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JRC serving policy and science at the HADES underground research facility

— a casebook

Interdisciplinary nuclear science
applications performed by
JRC-Geel in a 225-m-deep
underground, low-background-
radioactivity lab



Cover illustration: Aurora borealis (Northern lights) at Lofoten, Norway. Time lapse photo by Stein Egil Liland (<https://www.pexels.com/photo/time-lapse-photo-of-northern-lights-1933316/>). Northern lights are created by cosmic ray (mainly protons and alpha particles) and electron interactions with the Earth's atmosphere under certain conditions caused by the solar wind. It is an obvious display of the cosmic rays and from which the radioactivity measurements in the underground research facility HADES are well shielded. It is the protection from these cosmic rays and associated particles that enables the study of minute traces of radioactivity, which is important in a vast number of research fields that this report explains and exemplifies.

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Foreword

Speaking for the international, multidisciplinary team of the Joint Research Centre at Geel (JRC-Geel), it is a privilege for me to present this compilation of work done by the JRC at the low-background underground research facility (URF) high-activity disposal experimental site (HADES) in Mol, Belgium.

Not only has the JRC-Geel team working at the HADES URF been serving the EU and its Member States with commissioned analyses and dedicated reference materials, but it has also contributed to gathering new knowledge with colleagues from different areas of science, e.g. nuclear physics, radioecology and climate science. This research embraces a range of intriguing tasks, from analysing the events at Hiroshima in 1945 to the mapping of ocean currents. This work was strengthened further in 2014, as the JRC lab in HADES became a part of the open access research infrastructure of the JRC and later with the start of the JRC project science applications of radionuclides and actinide materials (SARA). Looking at the output (summarised in the appendices) one can truly call HADES an efficient factory for knowledge production.

This casebook also serves as a celebration of European scientific cooperation, as established by the Euratom Treaty in 1957. More specifically, it will show the reader how non-energy nuclear science applications can serve the science community and the society in many different ways. Most of all, we hope that it can inspire scientists in and new emerging underground laboratories.

Geel, March 2020

Mikael Hult

JRC Team Leader, Radionuclide Metrology

Vice-President of the International Committee for Radionuclide Metrology

Executive summary

In 1992, the Joint Research Centre (JRC) in collaboration with the Belgian nuclear centre StudieCentrum voor Kernenergie Centre d'Etudes Nucleaire (SCK CEN) put a high-purity germanium (HPGe) detector for gamma-ray detection in the 225-m-deep underground research facility high-activity disposal experimental site (HADES). This exploratory work showed that shielding the system from cosmic rays led to a remarkable reduction of the background count-rate in the detector of almost four orders of magnitude. The reduction in background noise opened up a new domain for the analytical technique called gamma-ray spectrometry. By measuring much lower activities than was possible before, a more or less infinite number of new nuclear science applications become possible. One reason for this is that radioactivity is present everywhere and there are 3 500 different radionuclides. Each radionuclide can tell a story, so by detecting and quantifying them we can learn about and trace both anthropogenic activities as well as natural processes.

This report gives a brief overview of work carried out by the JRC in HADES. This work includes the following.

- Detector developments. When the influence of cosmic rays disappears, there are much stricter demands on the radiopurity of the detector and the lead/copper shield around the detector. Furthermore, the quality of the HPGe detector when it comes to size and resolution has improved tremendously the past 20 years.
- Metrology. Quantifying results in Bq is much more challenging when measuring low levels of activities than at normal or high levels.
- Examples of nuclear science applications in the following fields.
 - Part I: Reference materials
 - 1. Rice from Japan and Korea, tea from Turkey
 - 2. Low-level IAEA reference materials
 - 3. The world's least contaminated soil
 - 4. Materials for nuclear decommissioning
 - Part II: Tracer studies
 - 1. Using emissions from Fukushima to trace currents in the Pacific Ocean
 - 2. Hydrothermal vents – uncharted territory of the ocean floor
 - 3. Cold-water corals (*Lophelia*) – the world's oldest living organism?
 - 4. Meteorites – messengers from space

- Part III: Fundamental physics and decay data
 - 1. The lowest decay energy known to humans
 - 2. The most long-lived isomeric state in the universe?
 - 3. Determining the nature of the neutrino
 - 4. Borexino – the least radioactive volume on Earth
- Part IV: Neutron and plasma physics
 - 1. Measuring charged particles inside a tokamak reactor
 - 2. A novel technique for neutron dosimetry, spectroscopy and retrospective assessment
 - 3. A novel technique for measuring neutron cross-section curves
 - 4. Neutron cross-section measurements
- Part V: Environment and industry
 - 1. Uptake of radionuclides in rice and other plants
 - 2. Novel types of concrete with low CO₂ footprint
 - 3. Support to a small or medium-sized enterprise in a semiconductor industry
 - 4. Radioactivity in human bones
 - 5. Geological applications
- Part VI: Radiation protection and emergency response
 - 1. Response to the criticality accident in Tokaimura
 - 2. Solving the Hiroshima enigma
- Part VII: Miscellaneous applications

The different case stories are presented in an order related to neither the magnitude of the project nor the impact. The governing idea has been to be able to explain a case story in an interesting way in 1 or 2 pages. The reader can look at individual case stories at random without having to read all the preceding text.

The underground facility HADES, partly through the JRC open access scheme and its exploratory research programme, has been leading the development of diversifying the science applications that can benefit from underground radioactivity measurements. Many institutes are now looking for existing nearby underground locations that can be used for this type of work, which is much less costly than constructing an underground laboratory from scratch. One can, for example, think of abandoned tunnels that were used as support tunnels during tunnelling work. Alternatives that can be put to use are abandoned mines, bomb shelters, etc.

Seeing that measurements of mBq levels of radioactivity require long measurement times, it is essential to have many detectors available. Judging from current developments in Europe, it is not unlikely that in 10 years from now there could be more than 100 underground HPGe detectors in operation in Europe. Given that researchers from all fields of science have access to such specialised equipment, one can expect the generation of knowledge in many fields to be substantial – as exemplified by the selection of cases in this report. Future developments of underground science are promoted by international collaboration. The network Collaboration of European low-level underground laboratories (CELLAR), for example, is working on promoting (i) mutual use of each other's infrastructure, especially when facing capacity challenges, (ii) sharing experiences of radiopure materials and (iii) sharing experiences linked to the operation of underground facilities.

A brief background

WHAT is HADES?

The high-activity disposal experimental site (HADES) is an underground research facility (URF) at a depth of 225 m, located near the municipality of Mol in northern Belgium. Here, the Joint Research Centre at Geel (JRC-Geel) operates a laboratory for low-background gamma-ray spectrometry and testing.

WHY is this JRC lab located underground?

The JRC's lab is located underground in HADES to minimise the amount of background radiation induced by cosmic rays. Compared with above-ground radioactivity measurements, the instrumental background count-rate is extremely low – in some cases representing as little as 0.02% of the normal reading above ground. These circumstances have made it possible to carry out specialised research in a wide range of projects, as demonstrated by the case studies in this report.

WHICH organisations are involved?

HADES is located at the premises of the Belgian nuclear research centre, SCK CEN, and is operated by the economic interest grouping (EIG) European underground research infrastructure for disposal of nuclear waste in a clay environment (EURIDICE). This unique underground research facility was created in order to develop and study methods for geological disposal of radioactive waste ⁽¹⁾. JRC-Geel rents space in HADES from EURIDICE to operate a gamma-ray spectrometry laboratory.

WHEN did it start?

SCK CEN began the construction of HADES in 1980. In 1992, the radionuclide metrology section of JRC-Geel (in those days the Central Bureau for Nuclear Measurements – CBNM) started a new exploratory research project together with SCK CEN. The aim of the project was to investigate the benefits of performing gamma-ray spectrometry under extremely low-background conditions.

(1) Note that there is no radioactive waste in HADES and never will be, as it will remain a research facility.

WHAT is the purpose of this publication?

This report gives an overview of nuclear science applications projects carried out in HADES since 1999. It aims both to give a historical account ⁽²⁾ to the broad scientific community and to serve as an example for laboratories that wish to start up similar infrastructures. We hope that the case stories in Parts I–VII will stimulate new exciting multidisciplinary work.

Websites

- The JRC's radioactivity laboratory in HADES: ec.europa.eu/jrc/en/research-facility/hades-underground-laboratory
- JRC open access to HADES: ec.europa.eu/jrc/en/research-facility/open-access/relevance-driven/2019-2-rd-eufrat-hades
- EU Science Hub: ec.europa.eu/jrc/en
- SCK CEN: www.sckcen.be
- EIG EURIDICE: www.euridice.be

Figure 1. Watercolour painting of the lift to the first shaft by Hidekazu Yoshida (Japan), 1988.



(2) This report is not a complete list of projects. To keep the report short and attractive to readers with different backgrounds, not many technical data or details are presented. For example, the varying amount of work required for each case study is not discussed. For more details, the reader is referred to the extensive list in Appendix 4.

The Euratom Treaty

The foundation for the international activities of JRC-Geel in HADES is the Euratom Treaty, signed in 1957. Here (and in Appendix 2) we explain how the text is applied.

In March 1957, the Euratom Treaty was signed (at the same time as the Treaty establishing the European Economic Community). It regulates how the Member States of the European Union work together for the peaceful use of nuclear energy – something that was quite ground-breaking at the time it was signed.

Today, there is a broad range of diverse opinions about nuclear energy. Is it a problem, or is it part of a solution when humankind encounters challenges regarding energy provision and environmental issues? As the debate goes on, some things are less politically controversial:

- the European Commission and the Member States must work together on assuring the health and safety of the population (Chapter 3, i.e. Articles 30–39);
- there must be an effective nuclear safeguards system in place;
- “Promotion of research”, also linked to “applications of radioisotopes”.

Those issues are also regulated in the Euratom Treaty.

The work of the JRC is explicitly defined in a number of articles of the Treaty. For the work in HADES, note above all Articles 4, 6, 8 and 30–39 and Annex I. In Appendix 2 in this report, there are extracts from the Treaty and explanations of how these articles relate to work that has been performed in HADES.

Introduction: Measuring low levels of radioactivity at the HADES URF

This report presents a long list of nuclear science applications that have been implemented in HADES over the past 20 years ⁽³⁾. What they have in common is that they are all based on the ability to measure radioactivity in the mBq range. This chapter gives an introduction to these laboratory techniques and the HADES URF.

Radioactivity is all around us

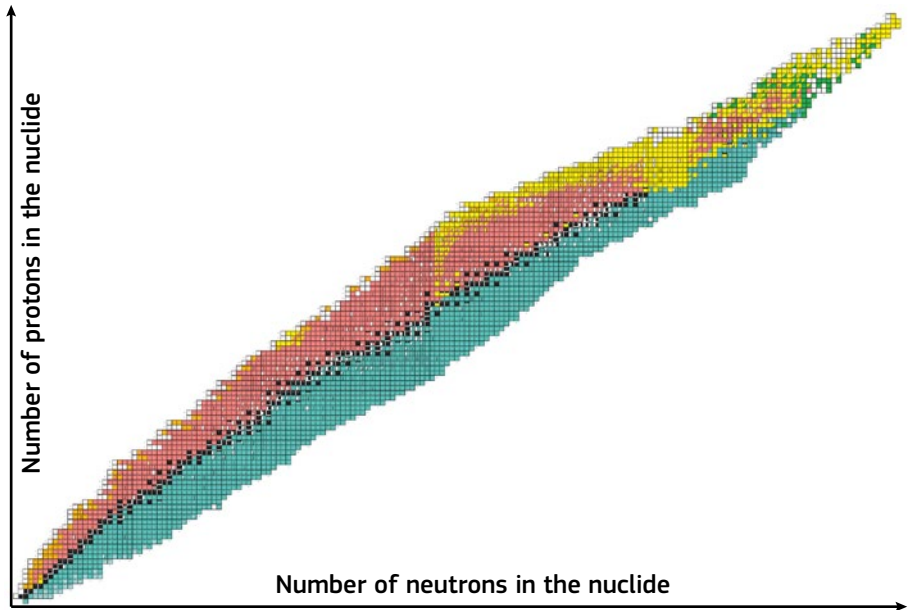
Natural radioactivity is present everywhere and relatively easy to detect as long as the levels are in the order of 1 Bq (the unit of radioactivity is the becquerel, one disintegration per second). However, major complications arise when one attempts to measure activities that are significantly lower, for example in the mBq range.

In most materials around us, as well as in most places on Earth, radioactivity is low and harmless to humans, animals and biota. Still, the detection of minute amounts of radioactivity (in the mBq and even μ Bq ranges) is very important today. For example, such measurements help scientists learn more about how nature works at a global scale (using tracer studies; see Part II). This relates both to natural processes as well as to those induced by human (anthropogenic) activities. It also enables studies of processes in industry to enable more efficient and better use of resources.

A benchmark of what one considers natural levels of activity is the natural concentration of ^{40}K in the human body, which is at a very stable physiological level: 60 Bq/kg. By also considering other natural radionuclides in the human body – such as ^{14}C and ^{210}Pb – one ends up with a natural activity per unit mass of almost 100 Bq/kg. This means that the amount of radioactivity of a person weighing 80 kg is roughly 8 000 Bq. This number is important to remember and relate to both when reading this report (which deals with mBq levels) and when hearing about activity levels reported in media.

(3) Note that the JRC's activities in HADES began in 1992 as exploratory research with one detector. This report focuses on work after 1999, when the work was taken up in the JRC work programme and more resources, for example additional detectors, were made available.

Figure 2. The Karlsruhe nuclide chart.



NB: Only the black squares are stable nuclides; all other squares represent radionuclides, all in all almost 3 500.

See also: https://www.epj-n.org/articles/epjn/full_html/2019/01/epjn180014/epjn180014.html.

The underground lab and the organisations behind it

The HADES URF is operated by the organisation EIG EURIDICE. Since 1999, JRC-Geel has formally rented space in HADES for its low-background gamma-ray spectrometry laboratory.

HADES – WHY IT WAS CONSTRUCTED

In 1974, SCK CEN embarked on a research project to assess the possibility of deep geological disposal of radioactive waste in clay. Back then, the clay layer ⁽⁴⁾, situated between 190 m and 290 m below the SCK CEN site in Mol, was already considered a potentially suitable geological host formation. To investigate the safety and feasibility of geological disposal in plastic clay at great depth, SCK CEN began building an underground research facility in 1980 at a depth of 225 m: HADES. This laboratory was gradually extended over the years and has been managed and operated by EIG EURIDICE since 1995. Since 1999, JRC-Geel has formally rented space in HADES for its low-background gamma-ray spectrometry laboratory.

(4) Called the Boom clay formation after the village of Boom, where the layer reaches the surface. Boom is located between Antwerp and Brussels about 60 km from Mol.

The HADES underground laboratory provides the ideal setting for researching the safety and feasibility of geological disposal. Experts use it to develop and test industrial technologies for building, operating and closing a waste repository in deep clay. Scientists conduct large-scale experiments under realistic conditions in the underground clay formation over a long period of time. The HADES URF is also open at international level to any organisation wanting to carry out *in situ* experiments. A large number of the experiments are carried out in an international context with financial support from the European Commission. Renowned worldwide, HADES is the oldest underground laboratory in the world built in a deep clay formation for studying the possibility of geological disposal. The International Atomic Energy Agency (IAEA) recognises it as a centre of excellence for waste disposal technologies and scientific training.

Note that HADES is, and always will be, a research facility and will never be used as a final repository for radioactive waste.

Figure 3. The way to HADES: looking down shaft 2.



Photo: Mikael Hult. Reproduced with permission from EURIDICE.

EIG EURIDICE

EURIDICE is an economic interest grouping, a joint venture between SCK CEN and the Belgian agency for radioactive waste and enriched fissile materials (ONDRAF/NIRAS). EURIDICE operates the HADES URF and conducts research on the safety and feasibility of geological disposal of high-level and/or long-lived radioactive waste in clay layers. In this way, EIG EURIDICE contributes to the Belgian national disposal programme run by ONDRAF/NIRAS.

After 30 years of research in the HADES URF, EURIDICE has obtained an important expertise concerning the thermo-hydro-mechanical behaviour of the clay, instrumentation techniques and monitoring, and excavation and construction techniques for disposal galleries.

It is impossible to dig tunnels in clay without causing local disturbance of the rock ⁽⁵⁾. A systematic study carried out during the construction indicated that this disturbed area only extends a limited distance into the clay. It was also found that the fissures in the disturbed area close by themselves thanks to the plastic behaviour of clay. As a result, the clay retains its low permeability. This is known as self-sealing.

High-level waste generates heat and, when placed in a repository, it will cause the clay around the disposal galleries to heat up initially. This heat input also affects the hydro-mechanical properties of clay. HADES experiments have made it possible to determine the clay's thermal conductivity and to study and model its corresponding thermo-hydro-mechanical behaviour. The purpose of the ongoing Praclay heater test is to confirm and refine this knowledge on a scale that is representative of a real repository.

EIG EURIDICE has more than 30 years' experience of using various measuring instruments and observation methods in the underground research laboratory and in the Boom clay formation in particular. The know-how acquired regarding the use of these instruments and observation methods will ultimately help ONDRAF/NIRAS to develop a monitoring programme for a real radioactive waste repository.

(5) In geological terms the word 'rock' relates to the medium that surrounds the underground research facility, which in the case of HADES is clay.

Figure 4. The building hosting HADES shaft 2.



Photo: Jan Rypens. © EIG EURIDICE

HADES – SOME TECHNICAL DETAILS

The radiation-shielding properties of the clay and sand surrounding HADES correspond to 500 m water equivalent (w.e.) ⁽⁶⁾. Inside the facility, the soft component and the nucleonic components of the cosmic rays are completely suppressed. What remains is a part of the hard component composed of muons (a heavy version of electrons). This remaining flux of muons is reduced by a factor of 5 000 compared with the conditions above ground.

The facility has the form of a tunnel, 4 m in diameter. On the level where the walking path is located, the width is about 3 m. An important limitation for the installation of detectors is that the central path (1 m wide) needs to be free from permanent installations. This limits the size of detectors that can be installed in HADES. However, for installing ultra low-background high-purity germanium (HPGe) detectors, the space is sufficient and appropriate.

(6) In other words, if all of the medium above HADES were water, a column of 500 m would be necessary to reduce the flux of cosmogenic radiation to the same level as it actually is.

HADES does not provide a clean-room environment but is perfectly adequate for gamma-ray spectrometry. Thanks to the powerful ventilation the average radon concentration is very low ($\sim 6 \text{ Bq/m}^3$). The dose rate in the part of the gallery hosting most of the JRC equipment is between 5 and 10 nSv/h, mainly because of natural radioactivity in the clay and construction materials.

The available space, the vast reduction of cosmic rays, the supply of fresh air and the temperature of 22–24 °C make HADES a comfortable workplace. All in all, it is exceptionally well suited for low-level radioactivity measurements.

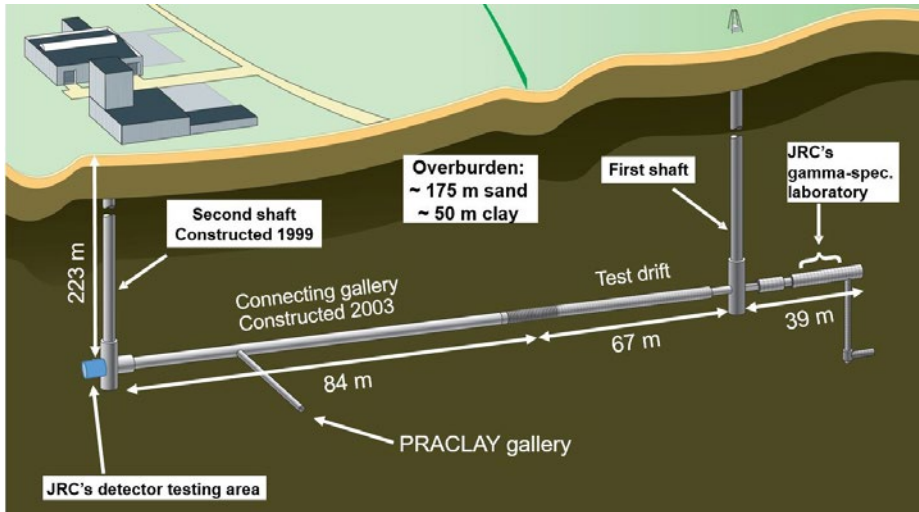
THE RADIONUCLIDE METROLOGY TEAM

Today, the JRC's radionuclide metrology team (RN) is part of Directorate G, Nuclear Safety and Security. The RN specialises in radioactivity measurements and has a large collection of unique instruments in its above-ground laboratories at the JRC-Geel site. Its key task is to realise the unit Bq and support a common system of radioactivity measurements through the Bureau International de Poids et Mesures (BIPM) ⁽⁷⁾. For accurate measurements, it is necessary to use many different instruments, for one obvious reason: no radionuclide decays in the same way as another, and altogether there are about 3 500 radionuclides.

It was after the Chernobyl accident in 1986 that the RN also began working on low-level measurements. At first, work took place only above ground, but in 1992 the RN began an exploratory project together with SCK CEN and placed its first HPGe detector in HADES. This turned out to be a good action and the number of projects that benefited from underground measurements has since grown. The JRC is now renting the area needed for operating its radioactivity laboratory inside HADES from EIG EURIDICE.

(7) Formally it is the BIPM's Consultative Committee for ionizing radiation (CCRI) that JRC-Geel's work supports.

Figure 5. Schematic view of the HADES Underground Research Facility.



© EIG EURIDICE.

Gamma-ray spectrometry

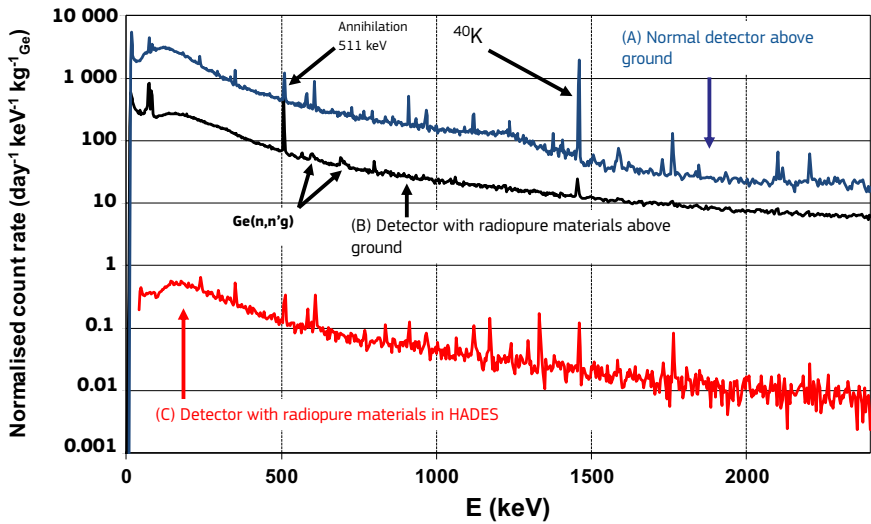
Gamma-ray spectrometry is the most frequently used analytical method in radiometric laboratories today. The JRC lab inside HADES currently uses 12 HPGe detectors.

At present, the radioactivity laboratory that JRC-Geel operates inside HADES uses 12 HPGe detectors. This includes three well detectors, as well as two dual-detector systems.

All detectors and shields are made from specially selected radiopure⁽⁸⁾ materials. The reason is that standard detectors, i.e. the type used above ground (see curve A in Fig. 6), contain materials with levels of natural radioactivity that we cannot accept underground. When the background from cosmic rays disappears, the dominating background will be from natural radioactivity inside the detector. Figure 6 shows gamma-ray background spectra (i.e. spectra collected without any sample or source near the detector) from detectors at different locations. It is obvious how big an improvement it makes to (i) select radiopure material in the detector and (ii) place it underground.

(8) By 'radiopure' we mean that the material has levels of natural radioactivity that can be considered to be far below the natural levels. It often requires several purification steps. An alternative is to look for old material from which the radioactivity has decayed away.

Figure 6. Normalised background spectra from three HPGe detectors. (A) Normal detector above ground. (B) Detector built using radiopure materials, and therefore many of the peaks seen in A have disappeared. (C) Located in HADES and therefore the continuum level is several orders of magnitude lower.



A key task of the RN is to perform reference measurements of radioactivity, covering many different fields in support of stakeholders and EU policies. Accordingly, the HPGe detectors are of different types, to produce optimal results for a number of situations:

- small as well as big samples,
- low as well as high gamma-ray energy,
- radionuclides that are single gamma-ray emitters,
- radionuclides with cascading gamma-rays (i.e. several gamma-rays emitted simultaneously).

Well detectors are optimal for small samples with single gamma-ray-emitting radionuclides. Multi-detector systems are better for radionuclides with cascading gamma-rays such as ^{134}Cs and $^{110\text{m}}\text{Ag}$.

Table 1. An overview of the HPGe detector systems presently operating in HADES

wName	Type	Relative efficiency (%)	Crystal mass (kg)
Ge-3	Coaxial	60	1.0
Ge-4	Coaxial	106	2.1
Ge-5	Planar - BEGe	50	0.8
Sandwich	Two coaxial detectors facing each other	175	3.7
Ge-8	Planar - BEGe	19	0.6
Pacman	Two coaxial detectors facing each other	144	3.5
Ge-12	Well-detector with a small planar crystal behind	n.a.	2.0
Ge-14	SAGE well	n.a.	2.5
Ge-16	SAGE well	n.a.	1.6

It has been shown repeatedly – e.g. during proficiency tests and interlaboratory comparisons – that it is much more difficult to get accurate values while measuring low levels of radioactivity. There are numerous reasons for this (see fact box below).

MEASURING ACCURATELY AT LOW LEVELS – MANY TECHNICAL CHALLENGES

- A smaller number of counts gives higher counting statistical uncertainty.
- A longer measurement time is needed to acquire a sufficient number of counts. This makes the measurement prone to instabilities in the measurement system.
- The background of the detection system is not negligible and can carry significant uncertainty.
- The background radiation changes over time. This includes (i) changes of muon flux, (ii) changes in radon concentration in the air, (iii) changes in background due to handling radioactive sources in the lab, (iv) changes due to decay of radionuclides in the shield, e.g. ^{137}Cs , ^{210}Pb , $^{56,57,58,60}\text{Co}$, $^{54,56}\text{Mn}$, ^{228}Ra , ^{228}Th , (v) changes in background due to the shielding effect of the (massive) sample itself.
- Measuring the sample in close geometry (e.g. placed directly on top of the HPGe detector) will render the coincidence-summing effect very strong.
- If a sample is not homogeneous, it is more critical for accuracy when the sample is located close to the detector.
- Electronic stability over time may not be possible and may result in reduced resolution, which will result in more interferences and higher detection limits.

In a special issue of *Metrologia* (the BIPM's own journal), Hult (2007) published a review on low-level gamma-ray spectrometry in which he defines the term 'low-level' that is used for low-level radioactivity measurements. It must not be confused with, for example, low-level radioactive waste measurements, which concern an entirely different radioactivity level. In another review paper, Hult and colleagues (2006) define the field of underground gamma-ray spectrometry, explaining, for example, why at least 10 m w.e. overburden (e.g. 4 m concrete or 1.5 m steel) is necessary to qualify for the use of the term 'underground'.

FURTHER READING: UNDERGROUND GAMMA-RAY SPECTROMETRY

- M. Hult, 2007, 'Low-level gamma-ray spectrometry using Ge detectors', *Metrologia*, Vol. 44, pp. S87–S94; Erratum, *Metrologia*, Vol. 44, p. 425.
- M. Hult et al., 2003, 'Underground measurements of environmental radioactivity in Europe', in: Naoto Kamata (ed.), *Proceedings of the International Symposium of the Kanazawa University 21st. Century COE Program, Kanazawa Univ. (Publs.)*, Vol. 1, pp. 18–23,
- M. Hult et al., 2006, 'Underground gamma-ray spectrometry', *Acta Chimica Slovenica*, Vol. 53, pp. 1–7.
- M. Laubenstein et al., 2004, 'Underground measurements of radioactivity', *Applied Radiation and Isotopes*, Vol. 61, pp. 167–172.
- JRC, 2020, video: [Youtube: Science in the deep underground \(https://www.youtube.com/watch?v=CqcT9nyOhZA\)](https://www.youtube.com/watch?v=CqcT9nyOhZA)

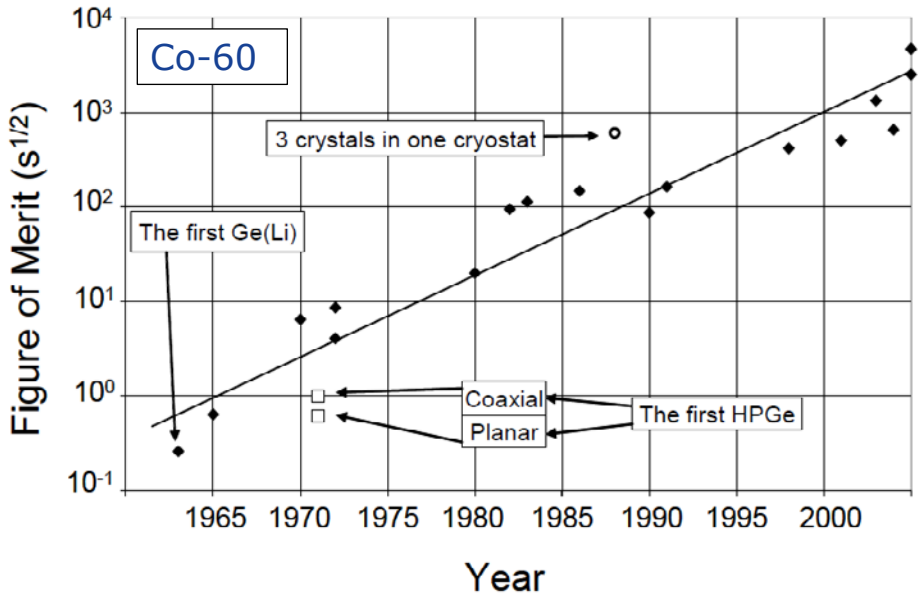
JRC-GEEL HAS DEVELOPED A NUMBER OF NOVEL DETECTOR SYSTEMS

In a review article in *Metrologia*, Hult (2007) defines the first low-level measurements as Willard Libby's ground-breaking measurements of ^{14}C in archaeological materials. For this, Libby received the Nobel Prize in 1960. He used Geiger–Müller tubes surrounding his proportional counter to detect the signal from cosmogenic radiation (muons).

Ever since, tremendous development has taken place in the field of low-level measurement techniques. Perhaps the single most important development was beginning to use underground laboratories. The first reported underground radioactivity measurements, according to Hult and colleagues (2006), took place in a derelict coal mine beneath Glasgow University, at a depth of 30 m.

Figure 7 shows the development of a FoM for Ge detectors that is related to the reciprocal of the detection limit: the higher the FoM, the lower activities can be measured. A similar development has taken place for other types of detectors. The most noteworthy are of course the large scale (kilotonnes) detector systems used to detect and search for neutrino interactions, dark matter interactions, double beta decays and other rare events.

Figure 7. The development of a figure of merit (FoM) for Ge-detectors using data found in scientific articles and combined with model calculations. The higher the FoM is, the lower activities can be measured.



Source: Hult et al. (2006). Printed under Creative Commons licence 4.0: <https://creativecommons.org/licenses/by/4.0/>.

FURTHER READING: GAMMA-RAY INSTRUMENTATION

- E. Andreotti et al., 2014, 'Determination of dead-layer variation in HPGe detectors', *Applied Radiation and Isotopes*, Vol. 87, pp. 331–335.
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- M. Hult, 2007, 'Low-level gamma-ray spectrometry using Ge detectors', *Metrologia*, Vol. 44, pp. S87–S94; Erratum, *Metrologia*, Vol. 44, p. 425, <https://doi.org/10.1088/0026-1394/44/4/S12>.
- M. Hult et al., 2006, 'Underground gamma-ray spectrometry', *Acta Chimica Slovenica*, Vol. 53, pp. 1–7, <http://acta-arhiv.chem-soc.si/53/53-1-1.pdf>.
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- J.S.E. Wieslander, 2009, 'A new gamma-ray spectrometry system for measurements of radioactivity in the micro-becquerel range', PhD thesis, Dept. of Physics, University of Jyväskylä, Jyväskylä, Finland.

Figure 8. Interior view of the JRC's gamma-ray spectrometry laboratory inside HADES.



Photo: Guillaume Lutter.

JRC-Projects at the HADES URF, 1999–2019 – a casebook

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The JRC activities of HADES

The main activities at HADES are chiefly of two types:

- supporting JRC projects (on neutron physics, climate change, international harmonisation, reference materials, etc.) performed either as exploratory research or in support of EU policies,
- serving scientists from Member State laboratories, either by providing access to use the detector systems or through collaboration in European projects.

The JRC's work programme is approved by the European Council as part of the framework programmes. In the past framework programmes, actions linked to nuclear safety and security were of paramount importance, and they remain so. The present programme, Horizon 2020 ⁽⁹⁾ (H2020), covers the 7-year period 2014–2020. In this, JRC actions towards studies of climate change are emphasised for the first time. This is even more pronounced in the proposed next research and innovation framework programme, Horizon Europe.

The types of external projects have varied over the years depending on priorities in the work programme. In the past in HADES, there have been (i) projects carried out for payment (third-party work), (ii) support to small and medium-sized enterprises (SMEs) and (iii) participation in what are called indirect actions, i.e. European research projects funded by the European Commission's Directorate-General for Research and Innovation ⁽¹⁰⁾ or the European Association of National Metrology Institutes (EURAMET) ⁽¹¹⁾, of which the JRC is an associate member. However, in H2020 the open JRC access scheme has been a high priority.

The examples (case stories) given in this report are taken from all types of projects, direct actions, indirect actions and open access projects.

(9) <https://ec.europa.eu/programmes/horizon2020/en>.

(10) https://ec.europa.eu/info/research-and-innovation_en.

(11) <https://www.euramet.org>.

Part I: Reference materials

Certifying or confirming the radioactivity in reference materials, such as food or sediment

A reference material (RM) contains a well-established activity of certain radionuclides. It must be very similar or identical to the samples that a laboratory normally analyses (milk, vegetables, plants, meat, etc.). Furthermore, it has been meticulously characterised, is homogeneous and has proven stability over time.

RMs are used for validating measurements and laboratory procedures and are therefore indispensable in any modern laboratory. Many laboratories produce their own reference sources, e.g. by spiking liquid acid solutions with commercially available radionuclides. Although such reference sources are useful for calibrations, they are not suitable for method validation, as they differ from the samples that the laboratory normally analyses (unless the laboratory normally measures acid solutions, of course). For high-quality analyses, it is crucial to have access to RMs with a similar matrix and density to those of the real samples.

JRC-Geel is equipped with a world-leading laboratory for the production and certification of certified reference materials (CRMs), following ISO standard 17034. The cooperation between the RM unit and the RN, which has a primary standardisation laboratory, creates a perfect platform for these activities. As an example, the RM unit has produced a CRM of bilberries harvested close to Chernobyl (JRC-Geel, IRMM-426 ⁽¹²⁾). Furthermore, a great number of RMs (maize, hay, water etc.) have been produced to support the proficiency tests that the JRC carries out for European monitoring laboratories in support of Euratom Treaty Article 35 (Hult et al., 2019) and for method validation for standardisation organisations (Sobiech-Matura et al., 2018).

Several reports point out that there is a lack of reliable CRMs in the world (Jerome et al., 2015). To produce such materials following ISO 17034 is a lengthy and expensive process, which is rarely profitable for a commercial enterprise and requires special capabilities. The global key producers are the IAEA and some of the major national metrology institutes. In addition, some other national laboratories (France, Japan, Korea, the United Kingdom) produce CRMs, but mainly to support their national monitoring labs.

Although most CRMs have activities that can be measured above ground, it is of course an asset to certify activity in a laboratory where the background can essentially be discarded. The data produced in such an environment are far more robust. The HADES laboratory has been used for certifying several reference materials that have inherently very low activities. One example is the glass beads used in the SuperNEMO collaboration (Povinec et al., 2016). A few more examples will follow in this casebook.

(12) <https://crm.jrc.ec.europa.eu/p/40455/40460/By-material-matrix/Plant-materials/IRMM-426-WILD-BERRIES-radionuclide-activity-concentration/IRMM-426>.

Figure 9. A wide-angle view of the central area of the JRC's RM-processing hall.



Photo: JRC.

FURTHER READING: REFERENCE MATERIALS

- T. Altitzoglou et al., 2004, 'Characterisation of reference materials for radioactivity with assigned values traceable to the SI units', *Applied Radiation and Isotopes*, Vol. 61, pp. 395–399.
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- M. Hult, V. Jobbagy, K. Sobiech-Matura, 2019, *How the JRC verifies the quality of results from radioactivity monitoring within the European Union*, Publications Office of the European Union, Luxembourg, (<https://dx.doi.org/10.2760/124550>).
- K. Sobiech-Matura et al., 2018, *Report on interlaboratory validation of gamma-ray spectrometry measurements*, Joint Research Centre, Geel.
- P.P. Povinec et al., 2016, 'Reference material for natural radionuclides in glass designed for underground experiments', *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 307, pp. 619–626.
- JRC-Geel, reference materials catalogue, 2019 (<https://crm.jrc.ec.europa.eu/>)

Reference materials, Case 1: Rice from Japan and Korea, tea from Turkey

In cooperation with three countries, scientists at HADES performed measurements that contributed to the certification of reference materials. The cooperation also helped establish international equivalence and comparability.

JAPAN

The nuclear accident in Fukushima, caused by the enormous earthquake and tsunami in March 2011, triggered a number of actions in Japan. To ensure safe food products, many new radiometric laboratories were started. To guarantee the quality of results from such laboratories, Japanese authorities briskly set up a quality control scheme. Accordingly, scientific institutes in different places swiftly began producing reference materials for these labs.

Although producing reference materials is normally a time-consuming procedure, several materials had to be developed against a tight deadline. It was important for Japan to seek unbiased confirmation of the reference values for these materials. The JRC contributed to this work by measuring several materials. As an example, the National Metrology Institute of Japan, together with its national food research institute, produced a brown rice RM⁽¹³⁾. The rice was sampled from fields contaminated by the Fukushima accident and contained both ¹³⁴Cs and ¹³⁷Cs in measurable amounts.

REPUBLIC OF KOREA

Most of the radioactivity from Fukushima entered the North Pacific Ocean, and was diluted and transported eastwards. However, some radioactivity was emitted into the air and transported westwards. As the Korean peninsula was left fairly unaffected, there was no need to start up new laboratories for monitoring, like in Japan. Still, the Korean Atomic Energy Research Institute took a similar action to Japan's and developed a rice RM for which it also sought international unbiased confirmation of the reference values⁽¹⁴⁾. It should be noted that the ¹³⁷Cs in the Korean material was added to the topsoil in the greenhouse where the rice was cultivated. This was necessary, as there was no contamination on the fields of the Korean peninsula.

(13) Interestingly, this work also contributed to better establishing international equivalence of the unit Bq: it was endorsed by the CCRI (Section II, measurements of radionuclides) of the BIPM as a supplementary comparison (No APMPRI(II)-S3.Cs-134.Cs-137).

(14) This work also contributed to better establishing international equivalence, as it was endorsed by the CCRI(II) of the BIPM to be a supplementary comparison (No CCRI(II)-S9 of 2013).

TURKEY

In the context of Turkey's close association with the EU, the JRC conducted a project called Europe and metrology in Turkey ⁽¹⁵⁾ between 2009 and 2012. The aim was to contribute to the better functioning of the EU–Turkey Customs Union. In the field of ionising radiation it is also evident that neighbouring countries mutually benefit from each other's reliable and robust measurements for radiation safety, emergency response and science. Scientists from the Turkish Atomic Energy Authority (TAEK) working both in Istanbul and in Ankara spent many months of training at JRC-Geel.

TAEK is now running a quality control scheme for Turkish environmental monitoring labs similar to what the JRC is doing for the EU Member States (Hult et al., 2019). A key activity is to organise proficiency tests. In the second proficiency test ever to be organised by TAEK, the JRC provided reference values. The material was black tea that grew in Turkey following the Chernobyl accident and consequently took up ¹³⁷Cs and ⁹⁰Sr.

FURTHER READING: RICE AND TEA REFERENCE MATERIALS

- BIPM supplementary comparison Korean rice, (<https://www.bipm.org/kcdb/comparison?id=1497>)
- BIPM supplementary comparison Japanese brown rice, (<https://www.bipm.org/kcdb/comparison?id=344>)
- M. Hult, V. Jobbagy, K. Sobiech-Matura, 2019, *How the JRC verifies the quality of results from radioactivity monitoring within the European Union*, JRC TECHNICAL REPORTS Joint Research Centre EUR 29509 EN (2019). (<https://dx.doi.org/10.2760/124550>).
- E. Yeltepe et al., 2018, 'A review of the TAEA proficiency test on natural and anthropogenic radionuclides activities in black tea', *Applied Radiation and Isotopes*, Vol. 134, pp. 40–44.

(15) (<https://ec.europa.eu/jrc/en/training-programme/europe-and-metrology-turkey>).

Reference materials, Case 2:

Low-level IAEA reference materials – mussels, shrimp and ocean sediment

The JRC and IAEA have a long-standing collaboration ⁽¹⁶⁾ in the nuclear field. Through the years, they have supported each other's work in producing certified reference materials. The common overarching goal is to better protect citizens and society from the dangers of ionising radiation.

A vital component for making good measurements is to have access to calibration sources and certified reference materials. IAEA has a mandate from member states all over the world to produce reference materials within its programme for reference products for science and trade. Within this context, IAEA has requested contributions to the certification of its low-level environmental reference materials from underground laboratories (such as HADES) that collaborate within the Collaboration of European low-level underground laboratories (CELLAR) network (see chapter 'Outlook').

MUSSELS AND SHRIMP

An example of a low-level reference material produced by the IAEA was Mediterranean mussels, collected close to Marseilles in France. Mussels are important natural archives and can provide information on changes in the surrounding water mass. Currently, IAEA is producing a shrimp RM for which it has asked for a HADES measurement.

SEDIMENT

One sediment RM was collected from a water depth of 4 500 m offshore from Bikini Atoll and another sample at a depth of 5 600 m, in the Pacific Ocean, just northwest of the Marshall Islands (Pham et al., 2016). These materials are important, as they contain anthropogenic radionuclides from nuclear weapon testing in the 1950s and 1960s. The sediment from Bikini Atoll contains higher levels of plutonium and americium than the other one, which on the other hand contains more ¹³⁷Cs. It is important for the monitoring labs to be able to measure these radionuclides at low activities.

(16) Within this collaboration, the IAEA has supported the certification of several JRC RMs with measurements both in its underground laboratory in Monaco as well as above ground in Seibersdorf (Austria).

FURTHER READING: IAEA REFERENCE MATERIALS

- M.K. Pham et al., 2010, 'A new reference material for radionuclides in the mussel sample from the Mediterranean Sea (IAEA-437)', *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 283, pp. 851–859.
- M.K. Pham et al., 2016, 'Certified reference materials for radionuclides in Bikini Atoll sediment (IAEA-410) and Pacific Ocean sediment (IAEA-412)', *Applied Radiation and Isotopes*, Vol. 109, pp. 101–104.

Figure 10. The Baker explosion, part of Operation Crossroads, a nuclear weapon test by the US military at Bikini Atoll, Micronesia, on 25 July 1946.



Source: United States Department of Defense. Public domain, <https://commons.wikimedia.org/w/index.php?curid=6931019>.

Reference materials, Case 3: The world's least contaminated soil

In specific locations in Peru, radioactive contamination is extremely low. This case story demonstrates that when it comes to radiation protection standards, National Metrology Institutes need to collaborate on a global scale.

The atmospheric testing of nuclear weapons in the 1950s and 1960s dramatically increased the levels of radioactive fallout all over the globe. This triggered the major states running a nuclear weapons program to sign the Partial Test Ban Treaty (PTBT) in 1963, which prevented testing in the atmosphere, at sea and in space. The Comprehensive Test Ban Treaty (CTBT) followed in 1996. It finally prevented the signing parties to carry out *any* nuclear weapons test explosion.

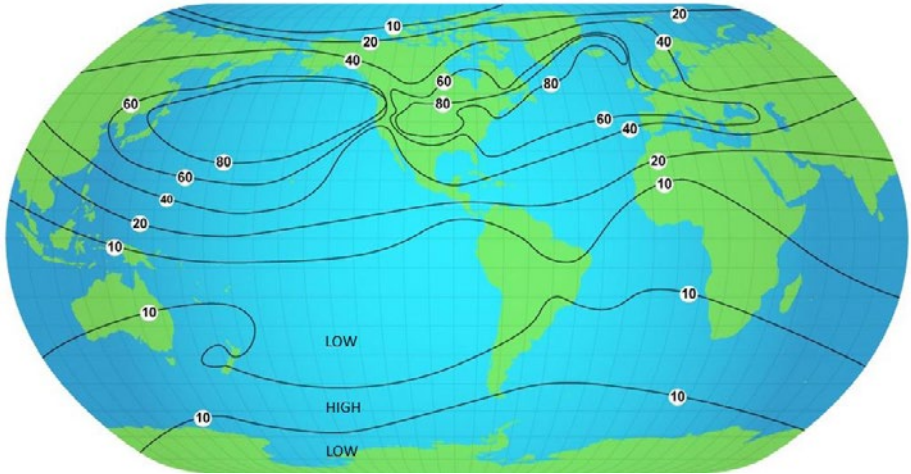
The effects of the PTBT were quickly perceived, as the activities of radionuclides like ^{137}Cs and ^{14}C started to decrease. Today, they are more or less back to the levels found before 1955. The fallout was not distributed homogeneously over the globe, due to air movements in the stratosphere and climatic conditions at different locations, e.g. the amount of precipitation. In general, the northern hemisphere received more fallout than the southern hemisphere.

Interestingly enough, at some specific locations in Peru, the concentration of fallout plutonium was extremely low (Hardy et al, 1973). The US National Institute for Standards and Technology (NIST) made use of this fact to produce a soil reference material based on Peruvian soil. Ever since, this material has served the radioanalytical community world-wide as an anthropogenic blank soil. In 2009, the material was sold out and NIST decided to produce a new batch. In the process, JRC-Geel was asked to perform low-level measurements in HADES. Other materials for which JRC has contributed to NIST certification are ocean shellfish (Altzitzoglou et al., 2000) and bone ash (Lin et al., 1998).

REFERENCES, LEAST CONTAMINATE SOIL

- T. Altzitzoglou, Low-level radioactivity measurements in an ocean shellfish matrix, *Applied Radiation and Isotopes*, 52, (2000), 539-544.
- Z. Lin et al., Development of the NIST Bone Ash standard reference material for environmental radioactivity measurement, *Applied Radiation and Isotopes*, 49, (1998), 1301-1306.
- E.P. Hardy et al., Global inventory and distribution of fallout plutonium. *Nature* 241 (1973) 444-445.

Figure 11. The curved lines are so-called isolines of cumulative ^{90}Sr deposits, based on analysis of soils collected 1965-67.



NB: The unit is mCi/km^2 , which equals $37 \text{ Bq}/\text{m}^2$. Note that the values in the northern hemisphere are higher than those in the southern hemisphere.

Source: UNSCEAR 1969 Report of the United Nations Scientific Committee on the Effects of Atomic Radiation- Annex A: Radioactive Contamination of the Environment by Nuclear Tests. Reproduced by permission of UNSCEAR. Colour version by Christopher Havenga (JRC).

Reference materials, Case 4:

Materials for nuclear decommissioning – a high-level material requiring low-level measurements

When dismantling nuclear facilities, materials from the decommissioned installations need to be tested before recycling. JRC-Geel developed a steel tube RM to aid this work.

Nuclear decommissioning is a hot topic in Europe today, as many old reactors are reaching the end of their designed lifetime. When a nuclear facility is dismantled, most materials are not contaminated or radioactive and can be recycled. It is, however, crucial that the methods for testing all materials (concrete, metals, plastic, etc.) be robust, as no contaminated materials must reach the general public. However, non-contaminated materials must not be treated as radioactive waste, as this handling is very costly.

Together with European partners, JRC-Geel developed (among other materials) a steel tube RM suitable for validating free-release measurement facilities. The tubes contain radioactive cobalt and silver, and the total activity for one steel tube is in the order of 100 kBq. They are produced using centrifugal casting. During this process, cobalt forms a solid solution in the material. However, that is not the case for silver.

To verify the distribution of silver inside each tube, swarf (chips) was taken from different locations. Owing to the small mass of these samples, low-level measurements were required. The results showed that cobalt was homogeneously distributed, while silver showed a small relative increase of about 3 % on the inside, compared with the outside. This was, however, within the design goals. The distribution along the tube axis, however, was homogeneous.

FURTHER READING: MATERIALS FOR NUCLEAR DECOMMISSIONING

- M. Hult et al., 2016, 'Distribution of radionuclides in an iron calibration standard for a free release measurement facility', *Applied Radiation and Isotopes*, Vol. 109, pp. 96–100.

Figure 12. A free-release measurement facility in which pallets filled with, for example, concrete or metal scrap can be tested for contamination.



NB: The facility was developed by the Czech Metrology Institute and Envinet within the EURAMET (EMRP)⁽¹⁷⁾ project Metrology for Radioactive waste Management, in which JRC was a partner. Photo: Mikael Hult.

Figure 13. Calibration standard for a free-release measurement facility. Left: a pallet filled with the reference tubes. Right: radioactive sources can be inserted in the pallet or in the radiopure steel spheres (pétanque balls in this case) to simulate different types of contamination.



Photo: Mikael Hult.

(17) European Metrology Research Project: MetroRWM (<https://www.euramet.org/research-innovation/emrp/?L=0>)

Figure 14. Interior view of JRC's gamma-ray spectrometry laboratory in HADES.



Photo: G. Lutter.

Part II: Tracer studies

Using naturally or accidentally emitted radioactivity as a tracer

Radioactive substances can be traced, even when they are far from their source or have been extensively diluted. Thus, they can tell stories of processes and past events.

For reasons of safety and ethics, this is hard to exploit in a planned experiment that includes the emission of radioactivity to the environment. However, there is great potential (and even a moral obligation) to use naturally or accidentally emitted radioactivity as a tracer. One example relates to the atmospheric nuclear weapon testing in the 1960s and 1970s from which fallout generated radioactive contamination worldwide. However, it also generated scientific knowledge about stratosphere–troposphere interchange processes. The fallout of ^{137}Cs from the same weapon tests can be used for establishing a time scale in samples such as Greenland ice cores that are important for many scientific studies ⁽¹⁸⁾. This chapter will show some more examples.

Figure 15. A Danish ice-core-drilling system in Greenland.



Photo: William Colgan, Geological Survey of Denmark and Greenland.

(18) (<http://www.campcenturyclimate.dk/ccc/>).

Tracer studies, Case 1:

Using emissions from Fukushima to trace currents in the Pacific Ocean

As ocean currents are one of the main driving forces of the climate, it is crucial to collect more knowledge about them. The Fukushima disaster opened up an unexpected opportunity to carry out global tracer experiments. Here, JRC researchers collaborated with both Japan and the United States.

The Fukushima nuclear accident in March 2011 led to the emission of several PBq⁽¹⁹⁾ of ¹³⁴Cs and ¹³⁷Cs⁽²⁰⁾. Most of the radiocaesium ended up in the North Pacific Ocean, but a limited amount was also emitted into the atmosphere.

This led Japan to start large-scale monitoring programmes of seawater and seafood. However, the radioactivity – which had high levels very close to the nuclear power plant – was quickly diluted by the huge water masses of the Pacific Ocean when transported away from the coast. At sea, it can serve as a tracer, making it possible to follow the movement of water masses and, accordingly, get more detailed information about ocean currents.

The exact nature of ocean currents is not very well known. They are complex and can change with time. Still, they are extremely important for life on Earth, as they transport huge amounts of heat and nutrients. To a large extent, they govern the climate, as well as aquatic life. By better understanding ocean currents, scientists may improve the computer models that describe how the climate changes over time.

The activity of radiocaesium in some parts of the Pacific – even far from Fukushima – reached levels of about 1 Bq/m³. However, it is not realistic to collect samples of 1 m³, particularly since commercial cargo ships were also asked by Japanese scientists to contribute to the collection of samples. Instead, they typically collected 2 litres from each sampling location. This means that one can expect approximately 2 mBq of radiocaesium in each sample – something that is very difficult to detect, unless one uses an underground laboratory.

Professor Michio Aoyama (Fukushima University and the Oceanographic Society of Japan) was in charge of a major operation collecting samples from all over the northern Pacific Ocean. He collaborated with Yosinoru Hamajima (Kanazawa University), who operates a shallow underground laboratory. However, owing to the large number of samples – some with very low activities – a collaboration started with JRC-Geel in order to measure samples in HADES.

The measurement campaign was successful and resulted in publications, which garnered praise as well as prizes (both the prestigious Hidoka medal and the

(19) Petabecquerel = 10¹⁵ Bq.

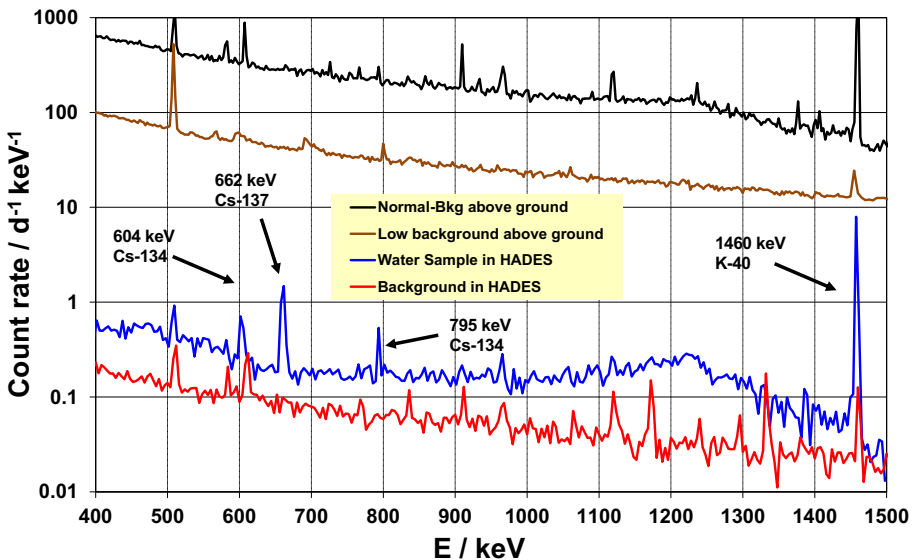
(20) Equal amounts of the two caesium radionuclides, as measured in Bq, were released in March 2011 (activity ratio: 1.0).

Udo medal) for Professor Aoyama. Furthermore, the article presented at the ICRM conference in Vienna in 2015 was the most downloaded from the journal *Applied Radiation and Isotopes* for a long time (Aoyama et al., 2016b). In addition, through the JRC open access programme, samples collected by the renowned Woods Hole Oceanographic Institution were analysed (Macdonald et al., 2020).

Some key conclusions of the study were the following.

- It was confirmed that surface water is subducted, down to a depth of 400 m, in an area close to the international date line.
- It was possible to make a better estimation of the total release of radioactivity from Fukushima.
- More knowledge was gained about the dynamics of eddy currents that appear near to coastal regions. Such current can have big impacts locally and may explain why, for example, schools of fish move in certain directions.

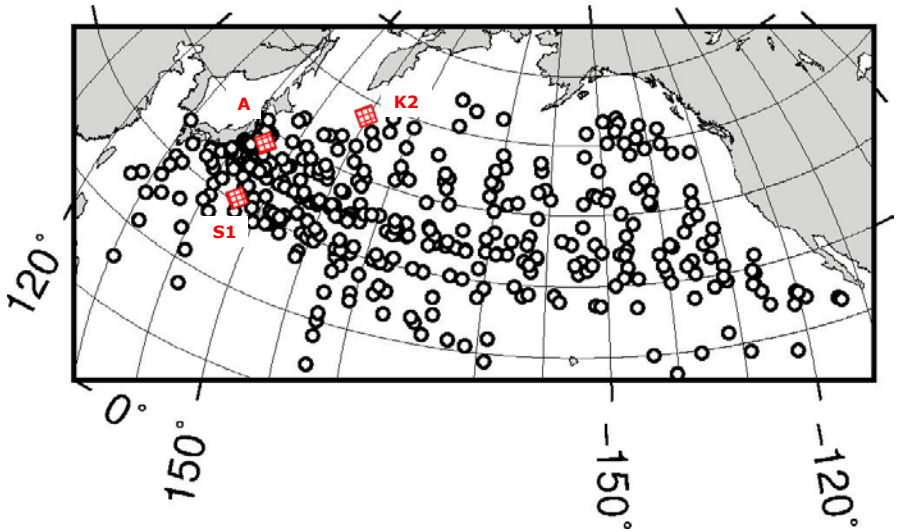
Figure 16. Gamma-ray spectra according to key in the figure.



NB: Bkg, background.

Source: Lutter et al. (2015, p. 547). Reproduced under Creative Common licence with *Nukleonika*.

Figure 17. Map of the Pacific Ocean with the 2011 and 2012 locations for surface sampling of water (black circles). Three locations for sampling plankton and particulate matter (red squares) are also given.



Source: Aoyama et al. (2016b, p. 436). Reproduced under Creative Commons licence 4.0 (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

FURTHER READING: USING FUKUSHIMA EMISSIONS TO TRACE CURRENTS

- M. Aoyama et al., 2016a, ' ^{134}Cs and ^{137}Cs in the North Pacific Ocean derived from the March 2011 TEPCO Fukushima Daichi Nuclear Power Plant accident, Japan. Part one: surface pathway and vertical distributions', *Journal of Oceanography*, Vol. 72, pp. 53–65.
- M. Aoyama et al., 2016b, 'Tracing radioactivity from Fukushima in the Northern Pacific Ocean', *Applied Radiation and Isotopes*, Vol. 109, pp. 435–440.
- G. Lutter et al., 2015, 'Measurement of anthropogenic radionuclides in post-Fukushima Pacific seawater samples', *Nukleonika*, Vol. 60, No 3, pp. 545–550.
- A. Macdonald et al., forthcoming, 'A Fukushima tracer perspective on four years of North Pacific mode water evolution', submitted to *Deep-Sea Research Part I*.

Tracer studies, Case 2:

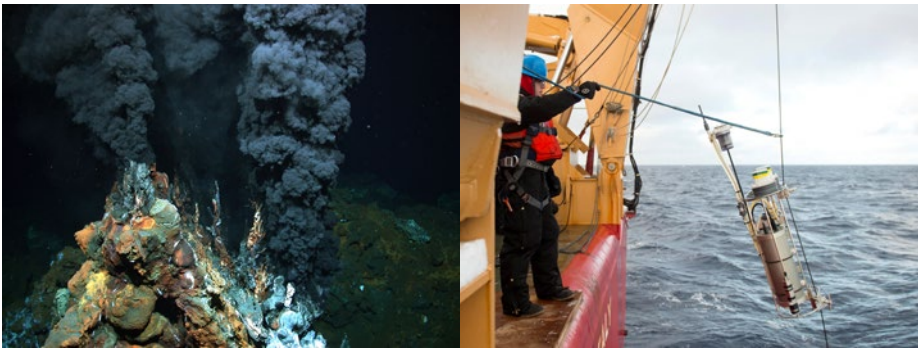
Hydrothermal vents – uncharted territory of the ocean floor

By measuring low levels of radium and thorium isotopes, JRC-Geel was able to help oceanographic researchers follow the plumes from hydrothermal vents. To detect them, a new type of HPGe well detector was used.

At the bottom of the oceans, along the edges of the tectonic plates, are hydrothermal vents, also known as black smokers. These are natural chimneys where hot, mineral-rich fluids enter the oceans from Earth's crust below. These naturally buoyant fluids can travel the oceans at great depths for decades.

Until now, not much is known about these fluids and their impact on marine life and the environment. Nevertheless, it is becoming increasingly clear that they have a great impact on ocean chemistry, due to their richness in minerals. Iron, for example, is important for photosynthesising organisms, which assimilate CO₂ from the atmosphere. Accordingly, by studying the emissions from hydrothermal vents we can learn more about the oceans' role as a buffer for atmospheric CO₂ and regulator of Earth's climate.

Figure 18. Left: photo of hydrothermal vent (black smoker) at depth of 2 980 m in the Mid-Atlantic Ridge. Right: photo from the GEOTRACES cruise GP16 with the McLane pump and the MnO₂-coated cartridge being deployed for collecting samples to be measured in HADES.



Sources: Left: photo from MARUM – Centre for Marine Environmental Sciences, University of Bremen (CC-BY 4.0). Right: photo by Cory Mendenhall, US Coast Guard.

In 2013, a US-led research cruise ⁽²¹⁾ towards the East Pacific Rise (circa 3 200 km west of Chile) followed a specific hydrothermal plume. At a depth of about 2 500 m, the plume travelled 4 300 km through the South Pacific.

(21) GEOTRACES expedition TN303, EPZT, GP16, (<http://geotraces.org>).

To monitor the travelling plume, the scientists aboard the research vessel used four radium isotopes with masses 223, 224, 226 and 228 as nuclear clocks. These four isotopes have very different half-lives: 11 days, 4 days, 1 600 years and 2 years, respectively. By measuring how their activity changes with the distance from the vent, one can determine for how long the plume has been travelling, affecting the surrounding water masses on its way.

Within the JRC-Geel open access programme, 30 measurements of hydrothermal plume samples were conducted in HADES in 2016. Because of the great impact worldwide of climate change studies, a continuation of this project was carried out under the JRC institutional programme in 2019.

A key finding from this study is that it is possible to detect very low amounts of ^{228}Ra . In the plumes, ^{228}Ra has a level of activity about a factor of 1 000 lower than that of ^{226}Ra , which makes detection very difficult. JRC studied the optimal conditions for detecting this radionuclide and concluded that it was possible, using a new type of detector, called a SAGe well detector. The measurement results obtained were used to determine the age of the plumes. It was also possible to detect new vents owing to an increase in the ^{228}Ra activity at a certain location. Furthermore, the samples in the water column above the hydrothermal plume showed a general decrease of activity with depth.

FURTHER READING: HYDROTHERMAL VENTS

- M. Hult et al., 2019, 'Underground gamma-ray measurements of radium isotopes from hydrothermal plumes in the deep Pacific Ocean', *Applied Radiation and Isotopes*, Vol. 153, <https://doi.org/10.1016/j.apradiso.2019.108831>.
- F. Pavia et al., 2019, 'Timescales of hydrothermal scavenging in the South Pacific Ocean from ^{234}Th , ^{230}Th , and ^{228}Th ', *Earth and Planetary Science Letters*, Vol. 506, pp. 146–156, <http://doi.org/10.1016/j.epsl.2018.10.038>.

Tracer studies, Case 3:

Cold-water corals (*Lophelia*) – the world’s oldest living organisms?

Another type of natural archive is corals. Cold-water corals of the Atlantic hold information for a better understanding for Earth and its ecosystems. JRC-Geel has been cooperating with German and Danish scientists.

Just like trees, corals can be seen as a natural archive. Most people would associate the word ‘corals’ with tropical underwater scenery with colourful fish. However, there are also cold-water corals, for example the species *Lophelia pertusa*. These are to be found in the Atlantic at depths down to 3 000 m.

Radiocarbon dating has shown that these coral colonies can be as old as 40 000 years and individuals over 1 000 years old. As living systems, these deepwater reefs can thus be considered Earth’s oldest living organisms. Accordingly, they may serve as an excellent natural archive that we must do our utmost to preserve.

In an open access project, the Alfred Wegener Institute, Bremen, and Bremen University have asked for the first ever underground measurements of cold-water corals in HADES. The aim of the measurements is to perform nuclear dating of each sampled layer (only about 1 g of material per sample) by measuring two radionuclides: ^{226}Ra and ^{210}Pb . As the latter is the last radionuclide in the ^{226}Ra decay chain, the ratio of the activities of the two will provide a means to establish the age of different parts of the coral reef structure.

In addition, the institute and university, in collaboration with the Danish Technical University, will measure other isotopes using mass spectrometric techniques. This serves to study environmental changes over time at the place of sample collection as well as anthropogenic input (plutonium, ^{129}I) after 1945. At the time of writing (spring 2020), the project is still ongoing but with the use of the newest detectors in HADES (SAGe well detectors) it has become possible to measure the very low activities (0.2 mBq) of ^{226}Ra and ^{210}Pb in the small samples.

Figure 19. Photo of cold-water corals (and sea lilies in the background) at a depth of 550 m in the western Atlantic.



Source: MARUM – Centre for Marine Environmental Sciences, University of Bremen (CC-BY 4.0).

FURTHER READING: OLDEST ORGANISMS

- M. Hult et al., 2022, 'Low-level radioactivity measurements of natural archives', paper accepted for presentation at the ICRM Low-Level Radioactivity Measurements Techniques conference, Gran Sasso, Italy, 2-6 May, <https://icrm2020.lngs.infn.it/>.

Tracer studies, Case 4: Meteorites – messengers from space

Radionuclides are produced by cosmic rays in meteoroids (see text box below) as they travel through space. Measuring the activity of meteorites will provide scientists that study them with essential information.

The geological and biological history of Earth is largely shaped by the violent impacts of huge meteoroids and asteroids.

- It is generally considered that the major extinction of the dinosaurs began with an asteroid impact on the northern edge of the Yucatán peninsula (the Chicxulub crater) 66 million years ago.
- Recent research points to an asteroid impact in Greenland as responsible for the Younger Dryas period (12 800 to 11 500 years ago) – essentially a thousand-year prolongation of the most recent ice age.
- Many other asteroid impacts are suspected of being responsible for sudden changes in Earth's climate in the past and for inducing floods.

Radionuclides, serving as internal clocks, can tell us a lot about meteorites. By studying the physics of the meteorites, the remains of meteoroids that continuously hit Earth, it is possible to better understand the nature of the enormous prehistoric impacts. Important information can include the following, for example:

- time of formation – typically 4.5 billion years ago, but recently objects not originating from our solar system have been detected;
- time of breakup, i.e. when a big asteroid broke up into smaller pieces;
- terrestrial age – how long it has been on Earth;
- pre-atmospheric size;
- path through space, e.g. how near to the sun.

To obtain all this information, many radionuclides need to be measured, both short- and long-lived ones. This includes the use of both radiometric and non-radiometric (typically mass spectrometry) techniques.

JRC-Geel has contributed to harmonisation work by supporting the cosmochemistry department of the Max Planck Institute for Chemistry in Mainz. The study in question compared gamma-ray spectrometry and mass spectrometry for measuring ^{26}Al (half-life: 717 000 years). The result confirmed that both techniques are useful and comparable.

In an ongoing study, as part of the JRC open access scheme, JRC-Geel measures meteorites for the Polish National Centre for Nuclear Research Radioisotope Centre. A very unusual detection was made, namely that of ^{60}Fe (half-life: 2.6 million years)

in a stony meteorite that is so recently discovered that it still has to be named. In the same project, small pieces of the Ghubara meteorite will be measured to obtain depth profile of radionuclides.

Figure 20. A piece of the Ghubara iron meteorite (compared with a dice of 1 cm³). It fell several thousand years ago and was found in 1954 in Oman. It contains unusual isotope ratios of trapped inert gases. There are diverging views on its original mass.



Photo: Tomasz Kubalaczak.

Meteoroid	A small rocky or metallic body in outer space.
Asteroid	Minor planet, especially of the inner solar system.
Meteor	The visible passage of a glowing meteoroid, a 'shooting star'.
Meteorite	A solid piece of debris from an object, such as a comet, asteroid or meteoroid, that originates in outer space and survives its passage through the atmosphere to reach the surface of a planet or moon.

Definitions from Wikipedia.

FURTHER READING: METEORITES

- P.N. Johnston et al., 2002, 'Measurement of low levels of ^{26}Al from meteorite samples', *Applied Radiation and Isotopes*, Vol. 56, pp. 399–403.
- S. Merchel et al., 2004, 'First international ^{26}Al interlaboratory comparison – Part II', *Nuclear Instruments and Methods in Physics Research Section B*, Vol. 239, pp. 217–226.
- Z. Tymiński et al., 2020, 'Underground radioactivity measurements of meteorites', paper in manuscript.

Part III:
Fundamental physics
and decay data

Obtaining basic physical data necessary for modern society and to study our universe

In total, there are about 3 500 radionuclides, all of which decay in their own special manner. For science, different decay parameters are very valuable to study and include in reference tables – for examples, see box ‘Interesting decay parameters’.

INTERESTING DECAY PARAMETERS

- Half-lives for radionuclides: the time it takes before 50 % of the atoms of the radionuclide in question have decayed.
- Half-lives (lifetimes) for excited states: like the previous but for the decay of an excited state instead of disintegration of the atom.
- Emission probabilities of different types of radiation (alpha particles, gamma-rays, etc.).
- Branching ratio: the fraction of the decay of a radionuclide that follows a certain path (electron capture, beta-plus, beta-minus or alpha-particle decay, spontaneous fission, etc.).
- Energy of emitted radiation.

However, as odd as it may seem, many of these decay parameters are still poorly known and many even unknown. Many were measured a long time ago with inadequate instrumentation, compared with the standards of today. The accuracy needed for modern science, medicine and industry is very different from that of a century ago.

Decay data is essential for a number of technical and scientific tasks, such as dating past events, calculating accurate radiation levels and understanding nuclear structure. For the proper storage of nuclear waste, we need to accurately calculate decay heat and activity. In medical research, we need to know precisely the decay parameters of radiopharmaceuticals before injecting them into patients.

There are also some grand challenges of basic research that can be probed in underground laboratories, such as determining the mass and the nature of the neutrino, thus deepening our understanding of the universe. We may note that it is actually the grand challenges that have enabled physics to push the limits for low-level measurements to uncharted territory. HADES has played a small but important role by performing detector development and testing – both for the development of the world’s most radiopure detector, Borexino (Case 4), and for the germanium detector array (GERDA) and large enriched germanium experiment for neutrinoless double beta decay (LEGEND) double beta decay experiments (Case 3).

Fundamental physics, Case 1:

The lowest decay energy known to humans

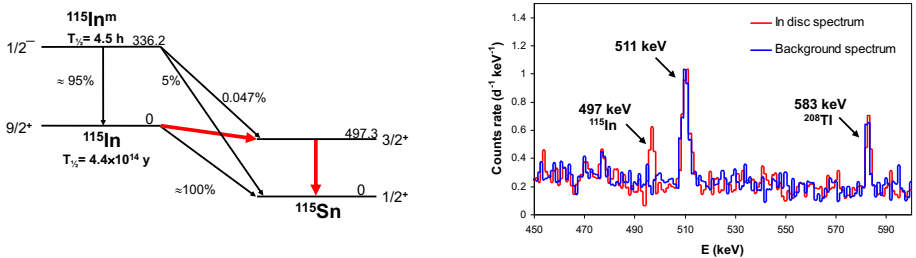
Monitoring a very special nuclear decay may give scientists a better understanding of the nature of the neutrino.

When an atom undergoes a radioactive decay, a certain, fixed amount of energy – the decay energy – is released to various particles (electrons, alpha particles, neutrinos, gamma-rays, etc.). By studying a few radionuclides (such as tritium and ^{187}Re) with extremely low beta-decay energy, scientists can obtain information about the mass and nature of the neutrino – one of the most enigmatic subatomic particles.

This is being explored by Karlsruhe Institute of Technology by the use of a gigantic detector (<http://www.Katrin.kit.edu>) that measures the decay energy of tritium (18.6 keV). In 2005, a research group from the Gran Sasso National Laboratory in Italy discovered a decay in ^{115}In , which has a much lower beta-decay energy (0.155 keV) than tritium. This decay was studied in HADES by measuring a 2.6 kg disc of pure indium for 215 days. The half-life of the rare decay – from ^{115}In to the first excited state in ^{115}Sn – is $4.3(5) \times 10^{20}$ years, building on the data from HADES.

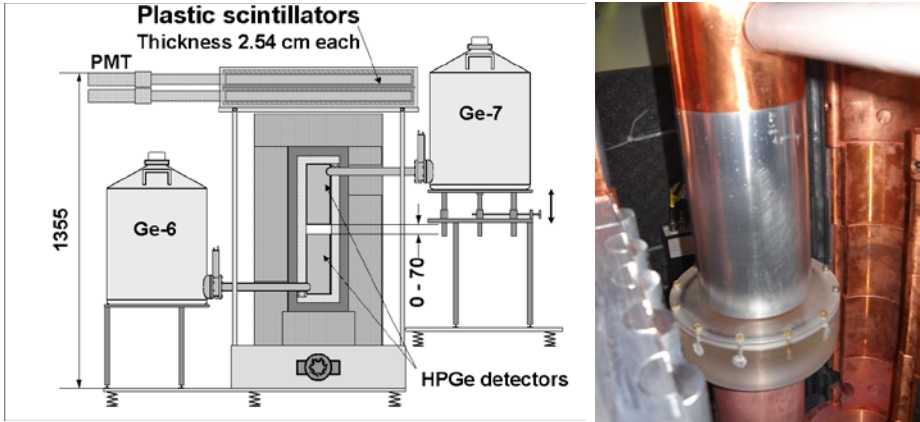
Some still preliminary data from HADES shows that there is no significant difference in the half-life of this decay when indium is in a different chemical environment, such as the salt InCl_3 . There are suspicions that decays with very low decay energy depend on the quantum mechanical state of the orbital electrons that are affected by chemical bonds.

Figure 21. Left: the decay scheme of $^{115\text{m}}\text{In}$ and ^{115}In . Right: part of the gamma-ray spectrum including the elusive peak at 497 keV.



NB: By measuring underground one effectively removes the influence of the decay of the metastable state, which has a short-half-life, as it can be induced by cosmic rays above ground.

Figure 22. Left: schematic drawing of the sandwich detector system. Right: photo of the two detectors with a sample in between when the shield is open.



FURTHER READING: LOWEST DECAY ENERGY

- E. Andreotti et al., 2011, 'The half-life of the beta decay $^{115}\text{In}(9/2^+) \rightarrow ^{115}\text{Sn}(3/2^+)$ ', *Physical Review C*, Vol. 84, No 4, 044605, doi: 10.1103/PhysRevC.84.044605.
- J.S.E. Wieslander et al., 2009, 'Smallest known Q value of any nuclear decay: The rare β^- decay of $^{115}\text{In}(9/2^+) \rightarrow ^{115}\text{Sn}(3/2^+)$ ', *Physical Review Letters*, Vol. 103, 122501.

Fundamental physics, Case 2: The most long-lived isomeric state in the universe?

^{180m}Ta is a peculiar radionuclide with some unique properties.

Tantalum is an intriguing element. It was discovered in 1802 by the Swedish chemist Anders Gustaf Ekeberg in the famous rock sample ⁽²²⁾ from Ytterby mine, Sweden. Ekeberg named the new element after the Greek demigod Tantalus, who was sentenced to an eternity of suffering from hunger and thirst. He wrote that this was 'partly in allusion to its incapacity, when immersed in acid, to absorb any and be saturated'. The isotope ^{180m}Ta counts as the rarest naturally existing isotope on Earth. Furthermore, ^{180m}Ta is the heaviest of all stable odd-odd nuclei (referring to the number of neutrons and protons), of which there are only nine.

The second excited state in ^{180m}Ta is a very long-lived metastable state (the 'm' stands for 'metastable'). It is in fact the most long-lived metastable state known to man. This metastable state has had implications for studies in building a gamma-ray laser as well as for models of its production in stars (stellar nucleosynthesis). The decay of this metastable state has never been detected. However, of the 10 attempts reported in literature, the 3 best attempts have been made in HADES.

- The first attempt was not planned but merely a spin-off (suggested by Professor Ron Fleming, University of Michigan) from a neutron experiment in which a piece of tantalum was used.
- The second attempt was a metrologically well-designed experiment.
- The third attempt was a development of the second attempt, in which some more measurement time was added and a combination of all measurements introduced using advanced statistical methods. This resulted in a lower limit of the half-life of 4.5×10^{16} years. This value can be compared with the first measurement in 1958, which gave a limit of only 1.5×10^9 years. It represents an improvement of more than seven orders of magnitude.

In addition, based on the three measurements in HADES, it has unexpectedly been possible to derive some interesting numbers related to detection of dark matter (Lehnert et al., 2020) that help to better constrain models of interactions of dark matter.

(22) The following elements were also discovered after analysis of the same rock that was found by Carl Axel Arrhenius in 1787: ytterbium (Yb), yttrium (Y), terbium (Tb), erbium (Eb), holmium (Ho), scandium (Sc), thulium (Tm) and gadolinium (Gd).

Figure 23. The decay scheme of ^{180m}Ta . (EC stands for Electron Capture.)

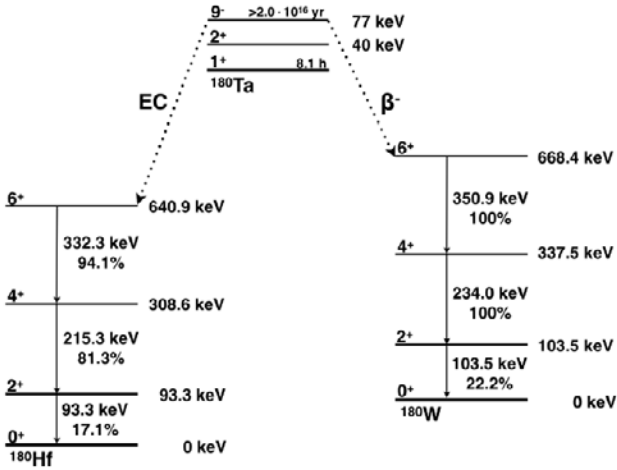


Figure 24. The six hyperpure tantalum discs used for the ^{180m}Ta measurements. The discs were cleaned by Italian National Institute for Nuclear Physics (INFN) Padua to remove surface impurities, then they were stored a year underground for ^{182}Ta to decay before starting measurements.

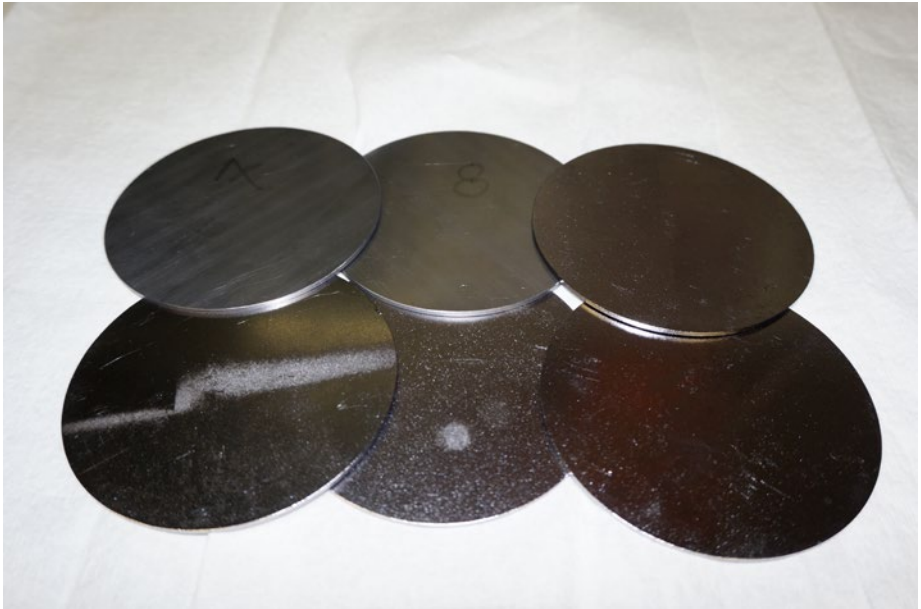


Photo: M. Hult.

FURTHER READING: MOST LONG-LIVED ISOMERIC STATE

- E. Conover, 2016, 'Rarest nucleus reluctant to decay', *ScienceNews*, 3 October, <https://www.sciencenews.org/article/rarest-nucleus-reluctant-decay>.
- M. Hult et al., 2006, 'Underground search for the decay of $^{180\text{m}}\text{Ta}$ ', *Physical Review C*, Vol. 74, 054311.
- M. Hult et al., 2009, 'Search for the radioactivity of $^{180\text{m}}\text{Ta}$ using an underground HPGe sandwich spectrometer', *Applied Radiation and Isotopes*, Vol. 67, pp. 918–921.
- B. Lehnert et al., 2017, 'Search for the decay of nature's rarest isotope $^{180\text{m}}\text{Ta}$ ', *Physical Review C*, Vol. 95, 044306.
- B. Lehnert et al., 2020, 'Search for dark matter induced de-excitation of $^{180\text{m}}\text{Ta}$ '. *Physical Review Letters*, Vol. 124, Iss. 1802-1, p. 1-7. <https://doi.org/10.1103/PhysRevLett.124.181802>

Fundamental physics, Case 3: Determining the nature of the neutrino

A grand challenge is to study physics beyond the standard model, which can give results that force us to reconsider physics as we know it today.

The particle named the neutrino interacts very little with matter. This also means that such particles (²³) can travel very far. Thus, by detecting them, we can learn about the origin of the universe (²⁴), conditions in stars, galaxies and even the centre of Earth. Until some 20 years ago, it was not known whether they had mass or not. Now, it is accepted that they have a mass but it is so low (< 0.2 eV) that it has not yet been determined. One ingenious experiment has been designed to determine the effective mass (a superposition of the mass of the three different types of neutrinos that exist) and simultaneously determine whether or not the neutrino is its own antiparticle. This experiment is based on the measurement of the neutrinoless double beta decay, which may occur in a few nuclides.

One of the most interesting nuclides in this context is ⁷⁶Ge. This is because it is possible to build detectors from germanium and thereby make the Ge crystals both sample and detector at the same time. A gigantic detector has been built in the Gran Sasso underground laboratory in Italy, called GERDA (GERmanium Detector Array). It is composed of an outer water tank of 390 m³ and an inner steel/copper tank holding 64 m³ of liquid argon. In the centre of the inner tank are the 39 germanium crystals that form the core of the experiment. All of them were exposed to careful and precise testing in the underground laboratory HADES before they were shipped to Gran Sasso for inclusion in the grand experiment.

There were numerous outcome of the GERDA experiment, for example the exact value of the half-life of the two-neutrino double beta decay of ⁷⁶Ge (1.0×10^{21} years). GERDA also significantly helped to improve the technology for producing Ge detectors.

Something that was not detected was the neutrinoless double beta decay. However, a limit on the half-life of 1.1×10^{26} years was published in *Nature* and all former claims of detection could be refuted. In spite of not detecting the decay, the experiment was a success, as it reached its design goals. A bigger experiment is needed to learn more about this possible decay and it is now being conceived by a bigger consortium, LEGEND. For this worldwide experiment too, the germanium crystals will be tested in HADES.

(23) We use the plural because there may be up to six types of neutrinos: one linked with each of the three leptons – electron, muon and tau – and (if the neutrino is a Majorana particle) also the corresponding anti-neutrino.

(24) See for example (https://en.wikipedia.org/wiki/Cosmic_neutrino_background).

THE NEUTRINO

Neutrinos are produced in nuclear reactions and in radioactive beta decay. Every second, Earth is hit by almost 10^{11} neutrinos per cm^2 . This means that the human body is penetrated by about 10^{14} neutrinos each second. This particle interacts extremely little with matter and is therefore very difficult to detect. It was not detected until 1956 by Reines and Cowan. Since then, Nobel prizes were awarded in 1988, 1995, 2002 and 2015 for work on neutrinos. Still, there is much to be learned.

Figure 25. The transport container for transporting a few germanium crystals (total volume of only about 30 litres) by road from HADES to Gran Sasso. The container was filled with water and iron to reduce the flux of muons and neutrons (with a factor 10) during the transport.



Photo: M. Hult.

FURTHER READING: NEUTRINO RESEARCH

- E. Andreotti et al., 2013, 'Characterization of BEGe detectors in the HADES underground laboratory', *Nuclear Instruments and Methods in Physics Research Section A*, Vol. 718, pp. 475–477.
- GERDA Collaboration, 2017, 'Background-free search for neutrinoless double beta decay of ^{76}Ge with Gerda', *Nature*, Vol. 544(7648):47-52. doi: 10.1038/nature21717
- GERDA Collaboration, 2019, 'Probing Majorana neutrinos with double beta decay', *Science*, Vol. 365, pp. 1445–1448.

Fundamental physics, Case 4: Borexino – the least radioactive volume on Earth

A ground-breaking detector design forced scientists to create the most radiopure volume on Earth and possibly the universe’s most radiation-free space.

As mentioned in the previous case-story about GERDA, the neutrino is an intriguing particle that is very difficult to detect because of its extremely low interaction probability. This means that any detector that is designed to detect neutrinos needs to be very big.

If the detector aims to detect low-energy neutrinos below 1 MeV (for example the pp neutrinos²⁵ below 420 keV), it is also necessary that the detector be extremely free of natural radioactivity. Unfortunately, uranium, thorium and other natural radionuclides are literally present everywhere (albeit in very low concentrations). Therefore, it is a formidable task to try to design a detector with an intrinsic activity concentration of uranium and thorium of less than 10^{-17} g/g (about 100 nBq/m³ for ²³⁸U).

This was indeed the goal of the Borexino detector installed in the Gran Sasso underground laboratory. To achieve this, all materials that went into constructing this 278-tonne detector needed to be measured and approved for low levels of radioactivity.

The Borexino collaboration needed support from JRC-Geel to measure materials such as stainless steel (for the tank), rubber O-rings, the liquid scintillation cocktail and nylon (a shroud to define the fiducial volume). In 1997 JRC joined the Borexino collaboration and contributed for several years with measurements that ultimately led to the successful inauguration of the detector

The Borexino detector can now be described as the most radiopure volume on Earth. It has successfully detected geoneutrinos (neutrinos from radioactive decay taking place at the centre of Earth ⁽²⁶⁾) as well as neutrinos from the pp and ⁷Be processes in the sun. The Borexino detector is also a vital part of the newly formed SuperNova Early Warning System, aiming for rapid communication within the scientific community whenever a supernova is detected.

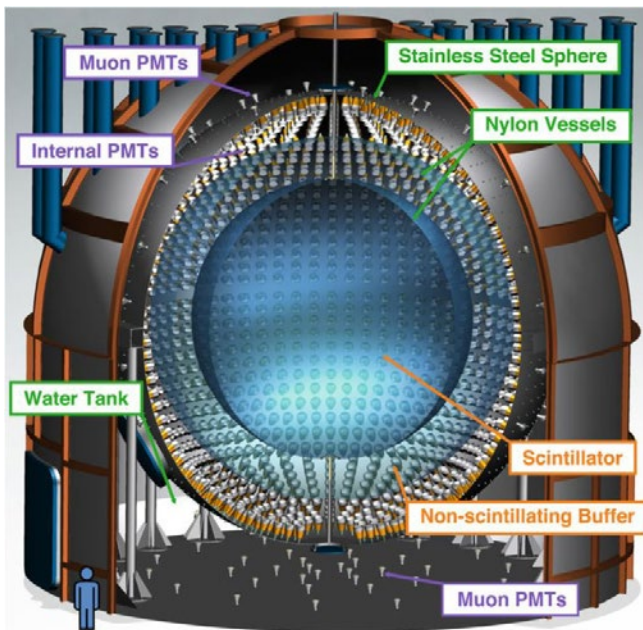
(25) The proton–proton chain reaction is one of two known sets of nuclear fusion reactions by which stars convert hydrogen to helium. (https://en.wikipedia.org/wiki/Proton–proton_chain_reaction)

(26) Radioactive decay is to a large extent responsible for the production of heat inside Earth (https://en.wikipedia.org/wiki/Earth%27s_internal_heat_budget).

FURTHER READING: BOREXINO

- Borexino Collaboration, 2002, 'Measurements of extremely low radioactivity levels in Borexino', *Astroparticle Physics*, Vol. 18, pp. 1–25.
- Borexino Collaboration, 2009, 'The Borexino detector at the Laboratori Nazionali del Gran Sasso', *Nuclear Instruments and Methods in Physics Research Section A*, Vol. 600, pp. 568–593.
- Borexino Collaboration, 2014, 'Neutrinos from the primary proton–proton fusion process in the Sun', *Nature*, Vol. 512, No 7515, pp. 383–386.

Figure 26. Schematic view of the onion-like Borexino detector. The outer diameter of the outer water tank is 18 m. The photomultiplier tubes (PMTs) detect light from nuclear interactions in the scintillator.



Drawing: Borexino collaboration.

Part IV: Neutron and plasma physics

Applications related to nuclear energy and nuclear safety

A key task for JRC-Geel since the start has been to measure neutron cross-sections using its two accelerator laboratories based on a 100 MeV linear accelerator and a 3.5 MV tandem accelerator. Neutron cross-sections are necessary to know precisely, in order to calculate the operational parameters of nuclear reactors, both those based on fission and those based on fusion. Better knowledge of cross-sections will also lead to greater safety margins and opportunities to realise more efficient modes of operation.

There are numerous ways of measuring such cross-sections. One way is to use the activation technique. In this, a well-known material (reference material) is irradiated with a known flux of neutrons. After that, the radioactivity induced in the material is measured using gamma-ray spectrometry. In cases with low yield (due to, for example, low isotopic abundance, small cross-section or low gamma-ray yield) it is necessary to measure the irradiated material in an underground laboratory.

Neutron and plasma physics, Case 1: Measuring charged particles inside a tokamak reactor

To study the flow of charged particles inside an experimental fusion reactor, pieces of material are placed close to the plasma. Activation induced in such materials has been measured in HADES.

Providing energy based on nuclear fusion (the same reactions as in all stars) has been a dream for many decades. However, it has proven to be very difficult, as it involves creating a plasma – something that requires the fuel to be heated to about 175 million °C (15 keV), confined in a small space for a sufficient time (many minutes ⁽²⁷⁾). One important kind of reactor design, where a magnetic field is used to confine the hot plasma in the shape of a torus, is called tokamak.

To this date, it has not been possible to build a reactor that can produce more power than it needs to operate. More research is necessary to be able to understand the fusion plasma dynamics in tokamaks (and other types of reactors). Many plasma physicists therefore eagerly await the ITER reactor, which is presently being built in France and will be the biggest tokamak ever. It is designed to generate a power of 500 MW from an input heating power of 50 MW.

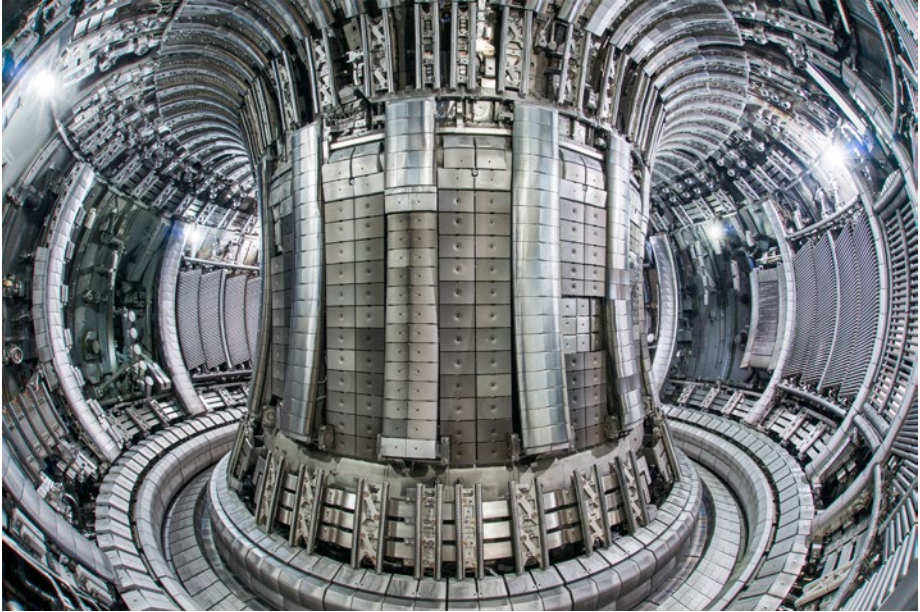
Because of the extreme environment inside the tokamak, no normal detectors can be positioned there to detect charged particles such as protons, deuterons, tritons and alpha particles. It is essential to monitor such particles, as they are necessary to keep the plasma ‘alive’. It is the speed of leakage of these particles that determine the time for which the plasma can be confined. Too high a leakage of these particles could also damage the walls of the tokamak.

Dr Georges Bonheure (at the time working for Plasma Physics Laboratory Brussels and for the Joint European Torus (JET)) developed a technique by which small (1 cm × 1 cm) pieces of materials can be placed close to the plasma. After one or several plasma pulses, the materials are removed via a port in the reactor. The small amount of activation induced by the charged particles is then measured in HADES. In a series of articles, the research group has shown that it is possible to do the following:

- measure the leakage of charged particles inside tokamaks;
- separately study 3 MeV and 14 MeV protons – this is important, as they are produced in different nuclear reactions;
- study individual plasma pulses by sequentially covering sets of materials;
- study the angular distribution of charged particles by placing the materials to face in different directions.

(27) ITER (<https://www.iter.org>) aims to achieve plasma pulses lasting up to 10 minutes.

Figure 27. Interior view of the JET tokamak in Culham, United Kingdom.



Source: EUROfusion.

Figure 28. Cross-sectional drawing (left) and side-view photo (right) of the probe used for measuring protons at JET.

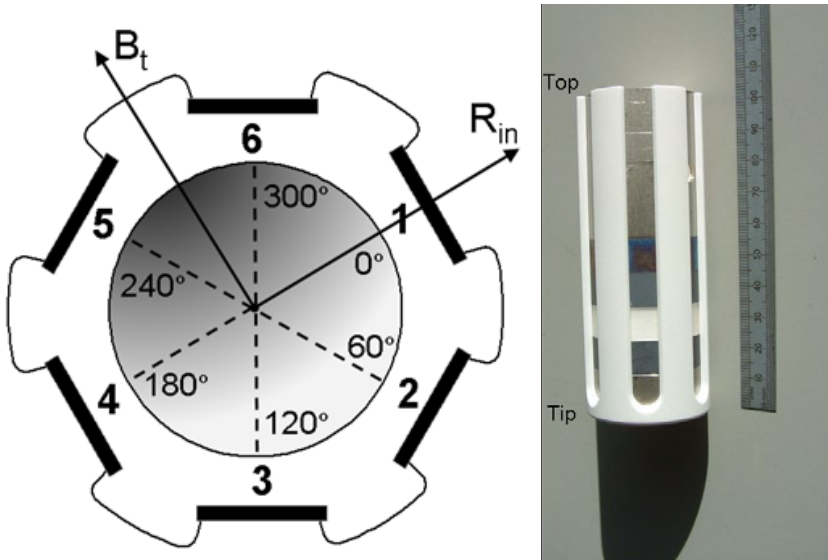


Photo: G. Bonheure. Licensed under CC BY 2.0. From González de Orduña et al. (2011).

FURTHER READING: MEASURING PARTICLES INSIDE A TOKAMAK REACTOR

- G. Bonheure et al., 2011, 'Fusion alpha loss diagnostic for ITER using activation technique', *Fusion Engineering and Design*, Vol. 86, pp. 1298–1301.
- G. Bonheure et al., 2012, 'Experimental investigation of the confinement of $d(^3\text{He},p)\alpha$ and $d(d,p)t$ fusion reaction products in JET', *Nuclear Fusion*, Vol. 52, 083004.
- J. Gasparro et al., 2006, 'A low-level activation technique for monitoring thermonuclear fusion plasma conditions', *Applied Radiation and Isotopes*, Vol. 64, pp. 1130–1135.
- R. González de Orduña et al., 2010, 'Angular distribution of proton leakage from a fusion plasma using ultra low-level gamma-ray spectrometry', *Applied Radiation and Isotopes*, Vol. 68, pp. 1226–1230.
- R. González de Orduña et al., 2011, 'Monitoring the leakage of 3.0 and 14.7 MeV protons from a fusion plasma', *Nuclear Instruments and Methods in Physics Research Section A*, Vol. 632, pp. 89–100.
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Neutron and plasma physics, Case 2:

A novel technique for neutron dosimetry, spectrometry and retrospective assessment

Neutrons are difficult to detect, as they lack electric charge. However, making use of the fact that they activate materials can help scientists look back at past events. A novel technique called Dosimetry using Neutron Activation (DONA) was developed at JRC-Geel.

The work following the Tokaimura accident and Hiroshima follow-up (see Part VI) pointed to the necessity to measure neutron flux retroactively. It triggered the development of a small well-defined probe with a set of metal discs (see Fig. 29) that can be placed at almost any location. After collecting these metal discs, the activity of many different radionuclides produced by neutrons is measured. The neutron energy spectrum and the neutron fluence rate can then be determined, and from this, dose rate can be calculated. This is done by performing a mathematical trick called unfolding (or deconvolution).

The great advantages of this technique are the following:

- The activation probe is completely passive and requires no electricity to operate.
- The probe is very durable and can be placed in harsh environments; it may even be designed to be dropped from a helicopter during an ongoing nuclear accident.
- As the sensitivity is high, relatively low neutron fluences can be measured. This enables dosimetric evaluation in areas where people are allowed to work.
- The probe requires very little space. The alternative technique, using Bonner spheres, requires an available volume that is 100 times bigger. Therefore, DONA also enables measurement of neutron spectra with high lateral resolution. This is important in certain applications, such as investigation of neutron spectra as a function of depth in certain materials.
- Representing a new, independent methodology, this technique can be used to validate or check other older techniques such as the use of Bonner spheres.

The DONA technique requires good knowledge of the neutron cross-section curves for the production of the radionuclides that are measured. The cross-section curves are input parameters together with the activities. Note that the detector can only provide retrospective data, as one has to measure the activity of radionuclides in each disc before a neutron spectrum can be produced.

Figure 29. A neutron detector based on 11 different metal discs.



Photo: M. Hult.

FURTHER READING: DONA-TECHNIQUE

- M.J. Martínez et al., 2000, "Measurements of activation induced by environmental neutron using ultra low-level gamma-ray spectrometry", *Applied Radiation and Isotopes*, 52,, p. 711-716.
- G. Lövestam et al., 2009, "Neutron fluence spectrometry using disk activation", *Radiation Measurements* 44, p. 72-79.
- E. Wieslander et al., 2010, "Validation of a Method for Neutron Dosimetry and Spectrometry Using Neutron Activation of Metal Discs", *Radiation Protection Dosimetry* 138, p. 205-212.

Neutron and plasma physics, Case 3:

A novel technique for measuring neutron cross-section curves

A novel technique, called Neutron Activation X-Section determined using UNfolding (NAXSUN), to measure neutron cross-section curves at accelerator labs has been developed. It is basically the reverse operation from the previous case (Case 2), the DONA technique.

It is always useful to have access to different techniques for measuring the same parameters. A certain technique may have a measurement bias, which may not be known until an alternative technique is used. Furthermore, it is technically difficult to measure some neutron cross-section curves with the traditional activation technique.

An alternative technique to measure neutron cross-section curves was conceived at JRC-Geel. It was further developed by the University of Novi Sad, Serbia. The principle of operation is as follows.

Step 1. Irradiation of several identical pure metal discs in different wide-energy neutron beams that overlap with each other. Only one disc is irradiated in each beam.

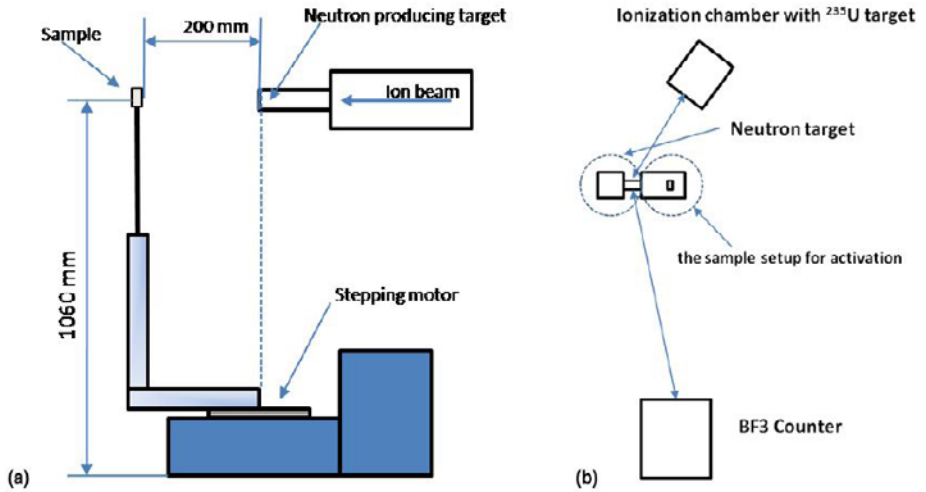
Step 2. Gamma-ray spectrometry of each disc to determine the activity of the radionuclides produced.

Step 3. Calculation of the neutron cross-section function using an unfolding procedure, in which the known spectra of the neutron beams and the activity of the radionuclides are input parameters.

To obtain an absolute value of the cross-section one needs to irradiate one more disc at one well-defined (not wide) neutron energy.

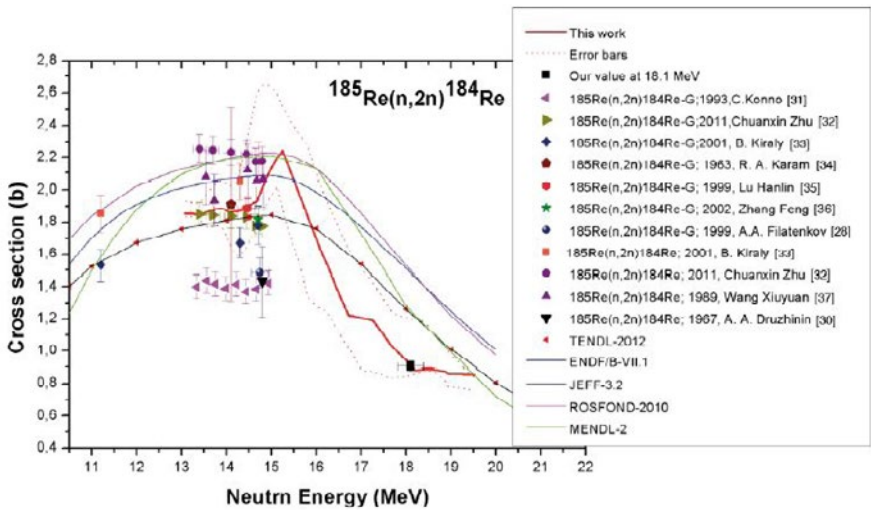
In essence, this technique is the reverse operation from Case 2 (in which the neutron cross-section is known but the neutron spectrum unknown).

Figure 30. Schematic drawing of the experimental set-up to activate one metal disc with a wide neutron beam.



Source: N. Jovančević et al. (2016).

Figure 31. Neutron cross-section curve measured (red solid line) using the NAXSUN technique. The other lines are from theoretical calculations. Only a few experimental data points exist around 14 MeV.



Source: N. Jovančević et al., 2016, European Physical Journal A, Vol. 52, 148.

FURTHER READING: NAXSUN TECHNIQUE

- N. Jovančević, et al., 2015, 'Modeling of neutron spectra based on activation analysis', *Physics Procedia*, Vol. 64, pp. 204–210.
- N. Jovančević, et al., 2016, 'The neutron cross-section functions for the reactions $^{187}\text{Re}(n, \alpha)^{184}\text{Ta}$, $^{187}\text{Re}(n, 2n)^{186}\text{Re}$ and $^{185}\text{Re}(n, 2n)^{184}\text{Re}$ in the energy range 13.08–19.5 MeV', *European Physical Journal A*, Vol. 52, 148–160.
- G. Lövestam, et al. 2007, 'Measurement of neutron excitation functions using wide energy neutron beams', *Nuclear Instruments and Methods in Physics Research Series A*, Vol. 580, pp. 1400–1409.

Neutron and plasma physics, Case 4: Neutron cross-section measurements

If the yield from a traditional neutron cross-section measurement is very low, working in the low-background environment of HADES may be the only way to get useful data.

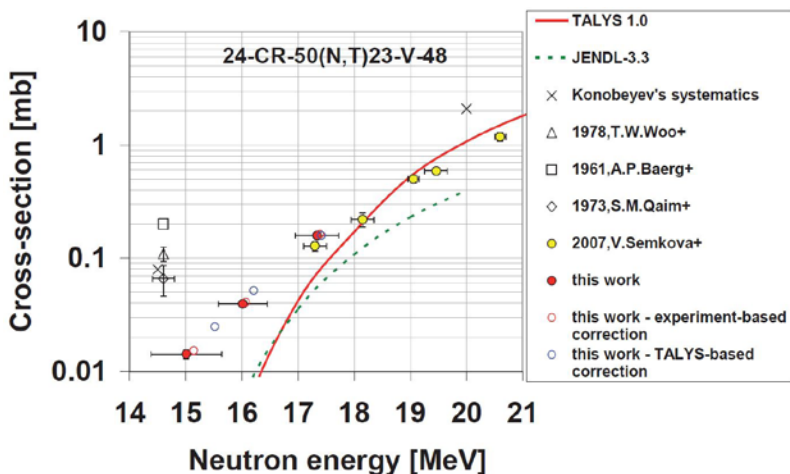
One common way of measuring neutron cross-sections is to irradiate a material with a well-defined mono-energetic beam of neutrons from an accelerator. For each new data point, the energy of the neutron beam has to be changed. Hence, many irradiations are necessary to create a curve.

After irradiation, the gamma-rays from the produced radionuclides are measured, typically by the use of a HPGe-detector. In some cases the number of gamma-rays emitted by the sample is very low, because, for example:

- the neutron flux from the accelerator was low;
- the amount of material was low;
- the isotopic abundance of the isotope of interest was low;
- the neutron cross-section was very low.

In such cases, it makes sense to measure the activity in HADES, given that the radionuclide has a sufficiently long half-life that there is time to transport the sample to HADES.

Figure 32. Tritium yield in a nuclear reaction. The red points correspond to low-yield data measured in HADES.



Source: A. Krasa et al., 2011, Nuclear Measurements, Evaluations and Applications - NEMEA-6 Workshop Proceedings pp. 225-237.

FURTHER READING: NEUTRON CROSS-SECTION MEASUREMENTS

- C. Ingelbrecht et al., 2002, 'An ^{26}Al target for (n, p) and (n, α) cross section measurements', *Nuclear Instruments and Methods in Physics Research Section A*, Vol. 480, pp. 114–118.
- A. Krasa et al., 2011, 'Tritium production in neutron induced reactions', Proceedings from NEMEA-6 Scientific Workshop on Nuclear Measurements, Evaluations and Applications – 6, OECD/NEA, Krakow, Poland, pp. 225-237.
- P. Reimer et al., 2002, 'Measurement of the ^{nat}Mo (n, x) ^{94}Nb cross section using ultra low-level γ -ray spectrometry at HADES', *Nuclear Physics A*, Vol. 705, pp. 265–278.

Part V:
Environment and industry

Natural radioactivity is present everywhere. Even in seawater there are traces of uranium. In any industry where huge amounts of materials are processed (mining, metallurgy, water purification, phosphate industry, etc.) there will be by-products or residue with elevated levels of natural radioactivity. Such materials are often called naturally occurring radioactive materials (NORMs). It is also possible to find natural materials that have elevated levels of activity without prior industrial processing. Accordingly, these materials leave traces enabling innumerable applications of environmental and industrial research that can benefit from low-level radioactivity measurements.

FURTHER READING: ENVIRONMENT AND INDUSTRY

- W. Bambynek, 1988, 'Radionuclide data', in: M. Garcia-Leon and G. Madurga (eds.), *Low-level measurements and their applications to environmental radioactivity, proceedings of the first summer school, La Rabida, Huelva, Spain, 1987*, World Scientific, Singapore, p. 37-41.
- M. Hult et al., 2003, 'Underground measurements of environmental radioactivity in Europe', in: *Proceedings of the International Symposium of the Kanazawa University 21st Century COE Program*, Kanazawa University, Vol. 1, pp. 18-23.

Figure 33. A typical pile of phosphogypsum, which is a common NORM.



Source: Wikipedia commons.

Environment and industry, Case 1: Uptake of radionuclides in rice and other plants

Understanding pathways for uptake of different radionuclides in crops will enable better instructions for population and farmers on how to act in the event of an accidental release of radioactivity.

In the wake of the Fukushima accident, much effort was made to remediate contaminated soil. This is very expensive and – as it turns out – not always necessary. By adapting farming techniques by, for example, performing extra ploughing and selecting plants that are suitable crops, it is possible to reduce the amount of radiocaesium in the end product.

To better understand the biochemistry in rice plants, the radioecology department of the Belgian nuclear research centre (SCK CEN) performed a study by cultivating rice in a contaminated solution. Instead of measuring the whole plant, the scientists measured the small uptake to different parts (root, stem, leaves and shoots) and tried to understand how much is taken up by the stem, compared with the root. This is the first study that can point to a significant uptake by the stem as well, called foliar uptake. It also shows that, by increasing the availability of potassium in the soil, the uptake of radiocaesium can be reduced (Uematsu et al., 2017).

In another study conducted under the JRC exploratory research programme, the uptake of natural radioactivity (from ^{238}U and ^{232}Th chains as well as ^{40}K) was studied in wheat (and to a lesser extent also in potato, horse radish and carrot) (Lindahl et al., 2007, 2011). The global aim was to investigate analytical methods to distinguish products from what is called organic farming (bioproducts) from conventional farming. Compared with manure, fertilisers based on phosphate ore contain elevated levels of natural radioactivity. Although greenhouse studies under very controlled conditions may generate results that correlate with fertiliser type, it is very difficult to use that as a sole flag for crops cultivated in fields. This is because the uptake depends on a great many factors such as the type of soil, weather, precipitation and unusual events.

However, the study performed was able, as a spin-off, to produce fundamental data on transfer factors (from soil to plant parts) of radionuclides, which are important for future radioecology studies.

FURTHER READING: UPTAKE OF RADIONUCLIDES IN PLANTS

- P. Lindahl et al., 2011, 'Natural radioactivity in winter wheat from organic and conventional agricultural systems', *Journal of Environmental Radioactivity*, Vol. 102, pp. 163–169.
- P. Lindahl et al., 2007, 'Improvements in underground gamma-ray spectrometry and the application of measuring radioactivity in agricultural samples', in: P. Warwick (ed.), *Environmental Radiochemical Analysis III*, Special Publication No 312, Royal Society of Chemistry Publishing, Cambridge, pp. 86–94.
- S. Uematsu et al., 2017, 'Foliar uptake of radiocaesium from irrigation water by paddy rice (*Oryza sativa*): An overlooked pathway in contaminated environments', *New Phytologist*, No 214, pp. 820-829, DOI: 10.1111/nph.14416.

Figure 34. A rice plantation.



Source: Wikipedia commons.

Environment and industry, Case 2: Novel types of concrete with small CO₂ footprints

By providing data about the natural radioactivity in alternative bulk products for construction materials, the JRC assisted research and development work aiming to reduce the amount of both hazardous by-products and CO₂ emission.

Bauxite residue, also known as red mud, is a major by-product during the refinement of aluminium ore. For every 1 t of alumina produced, 1–1.5 t of bauxite residue is generated. Worldwide, about 120 Mt is produced each year and only 4 % is utilised. The University of Hasselt together with Kiev National University of Construction and Architecture conducted a study in which they used red mud as aggregate in concrete products. The study involved chemical and mechanical investigations. At the radionuclide metrology laboratory at JRC-Geel (complemented by a few HADES measurements), the radiological properties of the starting material and final products were investigated.

As many industrial by-products have increased levels of natural radioactivity (from thorium and uranium decay chains), it is important to consider these aspects when utilising them in, for example, construction materials. The study shows that up to 90 % (by mass) of Ukrainian red mud can be incorporated in materials for road construction without jeopardising the health of workers and the public. When considering building materials, it is possible to use up to 75 %. In addition to reducing toxic waste such as red mud, it also cuts CO₂ emissions.

In concrete production, equal amounts (in mass) of CO₂ and concrete are produced. In another study, Hasselt University studied inorganic polymers that can replace concrete in construction materials and thereby vastly reduce CO₂ emissions. The work was presented in the PhD thesis of Tom Croymans.

FURTHER READING: NOVEL TYPES OF CONCRETE

- T. Croymans et al., 2017, 'Radiological characterization and evaluation of high volume bauxite residue alkali activated concretes', *Journal of Environmental Radioactivity*, Vol. 168, pp. 21–29.
- P. Krivenko et al., 2017, 'Development of alkali activated cements and concrete mixture design with high volumes of red mud', *Construction and Building Materials*, Vol. 151, pp. 819–826.
- T. Croymans et al., 2018, 'Valorization of Fe-rich industrial by-products in construction materials: A radiological assessment', PhD-thesis, Hasselt University, Belgium.

Environment and industry, Case 3: Support to a small or medium-sized enterprise in the semiconductor industry

The JRC assisted a high-tech European company in solving an analytical problem, where the challenge was to monitor the impurity level of semiconductor materials.

A world-leading European company in the semiconductor industry got different results from the two analytical techniques electron Raman scattering (ERS) and glow discharge mass spectrometry (GDMS) when checking their GaAs wafers for Zn impurities. The impurity level of semiconductors is of crucial importance, as it determines the electrical performance of the final products. At JRC-Geel, an advanced analytical method was conceived, which involved three steps:

1. neutron activation at the BR1 reactor at SCK CEN (300 m from HADES),
2. radiochemical separation,
3. gamma-ray spectrometry in HADES.

By using this technique, detection limits were better than GDMS by a factor of more than 1 000. The measurements at JRC-Geel concluded that the key problem was that the Zn levels were so low that they were close to or below the detection limits for ERS and GDMS. When measuring near the detection limits of a system, the uncertainty is very high. It was also concluded that GDMS performed better than ERS.

Table 2. Detection limits for Zn in GaAs using different techniques

Method	Detection limit (ng/g)
Glow discharge mass spectrometry	0.9
Radiofrequency glow discharge mass spectrometry	1.8
Inductively coupled plasma mass spectrometry	2.5
Spark source mass spectrometry	4
Electron Raman scattering	10
instrumental neutron activation analysis	9
Radiochemical neutron activation analysis + ultra low-level gamma-ray spectrometry in HADES	0.008

FURTHER READING: SUPPORT TO THE SEMICONDUCTOR INDUSTRY

- M. Köhler et al., 2000, 'Determination of Zn in high-purity GaAs neutron activation analysis', *Applied Radiation and Isotopes*, Vol. 53, pp. 197–201.

Figure 35. General HADES photos not directly linked to this case-story. Left: interior view of the Pacman dual detector system. Right: completing the shield of the well detector Ge12.



Photo: G. Lutter.

Environment and industry, Case 4: Radioactivity in human bones

Analysis of human bones can provide important information about a person. For example, the calcium to strontium ratio can give information about diet or at what age children were weaned. The level of ^{210}Pb can tell how much radon gas a person was exposed to.

Certain elements are bone-seekers and accumulate in the bones. Thorium is a typical example. The primordial radionuclide ^{232}Th is followed by a long decay chain. Thereby, one atom of ^{232}Th will eventually lead to 10 radioactive decays. ^{222}Rn is another atom that occurs naturally and is followed by a series of decays (in this case eight decays). Radon is a radioactive gas that typically enters the body via the lungs. A long-lived (22 years) daughter of ^{222}Rn is ^{210}Pb . Just like thorium, lead also accumulates in bones. By measuring ^{210}Pb in bones one can estimate the amount of radon gas that the person was exposed to. An example is the retrospective assessment of former uranium mine workers (SAG/SDAG Wismut company, see Part VII). Such work helps researchers to better assess the dangers of radon gas, impacting legislation on radiation protection.

In a study in collaboration with Hasselt University (Belgium), JRC researchers were able to show that ^{210}Pb is homogeneously distributed in the bone mineral (hydroxyapatite). As a consequence this means that ^{210}Pb is not homogeneously distributed in the bone, as the bone structure varies with depth. It is more dense on the outside and hollow within. In phantoms²⁸ used for calibrating whole-body counters at the time, ^{241}Am was used and it was generally homogeneously distributed in the phantoms. Since both ^{210}Pb and ^{241}Am emit gamma-rays with very low energy (46.5 keV and 59.5 keV, respectively), calibration using phantoms is very sensitive to the distribution of these radionuclides. The study served to make better calibrations of whole-body counters to determine the content of ^{210}Pb in bones.

FURTHER READING: RADIOACTIVITY IN HUMAN BONES

- P.N. Johnston et al., 2005, 'The distribution of ^{210}Pb in human bone and its impact on methods for the retrospective estimation of ^{222}Rn exposure from in vivo measurements', *Journal of Environmental Radioactivity*, Vol. 80, pp. 245–257.
- M.J. Martínez Canet et al., 2001, 'Ultra low-level γ ray spectrometry of thorium in human bone samples', *Radiation Protection Dosimetry*, Vol. 97, pp. 169–172.

(28) The word phantom is used in medicine to describe a specially designed object to mimic a human tissue or organ or complete body.

Environment and industry, Case 5: Geological applications

Underground gamma-ray spectrometry is useful when studying radioactive decay in geological structures, furnishing a better understanding of the planet we inhabit.

Geology is a field where radioactivity has contributed significantly to create knowledge and understanding of the history of Earth. For example, the decay of ^{238}U and the decay chain's end product, ^{206}Pb , tell us the age of Earth. The decay of numerous other radionuclides tells us the age of certain geological formations.

Many long-lived (greater than many millions of years) radionuclides are routinely measured using mass spectrometric techniques (counting atoms). However, the use of radiometric techniques (counting decays) is advantageous for certain problems. An important ingredient in a geological study is to study many different isotope ratios, which requires a battery of different analytical methods, underground gamma-ray spectrometry being one of them. This was evidenced in, for example, a study by Hoogewerff et al. (1997), in which HADES measurements shed light on geological dynamic processes in an arc-continent collision zone. Although HADES has not yet contributed significantly to such studies, it is a field that can be further exploited in the future.

FURTHER READING: GEOLOGICAL APPLICATIONS

- J. Hoogewerff et al., 1997, 'U-series, Sr-Nd-Pb isotope and trace-element systematics across an active island arc-continent collision zone – Implications for element transfer at the slab-wedge interface', *Geochimica et Cosmochimica Acta*, Vol. 61, pp. 1057–1072.
- D. Mouchel, R. Wordel, 1992, 'Measurement of low-level radioactivity in environmental samples by gamma-ray spectrometry', *International Journal of Radiation Applications and Instrumentation. Part A. Applied Radiation and Isotopes*, Vol. 43, pp. 49–59..
- R. Wordel et al., 1994, 'Investigation of the natural radioactivity in volcanic rock samples using a low background gamma-ray spectrometer', *Nuclear Instruments and Methods in Physics Research*, Vol. 339, pp. 322–328.

Part VI:
Radiation protection and
emergency response

For radioprotection and emergencies, it is important to measure not only high activities. There are many examples where low-level radioactivity measurements are also important. Cases 1 and 2 below will give two examples.

Another case is the following. In October 2017 many European labs started detecting ^{106}Ru in filters they used for monitoring radioactivity in air. This is an unusual radionuclide to detect in air, particularly when it is detected by itself and is not accompanied by other radionuclides. The activities were very low and posed no danger to human health in the areas in the EU where it was detected. Still, it was a mystery why it showed up. ^{106}Ru is used for the treatment of cancer of the eye, but it was unlikely that the release could come from production for such use. By collating data from European laboratories and comparing them with meteorological data, it was possible for researchers to claim with high probability that the release came from a specific region and was probably in the order of more than 100 TBq (100 trillion Bq) at the source of the release²⁹³⁰. So in more general terms one can state that detecting a very low level of activity could be indicative of a major problem further away.

(29) Institut de Radioprotection et de Sûreté Nucléaire, 2017, 'Detection of ruthenium 106 in France and in Europe: Results of IRSN's investigations', (https://www.irsn.fr/EN/newsroom/News/Documents/IRSN_Information-Report_Ruthenium-106-in-europe_20171109.pdf).

(30) O. Masson et al., 2019, 'Airborne concentrations and chemical considerations of radioactive ruthenium from an undeclared major nuclear release in 2017', Proceedings of the National Academy of Sciences of the United States of America, Vol. 116, No 34, pp. 16750–16759.

Radiation protection, Case 1: Response to the criticality accident in Tokaimura

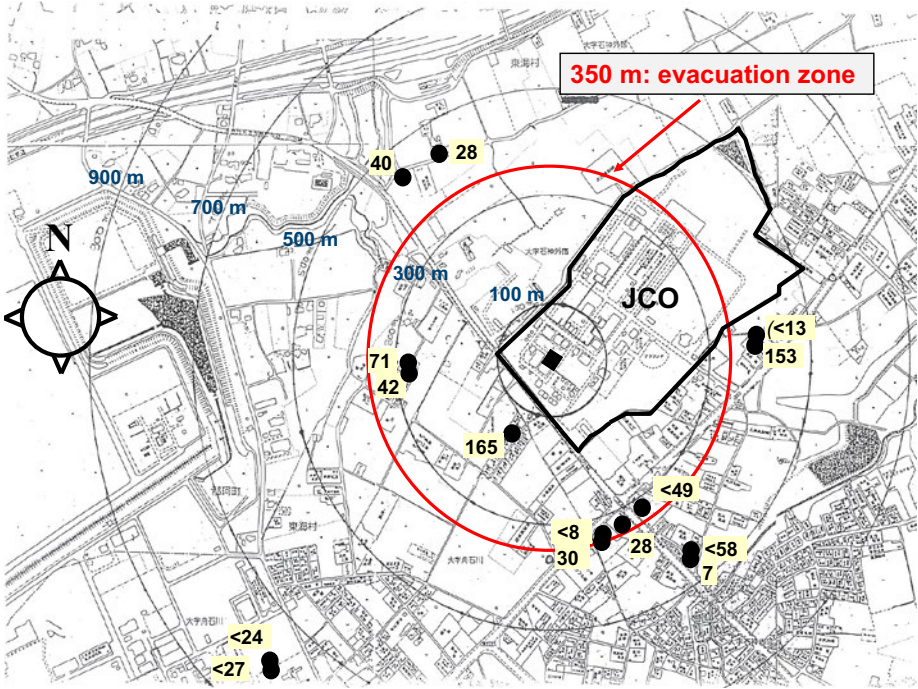
Following an accident in a nuclear fuel factory in Japan, the European Commission offered support. The only support Japan needed was underground measurements in HADES to determine the impact of the accident.

On 30 September 1999, a criticality accident took place in a fuel fabrication plant in Tokaimura, Japan. For about 20 hours, the plant and the surrounding village were irradiated with neutrons from a highly enriched uranium solution contained in a precipitation tank. This tank acted like a small reactor with a cooling mantle, which at least prevented contamination from spreading. The two workers who handled the solution received very high doses and died after a few weeks.

In response to previous nuclear accidents/incidents in Japan, an independent investigation team had been set up and led by Professor Komura of Kanazawa University. This team collected samples from houses in the surrounding village, and environmental samples such as soil. They concluded that contamination was very low but neutron irradiation was significant. To assess that, they performed retrospective analysis by measuring neutron-activated samples. On learning about HADES, Professor Komura enquired if it was possible to measure steel spoons and look for activation products. As it was, the President of the European Commission (at that time Romano Prodi) had already offered support. As short-lived radionuclides such as ^{51}Cr (28 days half-life) and ^{59}Fe (44 days half-life) were of interest, the measurements started immediately. The spoons were shipped by aeroplane, wrapped in cadmium and polyethylene to minimise activation from cosmogenic radiation during transport. The level of activation during transport was checked by sending blank samples, and was found not to influence the results. The long-lived radionuclide ^{60}Co (5.2 years half-life) was also used, even though it is common to find it as a contaminant in modern steel. However, the measured value of ^{60}Co would provide an upper limit for the neutron flux at the given locations.

The neutron flux varied greatly from one location to another, as the neutrons had to pass through walls, vehicles, vegetation, etc. Therefore, it was not possible to calculate the neutron flux in the different houses. Instead, measurements were necessary. The measurements triggered a special issue of the *Journal of Environmental Radioactivity*. Some of the results provided by the JRC can be seen in Figure 36.

Figure 36. Map of Tokaimura and the JCO factory. The numbers indicate neutron fluence values (in millions of neutrons per cm²) during the 20-hour accident that were derived from measurements of steel spoons in HADES.



Source: Gasparro et al. (2004). Licensed under CC BY 2.0.

FURTHER READING: CRITICALITY ACCIDENT IN TOKAIMURA

- J. Gasparro et al., 2004, 'Measurements of ⁶⁰Co in spoons activated by neutrons during the JCO criticality accident at Tokaimura in 1999', *Journal of Environmental Radioactivity*, Vol. 73, pp. 307–321.
- M. Hult et al., 2002, 'Thermal neutron fluence from ultra low level γ-ray spectrometry of spoons activated during the JCO criticality accident at Tokaimura in 1999', *Journal of Environmental Radioactivity*, Vol. 60, pp. 307–318.
- *Journal of Environmental Radioactivity*, 2000, Special issue on the Tokaimura accident, Vol. 50, No 1–2.

Radiation protection, Case 2: Solving the Hiroshima enigma

Validating data from Hiroshima helps to understand the effects of ionising radiation on the human body.

The cooperation between JRC and Japanese scientists during the measurements of samples from the Tokaimura accident (see Case 1) led to continued collaboration ⁽³¹⁾ in measuring samples exposed to the atomic bomb explosion in Hiroshima in 1945.

Humanity's understanding of the effect of ionising radiation on the human body is to a large extent based on the follow-up of 84 000 surviving victims from Hiroshima. By knowing the dose each of them received and following their medical history, it was possible to establish the relation between dose received and risk. Consequently, this research has greatly influenced the legislative process all over the world when it comes to radiation protection. Every time that a new reassessment of the victims' health and dose received was made, new recommendations for legislation were made.

Of course, none of the victims carried a dosimeter when the bomb exploded. However, everyone could say exactly where they were standing at the time of explosion. Japanese and American scientists in the Radiation Effects Research Foundation have been able to calculate the dose received by each survivor, based on knowledge of buildings.

These calculations need to be validated by measurements. The way to do that has been to collect samples from buildings and bridges that had an unobstructed view of the bomb when it exploded 580 m above the city. In these samples, scientists look for activation products, i.e. radionuclides produced by the neutrons from the bomb. Examples of such are ¹⁵²Eu in granite from bridges, ⁶³Ni from lightning rods and ⁶⁰Co in steel. Based on such measurements, scientists could produce curves in which the measured activity as a function of the distance from the hypocentre (the place on ground exactly below the point of explosion) can be studied and compared with values based on calculations.

After the reassessment in 1986 (Dosimetry System 86), it was clear that discrepancies of up to a factor of 100 appeared for ⁶⁰Co at locations 1 400–1 800 m from the hypocentre, where many of the survivors were located. In a first project in 2003, the JRC measured small (c. 100 g) samples of steel collected from bridges, handrails and pipes from roofs of buildings etc. The activity of ⁶⁰Co was very low, as 11 half-lives of ⁶⁰Co had passed since the activation. This meant that the ⁶⁰Co activity was about 2 000 times lower than in 1945. The measurements performed in HADES agreed perfectly with older measurements at distances from 0 m to 700 m from the hypocentre. However, at longer distances, JRC scientists could conclude that the actual ⁶⁰Co activity was lower than what was found in previous studies and

(31) This time the main collaboration was with the group of Professor Hoshi at Hiroshima University.

thus cast doubt on, and in fact refute the validity of, those former measurements. Furthermore, in a few cases, measurements in HADES could produce the distribution of ^{60}Co inside thick samples (Hult et al., 2012), which is important information when establishing the energy distribution of neutrons at a given location. All in all, the measurements in HADES managed to produce scientifically solid support for the presently used dosimetry system, Dosimetry System 02.

Figure 37. Left: this sample (before cleaning) from the city hall is a solid bar with a length of 418 mm. Right: this piece of a pipe from the Kyu-Fuzoku elementary school had a length of 298 mm, a diameter of 48 mm and a material thickness of 2 mm. It weighed 630 g. Before measurement in HADES it was cleaned and cut in smaller pieces.



Photo: M. Hult · J. Gasparro et al., 2010, JRC-Report EUR 24146 EN. Doi: 10.2787/22773.

FURTHER READING: HIROSHIMA ENIGMA

- J. Gasparro et al., 2010, *Measurements of ^{60}Co in massive steel samples exposed to the Hiroshima atomic bomb explosion*, JRC Report EUR 24146 EN. Doi: 10.2787/22773. Publication Office of the European Commission.
- J. Gasparro et al., 2012, 'Measurements of ^{60}Co in massive steel samples exposed to the Hiroshima atomic bomb explosion', *Health Physics*, Vol. 102, No 4, pp. 400–409, doi: 10.1097/HP.0b013e31823a172e.
- M. Hoshi et al., 2009, 'Atomic bomb radiation dosimetry in Hiroshima and Nagasaki and its meanings' (in Japanese), in: *Proceedings of the 52nd Annual Meeting of the Japan Radiation Research Society, Hiroshima, Japan*, p. 62.
- M. Hult et al., 2004, 'Deep underground measurements of ^{60}Co in steel exposed to the Hiroshima atomic bomb explosion', *Applied Radiation and Isotopes*, Vol. 61, pp. 173–177.
- M. Hult et al., 2012, 'Distribution of ^{60}Co in steel samples from Hiroshima', *Applied Radiation and Isotopes*, Vol. 70, pp. 1974–1976.

Part VII:
Miscellaneous applications

It is only human imagination and ingenuity that limit the usefulness of underground radioactivity measurements. Here follow examples of nuclear science applications different from those mentioned in Parts I–VI.

Detecting fission products in CTBTO filters

The preparatory commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO; <https://www.ctbto.org/>) was founded in 1996. Its main office lies in Vienna and is in charge of operating the International Monitoring System (IMS). This is a worldwide network of sampling and measurement stations. The purpose of the IMS is to enable detection of nuclear weapon testing.

One of the many analytical techniques in the IMS is the systematic measurement of air filters, using gamma-ray spectrometry. With gamma-ray spectrometry one can detect certain fission products with a high degree of sensitivity.

The CTBTO-approved laboratory in Richland, USA (Pacific Northwest National Laboratories), conducted a study in which it requested input from a number of underground laboratories. The aim was to investigate how much improvement in detection limits of fission products could be expected from typical CTBTO filters if they were to be measured in underground laboratories. The study gave the CTBTO a solid basis for understanding the achievable improvement in underground laboratories for its typical samples. This will help the organisation at a later stage to take a well-founded decision on whether to use underground laboratories or not.

FURTHER READING: DETECTING FISSION PRODUCTS

- C. Aalseth et al., 2009, 'Ultra-low background measurements of decayed aerosol filters', *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 282, pp. 731–735.

Monitoring radioactivity in lung cells and faeces from uranium miners

In 2003, the CELLAR collaboration was asked by Verein für Kernverfahrenstechnik und Analytik Rossendorf (VKTA) to provide underground measurements to support a project initiated by the German Federal Office for Radiation Protection. The aim was to verify radioanalytical methods used by the VKTA to analyse freeze-dried faeces from people who used to work in the former uranium mines in Saxony. The VKTA has a very important role in monitoring radioactivity in the people and environment around the former uranium mines operated by the former Wismut company.

^{238}U has a long decay chain and goes via the inert gas ^{222}Rn eventually to the long-lived (22 years half-life) ^{210}Pb . This lead isotope has been used by many scientists as a marker for retrospective assessment of radon exposure. In a pre-study (Mouchel and Wordel, 1996), the possibility of non-destructively detecting ^{210}Pb in lung cells from former uranium miners by using HADES was investigated. The positive outcome triggered measurements in 2001 of a few human lung cell samples from former Wismut miners that were provided by the German cancer research centre in Heidelberg via the VKTA. Each sample mass was only about 1.5 g, and the measured massic activities ranged from 1.7 mBq/g to 16 mBq/g. The conclusion was that it is possible to detect ^{210}Pb , but a larger study must not wait too long given that the half-life of ^{210}Pb is only 22 years.

FURTHER READING: DETECTING FISSION PRODUCTS

- J. Gasparro et al., 2003, *Results of ultra low-level γ -ray spectrometry measurements of freeze-dried human faeces*, JRC technical report GE/RN/04/2003/05/23. Printed by JRC-Geel.
- M. Hult et al., 2006, 'Underground gamma-ray spectrometry', *Acta Chimica Slovenica*, Vol. 53, pp. 1-7, <http://acta-arhiv.chem-soc.si/53/53-1-1.pdf>.
- D. Mouchel, R. Wordel, 1996, 'A low-energy HPGe detector dedicated to radioactivity measurements far below the environmental levels', *Applied Radiation and Isotopes*, Vol. 47, pp. 1033-1041.

Benchmarking German steel

In another CELLAR project from 2003, the VKTA was consulted by the German Federal Office for Radiation Protection. The project served to investigate the present situation of radioactivity contamination (mainly ^{60}Co) in German steel.

Every year, a number of radioactive sources are accidentally lost from regulatory control and some of them end up in metal industry in the form of recycled items. In Europe in general and Germany in particular, the control of radioactivity in scrap metals has been much improved. Therefore, it was expected that the ^{60}Co levels in steel should be low.

In fact, the ^{60}Co activity in German steel in 2003 was so low that in order to obtain numbers instead of just limits it was necessary to use underground laboratories for certain steel samples (Köhler et al., 2004). Furthermore, it was important to create a benchmark for the ^{60}Co activity in steel at a certain time for several reasons.

- International trade in scrap metal has increased, which is important for a circular economy.
- EU legislation allows the free release of scrap from nuclear decommissioning, an activity that is increasing in Europe today.
- Underground science experiments such as those using Borexino and GERDA as well as smaller ones require extremely radiopure metals. The study was useful to see if commercial German steel had activity levels that could be accepted in these experiments.

FURTHER READING: BENCHMARKING GERMAN STEEL

- M. Köhler, M. Hult, D. Arnold, M. Laubenstein, J.-L. Reyss, 2004, 'Reference measurements of low levels of ^{60}Co in steel', *Applied Radiation and Isotopes*, Vol. 61, pp. 207–211.

Creating new opportunities

Members of the network CELLAR meet regularly to discuss collaboration projects and new ideas.

One interesting suggestion is to use underground laboratories to check the activation of aeroplane parts in a manner similar to what was described in the chapter on neutron and plasma physics (Part IV, Case 2). By measuring the activity induced by cosmic rays in aeroplane parts, one can obtain information on how long a certain part has been in use. This is very important in order to check for possible fraud. Trade in used parts has a big market. In some cases (e.g. electronics) one can also mistake an old part for a new one, a case that can be resolved by analysing cosmogenic activation.

Another application that has been discussed is in nanoscience. It is very difficult to assess the impact of nanoparticles of titanium and silver when they enter the environment. How are they taken up by biota and wildlife? Which organs are most affected? One can design experiments in which, for example, mice are exposed to neutron-activated nanoparticles. The activity will be low and not affect the health of the animals. However, having gamma-ray-emitting nanoparticles will greatly facilitate detecting their distribution within the bodies of mice or within certain plants.

A third example, which is already being developed at the Canfranc underground laboratory in Spain, is to study the effect of very low doses, far below environmental levels. Could it be so that too low doses are detrimental for animals and biota? Or is it better to strive to minimise the exposure, no matter what? Performing such an experiment is not trivial, and will require selection of radiopure materials in the cages. Furthermore, all items, including food, will have to be checked for radioactivity. It also requires control groups exposed to an identical environment but with higher levels of radiation to be set up.

Furthermore, if we look at natural processes in nature and industrial processes, there is most likely a plethora of future work that will benefit from this relatively new technology. It is the experience of the JRC team that, when more information about the possibilities in underground science is spread, more useful applications turn up.

Figure 38. Photo not directly linked to this case study. All shields in HADES are composed of lead (old lead inside, newer lead outside) and freshly produced electrolytic copper. Here is the shield of Ge5 open when a sample is changed.

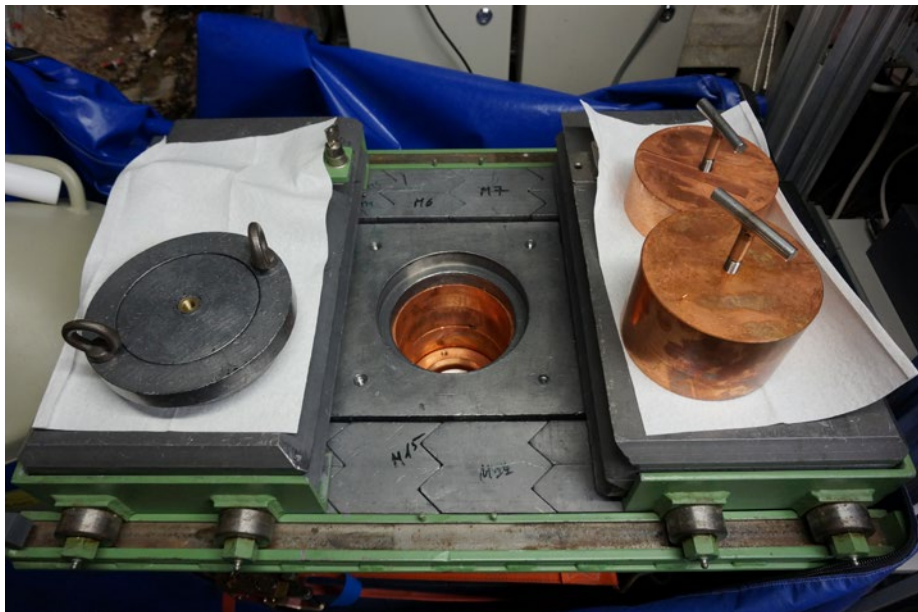


Photo: G. Lutter.

Outlook

Capacity building for knowledge production

COLLABORATION OF EUROPEAN LOW-LEVEL UNDERGROUND LABORATORIES

Since 2000, the European laboratories that perform dosimetry and radioactivity measurements in underground laboratories have collaborated in a network called CELLAR. Key aspects of the collaboration are linked to:

- mutual use of each other's infrastructure, especially when facing capacity challenges ⁽³²⁾;
- sharing of experiences of radiopure materials;
- sharing of experiences linked to operation of underground facilities.

WORLDWIDE LABORATORIES

Looking outside Europe, the main driving force behind new underground laboratories has been large-scale experiments in fundamental physics, studying neutrinos, dark matter, double beta decays and gravitational waves. We see increased interest in operating smaller laboratories for radioactivity measurements in a style similar to HADES. Work in such small labs has become essential for supporting large-scale experiments, and some are striving to become multidisciplinary.

MONTE CARLO SIMULATIONS

In the past 20 years, the use of Monte Carlo simulations has become increasingly important for low-level gamma-ray spectrometry. It is used for the following:

- calculating correction factors linked to efficiency transfer and/or coincidence summing,
- calculating locations of background radiation,

(32) The main drawback when measuring mBq levels of radioactivity or lower is the time-consuming measurement. A mBq is about one decay per hour. Assuming a 10 % detection efficiency, it takes 100 hours to collect 10 counts. Therefore, it is important to have many detectors.

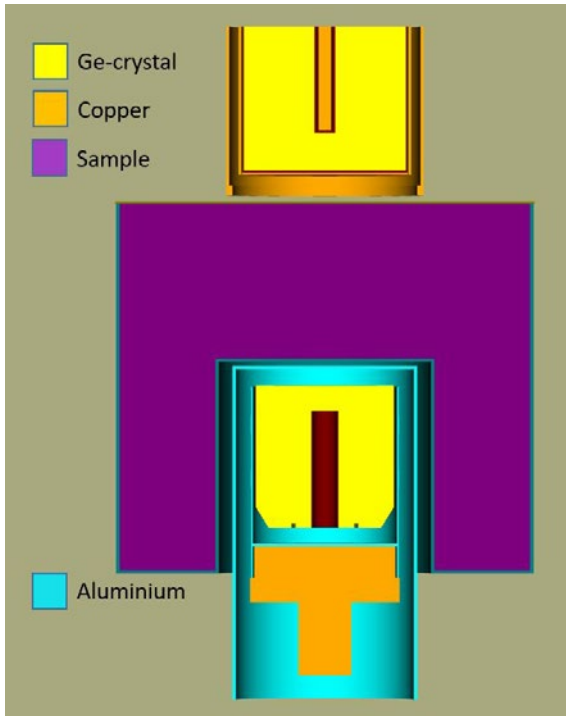
- calculating the optimal geometry for samples and detectors for different radionuclides,
- improving the design of detectors.

These simulations have led to measurements with increased efficiency, lower background radiation and increased accuracy of measurements.

There is still development ongoing in this field, which will eventually lead to better measurements. In the coming years we will see the following:

- simulations with higher numbers of histories and increased computing power;
- models with increased levels of detail;
- models that include inhomogeneous dead layer structures;
- models that include the electric field inside the crystals.

Figure 39. Cross-sectional view of a computer model of a sample in a Marinelli beaker measured in a dual detector system.



Source: G. Lutter.

Conclusions

Developing science to trigger new policies

Developments over the past 20 years show that underground gamma-ray spectrometry is not solely useful for physicists working on fundamental physics, although this was the prime driving force at the start of this field.

The underground facility HADES, partly through the JRC open access scheme and its exploratory research instrument, has been leading the development of diversifying the science applications that can benefit from underground radioactivity measurements. Many institutes are now looking for existing nearby underground locations that can be used for this type of work, which is much less costly than constructing an underground laboratory from scratch. One can, for example, think of abandoned tunnels that were used as support tunnels during tunnelling work. Alternatives that can be put to use are abandoned mines, bomb shelters, etc.

Seeing that measurements of mBq levels of radioactivity require long measurement times, it is essential to have many detectors available. Judging from current developments in Europe, it is not unlikely that in 10 years from now there could be more than 100 underground HPGe detectors in operation in Europe. Given that researchers from all fields of science have access to such specialised equipment, one can expect the generation of knowledge in many fields to be substantial – as exemplified by the selection of cases in this report.

Appendix 1:

Glossary

Anthropogenic	Induced by human activities.
BIPM	Bureau International des Poids et Mesures (www.bipm.org), the intergovernmental organisation that is in charge of defining and realising all the units in the international system of units (SI). It was created in 1875 following the signing of the Metre Convention.
CBNM	Central Bureau for Nuclear Measurements, the name of JRC-Geel 1960–1992.
CCRI	Comité Consultatif de Rayonnement Ionisant, the BIPM entity in charge of radioactivity, dosimetry and neutron metrology.
CELLAR	Collaboration of European Low-level Underground Laboratories, a European network collaborating on advancing underground science.
CRM	Certified reference material.
CTBT	Comprehensive Test Ban Treaty.
EIG	Economic interest grouping.
EUFRAT	European research infrastructure for nuclear reaction, radioactivity, radiation and technology studies in science and applications. The JRC open access scheme at the G2-unit of JRC-Geel including
EURAMET	European Association of National Metrology Institutes.
EURIDICE	European underground research infrastructure for disposal of nuclear waste in a clay environment.
GERDA	GERmanium Detector Array. An international science collaboration to measure the double beta decay of ⁷⁶ Ge in Gran Sasso National Laboratory, Italy.
H2020	Horizon 2020.
HADES	High-activity disposal experimental site.
HPGe	High-purity germanium.
IAEA	International Atomic Energy Agency.
IRMM	Institute for Reference Materials and Measurements, the name of JRC-Geel 1992–2016.
ILC	Interlaboratory comparison, defined in ISO 17043:2010 as the organisation, performance and evaluation of measurements on the same or similar items by two or more laboratories in accordance with predetermined conditions.

ISO	International Standardisation Organisation (www.iso.org), an international standardisation organisation with mandates within similar fields to those of the European Committee for Standardisation (CEN), i.e. standards linked to methods.
JRC	Joint Research Centre, a directorate-general of the European Commission.
Primordial	Something that has existed ever since the formation of Earth, 4.5 billion years ago.
PT	Proficiency Test. Defined in ISO17043:2010 as the evaluation of participant performance against pre-established criteria by means of ILC.
PTBT	Partial Test Ban Treaty.
RM	Reference material.
RN	Radionuclide metrology team.
SI	The international system of unit, which is maintained and realised by BIPM.
SCK CEN	StudieCentrum voor Kernenergie Centre d'Etudes Nucleaire (SCK CEN)
SMEs	Small and medium-sized enterprises.
TAEK	Turkish Atomic Energy Authority.
URF	Underground research facility.
VKTA	Verein für Kernverfahrenstechnik und Analytik Rossendorf.
w.e.	Water equivalent, used for comparing the overburden (e.g. sand, clay, rock, concrete) of different underground laboratories with regard to the reduction of the cosmogenic muon flux.

Appendix 2: The Euratom Treaty

Article 8 says that the Commission shall establish a Joint Nuclear Research Centre.

ARTICLE 8:

1. After consulting the Scientific and Technical Committee, the Commission shall establish a Joint Nuclear Research Centre.

This Centre shall ensure that the research programmes and other tasks assigned to it by the Commission are carried out.

It shall also ensure that a uniform nuclear terminology and a standard system of measurements are established.

It shall set up a central bureau for nuclear measurements.

2. The activities of the Centre may, for geographical or functional reasons, be carried out in separate establishments.

The Central Bureau for Nuclear Measurements (CBNM) was the name of JRC-Geel between 1960 and 1993. Ensuring a uniform nuclear terminology and standard system of reference has a wider meaning that encompasses providing radioactive reference measurements and reference materials in support of harmonisation and international equivalence, which JRC-Geel does in numerous ways. The RadioNuclide Metrology Team (RN) was the response to these tasks as well as to the specific phrase in the first programme of the Euratom treaty: 'to perform absolute measurements of radiation'. The RN was the first operational team of the CBNM, as it began its work as early as 1959 with offices located at the premises of the Belgian nuclear centre SCK CEN (<https://www.sckcen.be/en>). In 1960 the radionuclide metrology laboratory at the site of the CBNM commenced operation and staff moved from SCK CEN to CBNM.

In 1993 CBNM changed its name to the Institute for Reference Materials and Measurements (IRMM) to reflect the increased importance of non-nuclear activities on site. In 2016 the name changed again, this time to JRC-Geel to better promote the corporate identity of the JRC. Today, the JRC operates at several sites, namely in Brussels (Belgium), Karlsruhe (Germany), Seville (Spain), Ispra (Italy), Petten (Netherlands) and Geel (Belgium).

Article 6 says that the Commission is encouraged to place installations, equipment and expert assistance at the disposal of the Member States.

ARTICLE 6:

To encourage the carrying out of research programmes communicated to it the Commission may:

- (a) provide financial assistance within the framework of research contracts, without, however, offering subsidies;*
- (b) supply, either free of charge or against payment, for carrying out such programmes, any source materials or special fissile materials which it has available;*
- (c) place installations, equipment or expert assistance at the disposal of Member States, persons or undertakings, either free of charge or against payment;*
- (d) promote joint financing by the Member States, persons or undertakings concerned.*

In response to this article, the JRC has operated an open access scheme, in which HADES is an essential part, since 2014. In 2014–2019, there were 29 applications to use the detectors in HADES. The applications are evaluated twice a year by the open access panel of external experts for the European research infrastructure for nuclear reaction, radioactivity, radiation and technology studies in science and applications.

At the time of writing this report (April 2020), 12 projects have been accepted and finalised, 4 accepted projects are still ongoing and 4 new ones are about to start.

Article 4 and Annex I specify in which fields of nuclear research the Commission will be involved.

EXTRACT FROM ARTICLE 4:

1. The Commission shall be responsible for promoting and facilitating nuclear research in the Member States and for complementing it by carrying out a Community research and training programme.
2. The activity of the Commission in this respect shall be carried out within the fields listed in Annex I to this Treaty.

EXTRACTS FROM ANNEX I REFERRED TO IN ARTICLE 4:

V. Applications of radioisotopes

Application of radioisotopes as active elements or tracers in:

- (a) industry and science;
- (b) medicine and biology;
- (c) agriculture.

VI. Study of the harmful effects of radiation on living organisms

1. Study of the detection and measurement of harmful radiations.
2. Study of adequate preventive and protective measures and the appropriate safety standards.
3. Study of the treatment of radiation effects.

VII. Equipment

2. (a) instruments for radiation detection and measurement, used particularly in:
 - prospecting for minerals,
 - scientific and technical research,
 - reactor control,
 - health and safety

In many respects, HADES facilitates nuclear research for scientists in EU Member States, as shown in this casebook.

A detailed work programme with key orientations is established each year for the JRC by the Commission. These key orientations reflect the work the JRC will carry out and are structured in accordance with the political priorities of the Commission (<https://ec.europa.eu/info/strategy/priorities-2019-2024>) and following specific objectives set out in the framework programmes (https://ec.europa.eu/eurostat/cros/content/research-projects-under-framework-programmes-0_en and https://ec.europa.eu/info/horizon-europe-next-research-and-innovation-framework-programme_en).

Articles 30–39 (= Chapter 3) specify that basic standards are to be laid down within the Community for the protection of the health of workers and the general public against the dangers arising from ionising radiation.

ARTICLE 35:

Each Member State shall establish the facilities necessary to carry out continuous monitoring of the level of radioactivity in the air, water and soil and to ensure compliance with the basic standards.

The Commission shall have the right of access to such facilities; it may verify their operation and efficiency.

EXTRACT FROM ARTICLE 39:

The Commission shall set up within the framework of the Joint Nuclear Research Centre, as soon as the latter has been established, a health and safety documentation and study section.

This section shall in particular have the task of collecting the documentation and information referred to in Articles 33, 36 and 37 and of assisting the Commission in carrying out the tasks assigned to it by this Chapter [Chapter 3].

Article 39 basically says that the JRC shall support the Commission in its work on Chapter 3 (Articles 30–39). In HADES, work on characterising reference materials used in proficiency tests in support of Article 35 has been one such activity (Hult et al., 2019). In retrospect, one can conclude that it was a piece of great foresight and insight to introduce monitoring of the environment already in 1957 in the Euratom Treaty. It was, however, not until after the Chernobyl accident in 1986 that the importance of good-quality monitoring was fully realised and greater political acceptance was gained (Hult et al., 2019). Since then, environmental radioactivity monitoring has increased in importance all over the world. There are many positive effects from monitoring; for example:

- regular monitoring enables authorities to give good advice about sound actions at short notice whenever a nuclear accident takes place;
- the data from environmental monitoring (which are now increasingly made publicly available) are highly useful for many scientists in their work, e.g. on studying dispersion models;
- the data can contribute to checking undeclared nuclear weapon testing;
- having a good baseline (knowing the activity of different radionuclides present in the environment today) enables us to better assess the impact of future nuclear activities or accidents.

FURTHER READING: THE EURATOM TREATY

- Hult, M., Jobbagy, V. and Sobiech-Matura, K., 2019, Radioactivity monitoring: How the JRC verifies results from monitoring within the European Union — A quick guide, EUR 29509 EN, Publications Office of the European Union, Luxembourg, ISBN 978-92-76-08762-5, doi:10.2760/124550, JRC 117258. available for download from: <https://dx.doi.org/10.2760/124550>
- [Consolidated version of the Treaty establishing the European Atomic Energy Community \(https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A12012A%2FTXT\)](https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A12012A%2FTXT)

Appendix 3:

Thesis work and staff

MASTER OF SCIENCE (MSC) THESIS (WITH ASPECTS OF INPUT FROM HADES)

- **Hasselt University, Belgium.** Quinten Remijssen (2015), Stef Geelen (2018)
- **Lund University, Sweden.** Christoffer Ellmark (2005)

DOCTORAL (PHD) THESIS, EMPLOYED BY JRC (WITH ASPECTS OF INPUT FROM HADES)

- **Valencia University, Spain.** Maria José Martínez Canet, 'Measurement of low neutron fluxes using activation technique and ultra low-level gamma-ray spectrometry' (1997–2001)
- **Jyväskylä University, Finland.** Elisabeth Wieslander, 'A new gamma-ray spectrometry system for measurements of radioactivity in the micro-becquerel range' (2005–2009)

PHD THESIS, NOT EMPLOYED BY JRC (WITH ASPECTS OF INPUT FROM HADES)

- **Ruperto-Carola-University of Heidelberg, Germany.** Marco Salathe, 'Study on modified point contact germanium detectors for low background applications' (2015)
- **University of Padua, Italy.** Katharina von Sturm, 'Confined event samples using Compton coincidence measurements for signal and background studies in the Gerda experiment' (2016)
- **Technical University Dresden, Germany.** Björn Lehnert, 'Search for $2\nu\beta\beta$ excited state transitions and HPGe characterization for surface events in GERDA Phase II confined event samples using Compton coincidence measurements for signal and background studies in the GERDA experiment' (2016)
- **Technical University, Munich, Germany.** Thomas Bode, 'The neutrinoless double beta decay experiment GERDA Phase II: A novel ultra low background contacting technique for germanium detectors and first background data' (2016)
- **KU Leuven, Belgium.** Shinichiro Uematsu, 'Radiocaesium transfer to crops in

the Fukushima affected environments: A soil chemical and plant physiological approach' (2017)

- **Technical University, Munich, Germany.** Heng-Ye Liao, 'Development of pulse shape discrimination methods for BEGe detectors' (2017)
- **Danish Technical University, Denmark.** Nikola Markovic, 'Coincidence methods in gamma-ray spectrometry for radioecological applications' (2018)
- **University of Salamanca, Spain.** Begoña Quintana Arnes, 'Optimisation of gamma spectrometry techniques for the measurement of low level activities' (1994)
- **Jagiellonian University, Krakow, Poland.** Krzysztof Panas, 'Background suppression in high purity germanium detectors used in the GERDA experiment' (2018)
- **Hasselt University, Belgium.** Tom Croymans, 'Valorization of Fe-rich industrial by-products in construction materials: A radiological assessment' (2018)

JRC STAFF ⁽³³⁾ WORKING IN HADES

- **Present HADES group.** Gerd Marissens (lab responsible), Heiko Stroh (deputy lab responsible), Guillaume Lutter (scientist), Mikael Hult (group leader)
- **Former postdoctoral contractors.** Faidra Tzika, Patric Lindahl, Erica Andreotti, Raquel Gonzalez-Orduña, Joël Gasparro
- **Former PhD students.** Maria José Martínez Canet, Elisabeth Wieslander
- **Former visiting scientists.** Matthias Köhler (VKTA, Germany), Peter N. Johnston (RMIT University, Australia), Werner Preusse (Chemnitz 2, Landesmessstelle für Radioaktivität, Germany), Namik Sahin (TAEK, Turkey), Ayan Yüksel (TAEK, Turkey)
- **Former group leader 1992–1998.** Rainer Wordel
- **Former HADES technicians.** Daniel Mouchel, José das Neves, Roberto Vasselli

(33) All from the RN of Unit G2 (formerly D4), 'Standards for Nuclear Safety, Security and Safeguards'.

Appendix 4: References

A list of references to scientific articles published in international refereed journals involving HADES (completely or to some extent). The references are listed in chronological order, following the organisation of chapters in this report.

INSTRUMENTATION, METROLOGY, HADES DESCRIPTIONS, LOW-LEVEL OVERVIEWS

1. D. Mouchel et al., 1995, 'A low-energy HPGe detector dedicated to radioactivity measurements far below the environmental levels', *Applied Radiation and Isotopes*, Vol. 47, pp. 1033-1041.
2. R. Wordel, 1996, 'Study of neutron and muon background in low-level germanium gamma-ray spectrometry', *Nuclear Instruments and Methods in Physics Research Section A*, Vol. 369, pp. 557-562.
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4. B. Quintana et al., 1996, 'Automatic analysis of continuum phenomena in gamma spectra', *Applied Radiation and Isotopes*, Vol. 47, pp. 911-917.
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6. M. Hult et al., 2003, 'Underground measurements of environmental radioactivity in Europe', in: N. Kamata (ed.), *Proceedings of the International Symposium of the Kanazawa University 21st Century COE Program*, Kanazawa University, pp. 18-23.
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