the NPs to make them distinctive and thus easily detectable, is one very smart way to solve the problem especially because after neutron or proton activation the physicochemical characteristics in terms of size distribution and Z potential are maintained as the “cold” ones. One of the debated themes is the influence of NPs on the human reproductive system. In this contest a way to have experimental evidence is the study of AuNPs and AgNPs passage through the blood-testicular barrier in vivo. In this work some preliminary results obtained by NAA of the sperma (seminal liquid plus sperms) from selected strain of rabbits are presented. Samples were taken from animals exposed for different time (3 and 7 days) to 5 mg AgNPs and AuNPs $\text{kg}^{-1}\text{b.w.}$

Log: 504. **MEASUREMENTS RESULTS OF THE FALLOUT AT NORTH REGIONS OF ITALY AFTER FUKUSHIMA INCIDENT.** Groppi, F(1), Manenti, S (1), Gini, L (1), Ioannidou, A (2,1), (1) L.A.S.A. - Radiochemistry Laboratory, Università degli Studi di Milano and INFN Sez. di Milano, via F.lli Cervi 201, I-20090 Segrate (MI), Italy, (2) Nuclear Physics & Elementary Particle Physics Division, Department of Physics, Aristotle University of Thessaloniki, 54124, Thessaloniki, Greece.

Radionuclides from Fukushima were first detected at Milano region in a rain water sample, collected during March 27-28, with rainwater activities to be 0.89 Bq L^{-1} and 0.12 Bq L^{-1} for ^{131}I and ^{137}Cs respectively. During the same days a snowfall sample was collected from Monte Rosa mountain at a height of 3000 m, with the concentrations of ^{131}I and ^{137}Cs in snowfall to be lower than that in rainwater sample. A sample of dry deposition that was collected 9 days after the first rainfall event of 27-28 March, 2011 showed that the dry deposition of ^{131}I and ^{137}Cs was 0.40 Bq m^{-2} and 0.30 Bq m^{-2} respectively. The concentrations of ^{131}I in sheep and cow milk samples collected on 30 March, 2011 from a farm at Macugnaga, Monte Rosa mountain, were 0.30 Bq L^{-1} and 0.37 Bq L^{-1} respectively. Daily monitoring of the airborne activity levels was carried out with a high volume air sampler and Glass Fiber filters as the collection substrate. Increased atmospheric radioactivity was detected on air filter taken on 30 March 2011, while the maximum activity of $467 \mu\text{Bq m}^{-3}$, occurred at April 3-4, 2011. A week later the activities had fallen to about 50% of peak values, with a general decreasing trend over the following days. In the time period of one month after the nuclear accident, concentrations of ^{137}Cs and ^{134}Cs in air as high as $88 \mu\text{Bq m}^{-3}$ and $78 \mu\text{Bq m}^{-3}$, respectively were recorded. According to the measurements, airborne activity levels remains of no concern for public health in Italy.

Log: 506. **DEVELOPING AND EVALUATING HDEHP, HEH[EHP], AND H[TMPEP] BASED POLYMER LIGAND FILM (PLF) FOR EXTRACTING PLUTONIUM.** Rim, J (1,2); Peterson, D (2); Armenta, C (2); Gonzales, E (2), Unlu, K (1), (1) Pennsylvania State University, (2) Los Alamos National Laboratory.

This paper presents a development of HDEHP, HEH[EHP], and H[TMPeP] based polymer ligand film (PLF) for rapid extraction of plutonium from solution. The ligands were incorporated into a polymer matrix to generate a thin film substrate. A polymer matrix provided a smooth surface and eliminates need for extensive and time consuming chemical processes to count sample with alpha spectroscopy. PLFs were tested using ^{239}Pu and ^{240}Pu tracer. The pH of plutonium solution and total activity was varied to study effectiveness of PLF in different conditions. Entire samples were analyzed with alpha spectroscopy to measure recovery of plutonium by particular ligand. Some samples were further studied with Thermal Ionization Mass Spectrometry (TIMS). For TIMS analysis, plutonium was stripped from a sample and column chromatography was performed to remove any impurities.

Log: 507. **STUDY ON SEPARATION METHOD OF THE 178M2HF ISOMER FOR INDUCED GAMMA EMISSION EXPERIMENT.** JIANG Tao, HE Yu-hui, YANG Jun, YANG Tianli; Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics, Mianyang 621900, China.

As the target material for induced Gamma emission experiment, 178m2Hf (OH) 4 was obtained in using several separation methods, such as deposition, extraction and anion/cation exchange from the 178m2Hf sample, which was prepared by radiating Yb target of Cu-base magnetron sputtering with α particles. The integrity moulding target was prepared by vacuum filtration technology. Chemical yield and decontamination coefficient were measured with tracing technology. The results show that the chemical yield of the process is about 69.3% for Hf and the process's decontamination performance is high for ^{65}Zn and better than for ^{173}Lu . The decontamination coefficients for ^{65}Zn and ^{173}Lu are about 105 and 103, respectively.

Log: 508. **A NOVEL RAPID DETECTION APPROACH FOR THE ANALYSIS OF RADIONUCLIDES IN ENVIRONMENTAL SAMPLES USING GRAPHITE MALDI MASS SPECTROMETRY.** Samantha Walton and David Mitchell, AWE Aldermaston.

Graphite - Matrix Assisted Laser Desorption Ionisation Mass Spectrometry (G-MALDI-MS) has been shown to be a more effective method for the generation and detection of various actinide and lanthanide elements compared with that observed by direction laser irradiation. In contrast to the use of conventional matrices for MALDI-MS, graphite is found to reduce the extent of isobaric interference across the mass range of interest, providing enhanced detection limits. Research undertaken at AWE has shown that G-MALDI-MS provides a rapid means of isotopic signature determination at an acceptable level of accuracy and precision for its application as a potentially rapid radionuclide screening method for environmental samples. As such G-MALDI-MS offers potential as an additional tool (implemented in conjunction with radiometric methods) for Nuclear Forensic investigations where the low level detection and identification of trace amounts of radionuclides and high explosives is of paramount importance. The ability of G-MALDI-MS to rapidly detect trace levels of radionuclides and high explosives is demonstrated with an assessment of the achievable sensitivity, resolution and quality of isotope ratio determination. Consideration is also given to the likely detection limits within environmental samples, in which isobaric interference will be more problematic; in the light of this an evaluation of the future use of G-MALDI-MS for these applications is provided. © British Crown Owned Copyright 2011/AWE
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Log: 509. **SIMULATION OF 2-SITE LANGMUIR MODEL FOR CHARACTERIZING THE SORPTION CAPACITY OF CS AND SE IN CRUSHED MUDROCK UNDER VARIOUS IONIC STRENGTH EFFECTS.** Chuan-Pin Lee(Department of Earth Science, National Cheng Kung University) ; Ming-Chee Wu (Department of Earth Science, National Cheng Kung University) ; Tsuey-Lin Tsai(Chemical Analysis Division, Institute of Nuclear Energy Research) ; Hwa-Jou Wei(Chemical Analysis Division, Institute of Nuclear Energy Research) ; Lee-Chung Men(Chemical Analysis Division, Institute of Nuclear Energy Research).

The sorption capacity of cesium and selenium in crushed mudrock was demonstrated in this study through a 2-site Langmuir model. To employ such numerical analysis, this study applied batch tests with kinetic sorption experiments (time-dependent) and different concentrations (10^{-2} ~ 10^{-7} M) in synthetic groundwater and seawater. Moreover, the distribution coefficients (K_d) in linear sorption assumption only be demonstrated under 10^{-7} M for Cs and 10^{-5} M for Se with a solid to liquid ratio (1g : 30mL). In fact, the kinetic experiments after 14 days failed to fit the K_d models due to the test condition (10^{-4} M) in non-linear range. The fitting results showed a two-site Langmuir model has been found to be capable of appropriately describing Cs and Se sorption in mudrock under test conditions(10^{-2} ~ 10^{-7} M). Consequently, the sorption capacity was about 0.06mol/kg for Cs and 0.015 mol/kg for Se and indicated the less relation to ionic strengths and chemical compound in liquid except Ca^{2+} ion.

Log: 510. **BEHAVIOR OF FISSION PRODUCTS IN THE COURSE OF DISSOLUTION OF SIMULATED SPENT NUCLEAR FUEL IN IRON NITRATE SOLUTIONS AND OF RECOVERY OF URANIUM AND PLUTONIUM FROM THE RESULTING SOLUTIONS.** Yu. M. Kulyako(1), S. A. Perevalov(2), T. I. Trofimov(3), B. F. Myasoedov(4), V.I. VERNADSKY INSTITUTE OF GEOCHEMISTRY AND ANALYTICAL CHEMISTRY RAS (GEOKHI RAN).

Experiments aimed to examine the spent nuclear fuel dissolution in iron(III) nitrate solutions and to elucidate the behavior of fission products in the process were performed with simulated fuel corresponding to spent nuclear fuel of a WWER-1000 reactor. In Fe(III) nitrate solutions, U is quantitatively transferred from the fuel together with Cs, Sr, Ba, Y, La, and Ce, whereas Mo, Tc, and Ru remain in the insoluble precipitate and do not pass into the solution, and Nd, Zr, and Pd pass into the solution to approximately 50%. The recovery of U or jointly U + Pu from the solution after the dissolution of oxide nuclear fuel is performed by precipitation of their peroxides, which allows efficient separation of actinides from residues of fission products and iron.

Log: 511. **WORLDWIDE MEASUREMENTS OF RADIOXENON BACKGROUND NEAR ISOTOPE PRODUCTION FACILITIES, A NUCLEAR POWER PLANT AND AT REMOTE SITES – THE “EU/JA-II” PROJECT.** P.R.J. Saey^{1a}, A. Ringbom², T.W. Bowyer³, M. Zähringer^{4b}, A. Faanhof⁵, C. Labuschagne⁶, M. Al-Rashidi⁷, J. Scheer⁸, U. Tippawan⁹, B. Verboomen¹⁰, 1: Vienna University of Technology, Atomic Institute of the Austrian Universities, Stadionallee 2, 1020 Wien, Austria, 2: Swedish Defence Research Agency (FOI), Systems Technology Division, 172 90 Stockholm, Sweden, 3: Pacific Northwest National Laboratory (PNNL), P.O.Box 999, Richland, WA 99352, USA, 4: Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organisation, Provisional Technical Secretariat, PO Box 1200, 1400 Wien, Austria, 5: South African Nuclear Energy Corporation (NECSA), Pretoria, South Africa, 6: South African Weather Service (SAWS), Stellenbosch, South Africa, 7: Kuwait Institute of Scientific Research (KISR), Coastal & Air Pollution Department, Safat, Kuwait, 8: E.ON, Kernkraftwerk Isar, Dammstraße, 84051 Essenbach, Germany, 9: Chiang Mai University, Physics Dept., Fast Neutron Research Facility (FNRF), 50200 Chiang Mai, Thailand, 10: National Institute for Radioelements (IRE), Avenue de l'Espérance 1, 6220 Fleurus, Belgium | a: Now at International Atomic Energy Agency, Wagramer Strasse 5, P.O. Box 100, 1400 Vienna, Austria, b: Now at Bundesamt für Strahlenschutz, (BfS), Rosastr. 9, 79098 Freiburg, Germany.

Radioxenon measurements are one of the principle techniques used to detect underground nuclear explosions and specifically, the presence of one or more radioxenon isotopes allows one to determine whether a suspected event was a nuclear explosion or originated from a civilian source. During the design of the International Monitoring System (IMS), which is part of the verification regime for the Comprehensive Nuclear-Test-Ban Treaty (CTBT), it was determined, that radioxenon monitoring should be performed at 40 or more stations worldwide. At the time of the design of the IMS, however, very few details about radioxenon background were known and it is now recognized that the backgrounds are probably evolving anyhow. This paper presents the results of measurement campaigns to study the worldwide concentrations of radioxenon isotopes. It contributes essential data to characterize civil sources and to distinguish from signatures from nuclear explosions. It focuses in locations where there was either very little information or there was a unique opportunity to learn more about emissions from known large sources. The locations reported here are Belgium, Germany, Kuwait, Thailand and two locations in South Africa. A mobile sampler and analyzer was transported to the vicinity of specific known sources in these countries and 12 hour samples were taken in sampling periods between 5 days and 3 months. The findings corroborate the hypothesis that a few major radioxenon sources contribute in great part to the global radioxenon background. Measurements from locations very close to the source provide unique data on isotopes other than Xe-133 which are sparsely detected in the CTBT monitoring network. Nuclide ratios are used in order to better characterize the sources and to understand and model xenon production processes. This and good quality meteorological transport modelling provides the toolbox to reliably distinguish civil sources from nuclear explosions. Another finding was the discovery of relevant independent sources of Xe-131m (the daughter of I-131), which has potential to degrade isotopic discrimination, a tool for screening out civilian isotopic uses from treaty verification monitoring. As a consequence, there is some need to investigate local potential sources in the area of medical applications of I-131.

Log: 512. **WOSMIP II - WORKSHOP ON SIGNATURES OF MEDICAL AND INDUSTRIAL ISOTOPE PRODUCTION.** Paul R.J. Saey(1), Theodore W. Bowyer(2), Ian M. Cameron(2), Judah Friese(2), Rosara F. Kephart(2), Murray Matthews(2), Harry S. Miley(2) | 1: Vienna University of Technology, Atomic Institute of the Austrian Universities, Stadionallee 2, 1020 Vienna, Austria, 2: Pacific Northwest National Laboratory, PO Box 999, Richland, WA, United States.

Medical and industrial isotopes are fundamental tools used in science, medicine and industry. Their principal use is in diagnosis and treatment planning. Tc-99m, derived from Mo-99, is by far the most heavily utilized isotope. It has broad applications, like e.g. evaluating the function of heart, liver, thyroid, blood flow and detection of prostate, breast and bone tumours. This method has around 30 million procedures per year. The main production of Mo-99 is reactor based, i.e. fission of U-235. In this process, the uranium targets are irradiated, mostly in research reactors, then cooled for a short time before being dissolved and then the Mo-99 separated from the rest of the fission products. The fission gases (e.g. radioxenons) are set free during the dissolution. They are often trapped, which is done differently at each facility before being released into the atmosphere. Measurements of ultra-low concentrations of radioxenon isotopes, however, are also an important verification tool for e.g. identifying underground nuclear explosions as well as for undeclared nuclear activities. The "Workshop on Signatures of Medical and Industrial Isotope Production" (WOSMIP) initiative was founded in 2009. Its goal is better understand the isotopic and chemical signatures created through isotope production mechanisms. The main producers are currently situated in Canada (Chalk River), The Netherlands (Petten), Belgium (Fleurus), South Africa (Pelindaba), Indonesia (Serpong), Argentina (Eizeza) and Australia (Lucas Heights). Six of these seven producers were present at the second WOSMIP workshop to discuss several operational topics with the monitoring and verification community (71 attendees from 21 countries). This paper presents the main outcomes of the 2011 workshop on topics like emission and background reduction presented by different producers, recent and planned activities at isotope production facilities and the experience in monitoring emissions. The workshop concluded: (1) WOSMIP 2011 attendees recognized that medical isotope production is a global problem for the verification community, (2) The background from present-day medical isotope production efforts can be managed and should not detract from progress towards entry-into-force of the CTBT, (3) However, given the signaled increase in global production, steps must be taken now to manage the issue, (4) Participants at WOSMIP-2011 discussed ways to raise visibility of the issue within the appropriate communities and to form joint experiments examining effluent monitoring, etc., (5) It was suggested that the group works towards establishing a proposed baseline acceptable level of emissions for medical isotope production – one that was internationally accepted, but voluntary. WOSMIP III will take place in Strassoldo, Italy, between Tuesday 18 – Friday 22 June 2012. More information will be available at: <http://wosmip.pnnl.gov/>.

Log: 513. **CHEMICAL YIELD TECHNIQUES FOR RADIONUCLIDES AT AWE.** DR ANDREW R KING, AWE plc.

AWE has had the capability to separate and purify radiochemistry samples for many years. To determine the effectiveness of the separation, known amounts of either radioactive tracers or inactive carrier solutions are added at the start of the method. These isotopes/elements are then measured at the end of the scheme to calculate the chemical yield. Corrections to the counting data of the analytes of interest are subsequently calculated using the chemical yield data. Recent improvements have been made at AWE in the techniques used to ascertain the chemical yields. A summary of x-ray fluorescence (XRF) and inductively coupled plasma atomic emission spectrometry (ICP-AES) procedures will be provided, including a comparison of results and uncertainty of measurements. In addition, the potential benefits of ICP-Quadrupole Mass Spectrometry will be considered in terms of future applications to the current procedures.

Log: 514. **SOURCE TERM ANALYSIS OF URANYL NITRATE TO DETECT DIVERSION AT NATURAL URANIUM CONVERSION FACILITIES.** Dewji, S. A. (1)(2); McElroy, R. (1); Lee, D. (1); Chapman, J. A. (1); Hertel, N. E. (2). (1) Oak Ridge National Laboratory; (2) Nuclear and Radiological Engineering Program, Georgia Institute of Technology.

Historically, safeguarding nuclear material in the front-end of the fuel cycle was implemented only when UF₆ was declared as feedstock for enrichment plants. With each step in the conversion process from uranium yellowcake to feedstock for UF₆, intermediary uranium oxide and fluoride compounds become progressively attractive products for diversion. The diversion of this product material could potentially provide feedstock for clandestine or undeclared enrichment for weapons development for state or non-state entities. Recent International Atomic Energy Agency policy papers have sought to implement safeguards when any purified aqueous uranium solution or oxides suitable for isotopic enrichment or fuel fabrication exist. Oak Ridge National Laboratory has developed the Uranyl Nitrate Calibration Loop Equipment (UNCLE) facility to simulate the full-scale operating conditions of a purified uranium-bearing aqueous stream exiting the solvent extraction process in a natural uranium conversion plant (NUCP). UNCLE has recently tested neutron-based detectors for operations monitoring of NUCP activities. Currently, gamma-ray signatures are being investigated to detect undeclared activities in a timely manner. The preliminary results of gamma-ray source term modeling will be presented as part of a comprehensive validation effort employing gamma-ray detection instrumentation for the detection of diversion from declared conversion activities.

Log: 515. **SPECIATION ANALYSIS OF ¹²⁹I IN THE ENVIRONMENT.** Xiaolin Hou, Center for Nuclear Technology, Risø DTU, Technical University of Denmark.

¹²⁹I (15.7 Ma) is a naturally occurring radioisotope of iodine, while releases from human nuclear activities dominate the present ¹²⁹I level in the environment, where the nuclear reprocessing facilities are responsible for about 90% of the anthropogenic releases. These sources provide a unique tracer for researches on oceanographic, atmospheric, and environmental processes by chemical speciation analysis of ¹²⁹I and stable iodine. Various methods for speciation analysis of ¹²⁹I and ¹²⁷I in environmental samples such as seawater, fresh water, atmosphere, soil and sediment have been established in our laboratory by chemical separation coupled with AMS and ICP-MS measurement. For fresh water and seawater, solvent extraction as well as anion exchange chromatography combined with decomposition of organic matters with chemical oxidation have been applied to separate iodide, iodate and total iodine. For speciation analysis of ¹²⁹I in low level ¹²⁹I, anion exchange chromatography followed by coprecipitation is employed for the speciation analysis of ¹²⁹I. A rapid method has also been developed for the separation of carrier free ¹²⁹I from seawater samples by coprecipitation of AgI-AgCl in suitable conditions. The speciation analysis of ¹²⁹I in atmosphere is carried out by collecting particle associated, gaseous inorganic and gaseous organic iodine using a special designed air sampler, with combustion decomposition, coprecipitation separation and AgI for AMS measurement. A modified sequential extraction has been employed for the speciation analysis of ¹²⁹I in soil and sediment. This presentation will summarize all these methods, as well as demonstrate their environmental tracer applications.

Log: 516. **ACCURATE DETERMINATION OF RARE EARTH ELEMENTS IN COSMOCHEMICAL SAMPLES BY RADIOCHEMICAL NEUTRON ACTIVATION ANALYSIS.** Aihara, H.; Ebihara, M. Tokyo Metropolitan University.

Rare earth elements (REEs) are the most important elements in geochemistry and cosmochemistry. In general, geochemical samples contain higher amounts than those in extraterrestrial cosmochemical samples. In determining REEs, inductively coupled plasma mass spectrometry (ICP-MS) is nowadays commonly applied to geochemical samples even for some cosmochemical samples including chondritic meteorites, whose elemental abundances are essentially the same as those of the solar system. Radiochemical neutron activation analysis (RNAA) can be applied to such solid samples for obtaining reliable data of REE contents. We

have been developing the radiochemical separation procedure of REEs usable in RNAA of geochemical and cosmochemical samples. The procedure consists of alkaline fusion, precipitations of hydroxide and fluoride of REEs and column separations using anion exchange resin and chelate resin. Chemical yields are determined by ICP-atomic emission spectrometry. In RNAA for REEs, La, Ce and Nd can be influenced by neutron-induced fission reactions on U and their contributions need to be evaluated and collected if necessary. In this study, U contents were simultaneously determined by measuring radioactivity of ^{140}Ba and ^{140}La . Applying the procedure developed in this study, REEs could be reliably determined for geochemical peridotite samples and ureilite meteorites, which contain REEs at the level of about 10^{-2} x chondritic level (CI). The present procedure was proved to be effective in determining REEs at the level of CI x 10^{-4} to 10^{-6} , which cannot be determined by any other analytical methods including ICP-MS.

Log: 517. **FISSIONABLE PARTICLE IDENTIFICATION AND ANALYSIS FOR TREATY VERIFICATION AND NUCLEAR FORENSICS.** Morey, MS(1); Russo, RE(2); Havrilla, G(3); Manard, M(4) (1,4) Special Technologies Lab, (2) Applied Spectra, (3) Los Alamos National Laboratory.

Since the release of the genie from the bottle decades ago followed by the end of the Cold War, the threat of nuclear nonproliferation and risk of nuclear terrorism has heightened significantly. An appropriate response to such actions requires quick analysis and screening of trace evidence in the field to determine attribution. This forensic can include the bomb type (Pu or U) and isotopic ratios which could give clues to the fissile material's origin. Advances in laboratory and potentially fieldable analytical tools have made rapid sample attribution on location closer to reality. We have completed preliminary studies pushing the lower size limits of newer and more established analytical techniques such as Raman, XRF, Laser Ablation (ICPMS, Ion Mobility MS), LIBS, and the recently reported Laser Ablation Molecular Isotopic Spectrometry (LAMIS). Initial results of these studies on actinide-containing mineral particles will be presented and show the ability to collect spectral and compositional information on particles below 5 microns. The techniques are complementary and yield a powerful methodology for fissionable particle identification, analysis, and ultimately, determination of lineage.

Log: 518. **PROFILING DNA RECOVERED FROM A RADIATION OR RADIOACTIVITY INCIDENT.** Andrew Hodgson, AWE plc.

The examination and profiling of human DNA recovered from a scene of crime is an essential aspect of any UK criminal investigation. However, it is currently not known whether DNA recovered from a scene where an ionising radiation source or radioactive contamination is present can be successfully profiled. Furthermore, the direct examination and analysis of radioactively contaminated DNA is not possible using the current procedures employed by forensic service providers, due to the risk of operator exposure and the contamination of essential forensic equipment. AWE is therefore putting in place an extensive research and development programme to address this gap in the UK's CBRN capability by enhancing the understanding of the effects that radiation has on the ability to profile human DNA, and assessing the associated retention of different radioactive contaminants within each step of the profiling procedure. A summary will therefore be provided on the project's aims and progress to date; together with a discussion of the lessons that have been learnt during the programmes development.

Log: 519. **VARIABILITY OF PROCEDURAL BLANKS LEADS TO GREATER UNCERTAINTY IN ASSESSING DETECTION LIMITS FOR THE MEASUREMENT OF POLONIUM-210.** Peter Medley (Environmental Research Institute of the Supervising Scientist), Andreas Bollhoefer (Environmental Research Institute of the Supervising Scientist), Paul Martin (Australian Radiation Protection and Nuclear Safety Agency).

Accurate determination of detection limits for the measurement of low-level radioactivity is important for many applications, from environmental tracer studies to the regulation of mining activities. For the determination of Po-210 by alpha spectrometry, factors influencing the detection limit include the sample mass, chemical recovery, detection efficiency, instrumental background, and the chemical or procedural blank. The procedural blank signal can arise from a number of sources, including low-level contamination in the reagents used for chemical separations. Unfortunately, there is little published information concerning the contribution from, and variability in, the blank in alpha spectrometry for most common methods and most radionuclides of interest. Low-level measurements of Po-210 using alpha spectrometry have been conducted at the Environmental Research Institute of the Supervising Scientist (eriss) for many years, on samples with a range of activity concentrations spanning several orders of magnitude. These samples originated from a wide range of research and monitoring projects, and included a wide variety of traditional foods consumed by the indigenous population in Australia's Northern Territory. Many native plant species have relatively low activity concentrations of Po-210 in their edible portions, and effective separation of Po-210 from the sample matrix can be difficult, with the volatility of polonium at relatively low temperatures contributing to this problem. The amount of sample that can be used for analysis is therefore constrained by two main factors – it must be small enough to avoid a reduction in the recovery of Po-210 during chemical separation and source preparation, yet large enough to allow sufficiently accurate and precise quantification of the activity concentration of Po-210. Analysis of eriss' long-term data set of instrument backgrounds and procedural blanks for Po-210 will allow the contribution of procedural blanks to the overall background noise to be better quantified. In turn this will enable better specification of the optimum sample sizes required for a range of environmental samples to produce results with good accuracy and precision from low-level measurements of Po-210.

Log: 520. **ASSESSMENT OF FUKUSHIMA RADIOLOGICAL SOURCE TERM COMPOSITION.** Wimer, N. LLNL.

U.S. Department of Energy radiological measurements of Fukushima Daiichi Nuclear Power Plant releases from March to May 2011 reflect in part ground deposition concentrations across Japan. Dispersed major dose contributors shifted significantly in composition and physical form between early and late response phases. Estimates are made of initial source-term composition as of reactor release time, and of average fuel-assembly burnup reflected in the release. Comparisons are drawn to nominal boiling water reactor radionuclide compositions for volatile and refractory radionuclides. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL-ABS-523917.

Log: 521. **INVESTIGATION OF THE ORIGIN OF ENVIRONMENTAL COMPOUNDS FROM INDOOR AIR SAMPLES VIA ACCELERATOR MASS SPECTROMETRY.** Kretschmer, W, Schindler, M, Scharf, A, Stuhl, A, Physics Institute, University of Erlangen, Germany.

Many organic environmental compounds are potentially dangerous due to their allergic or carcinogen impact on humans. For an effective program to reduce their concentration in houses, their sources have to be detected. Our investigation is focussed on aldehyde compounds since their indoor concentration is relatively high and since they originate from biogenic or anthropogenic sources. Both types of sources can be distinguished by their different ^{14}C content which can be measured via accelerator mass spectrometry (AMS). For the collection and separation of these gaseous substances they have to be converted into liquid or solid phase by derivatization. This leads to the incorporation of up to six additional carbon atoms into the derivatized sample and hence to a reduced ^{14}C content and to an increased uncertainty for the deduced ^{14}C concentration. To minimize the number of additional carbon atoms, different derivatization compounds and methods have been tested with acet- and formaldehyde of known ^{14}C content. The ^{14}C concentration of the calibration samples and from various indoor air samples have been determined by AMS, the corresponding results are discussed with regard to potential sources of aldehydes.

Log: 522. **THE ENEA NOBLE GAS LABORATORY: STATUS OF IMPLEMENTATION.** Baldi, A; Bartolomei, P; Lorenzelli, R; Nava, E; Padoani, F; Rizzo, A; Salvi, S. ENEA.

The strategic value of noble gas capability has been recognized by ENEA. Within the framework of institutional agreements, a noble gas measurement laboratory is under construction for environmental and monitoring purposes, in connection with studies for a near-surface repository for radioactive waste. This research is intended to contribute to the international effort to support CTBT verification and to improve Italian NDC capability. The ENEA noble gas laboratory has three separate components: sampling, extraction and gamma spectrometry analysis. The separation of sampling equipment from the analysis is seen as necessary for the effectiveness of extensive sampling campaigns, as required in monitoring programs. Sampling is based on cryogenic adsorption on activated charcoal particles and the extraction on the temperature-controlled desorption with Helium carrier. The gamma analysis has been carried out so far using a low background HPGe detector (Al endcap with Be window). Some preliminary results and evaluations will be presented on extraction efficiency and nuclide identification. Refurbishment is currently under way to accommodate a more sensitive acquisition system: the goal is a very low background (also thanks to lead-shielding), and higher efficiency by using a different HPGe detector (Al endcap with carbon fiber window). A further improvement will consist of coupling the HPGe detector with a phototube-scintillator system to perform beta-gamma coincidence measurements and anticoincidence with a guard detector to achieve an ultra-low background. The efficiency performance of different materials (i.e. polycarbonate and carbon fiber) for gas sample containers will also be evaluated.