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Abstract

The present overview report show that there are many existing and potential sources of radioactive particle contamination of relevance to the Nordic countries. Following their release, radioactive particles represent point sources of short- and long-term radioecological significance, and the failure to recognise their presence may lead to significant errors in the short- and long-term impact assessments related to radioactive contamination at a particular site. Thus, there is a need of knowledge with respect to the probability, quantity and expected impact of radioactive particle formation and release in case of specified potential nuclear events (e.g. reactor accident or nuclear terrorism). Furthermore, knowledge with respect to the particle characteristics influencing transport, ecosystem transfer and biological effects is important. In this respect, it should be noted that an IAEA coordinated research project was running from 2000-2006 (IAEA CRP, 2001) focussing on characterisation and environmental impact of radioactive particles, while a new IAEA CRP focussing on the biological effects of radioactive particles will be launched in 2008.

Key words

radioactive particles, sources, characteristics

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Overview of sources of radioactive particles of Nordic relevance as well as a short description of available particle characterisation techniques

an NKS HOT II report

by

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1 Introduction

Contamination of the environment from anthropogenic (man-made) radionuclides commenced with the nuclear era. The major source of fission and activation products in the environment is the global fallout from 543 atmospheric nuclear weapon tests conducted during 1945 – 1980 (UNSCEAR, 1993; UNSCEAR, 2000). Other important anthropogenic sources include accidental and authorised releases at sites associated with the nuclear fuel cycle as well as releases from satellite, aircraft and submarine accidents and dumping of radioactive waste at sea. Among artificially produced long-lived radionuclides released to the environment, ^{137}Cs, ^{90}Sr, ^{241}Am and Pu isotopes are of major radiological concern, even though several other radionuclides (^{99}Tc, ^{129}I, ^{237}Np, ^{3}H, ^{14}C, etc.) are also released from nuclear installations (Salbu, 2000b). As contended by Salbu (2000a), a significant fraction of the refractory (i.e. non-volatile) radionuclides released during nuclear events are associated with radioactive particles.

This report provides an overview of sources reported to have contributed to radioactive particle contamination in the Nordic environment or in areas of relevance to the Nordic countries and the characteristics of observed particles.

2 Nuclear weapon test fallout

The main source of anthropogenic radionuclides in the environment is fallout from 543 atmospheric nuclear weapon tests (UNSCEAR, 1993; UNSCEAR, 2000). Authorized or accidental releases from the nuclear fuel cycle, especially reprocessing plants and nuclear reactors also contribute significantly. Releases from satellite, aircraft and submarine accidents and dumping of radioactive waste at sea have a local impact only.

History of nuclear weapons testing

The first nuclear device ever detonated was the Trinity test performed by the US on the 16th July 1945. Most of subsequent tests were performed by the US and former USSR (UNSCEAR, 2000). In 1963, these two countries (and others) entered the partial test ban treaty (PTBP) prohibiting all testing of nuclear weapons/devices in the atmosphere, outer space and under water. The PTBT permits underground testing if the explosions do not cause radioactive debris to be present outside the territorial limits of the state conducting the tests, and hence the majority of the tests after 1963 have been performed underground. Two countries, France and China, conducted atmospheric tests after 1963. France ended their atmospheric testing in 1970 and China in 1980 (UNSCEAR, 2000). Fig. 2-1 displays the number and yields of tests performed underground and in the atmosphere until year 2000. The sites used for atmospheric testing are shown in Fig. 2-2.

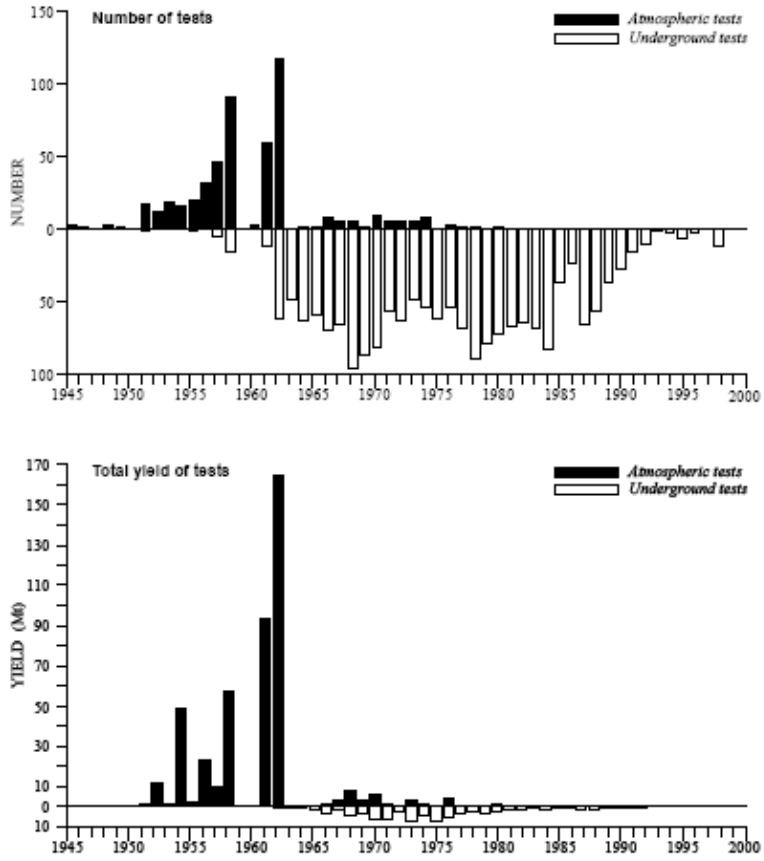

Fig. 2-1. Tests of nuclear weapons in the atmosphere and underground (UNSCEAR, 2000).

Fig. 2-2. Sites where atmospheric tests of nuclear weapons have taken place since 1945 (AMAP, 1998). The sites include: Nevada test site, Johnston Atoll (not shown), Christmas Island (United States (US)); Bikini and Eniwetok Island (US tests in Marshall Islands); Novaya Zemlya (Former Soviet Union (FSU) tests in Russia); Semipalatinsk (FSU tests in Kazakhstan); Monte Bello and Maralinga (UK tests in Australia); Mururoa (France); Reggane (French tests in Algeria); Lop Nor (China).

Particle formation and dispersion

The predominant part of radioactive material produced in the explosion of a nuclear device is associated with particles. Atmospheric explosion of a nuclear device produces a cloud of vaporized material (fireball). The cooling of the fireball results in formation of particles via

nucleation and condensation in the gas phase. This is followed by growth of the particles by coagulation of further material on the particles (Edvardson, 1965). Convective forces initiated by the fireball results in transport of air and debris into the fireball. In low altitude detonations, large amounts of ground material is sucked into the fireball and completely or partially vaporized or melted. Particles of this material also serve as nuclei on which condensation of radioactive nuclides can occur. The size distribution of the formed particles is highly dependent on the altitude, explosion yield and the environment of the detonation (i.e., the composition of the ground material). In general, the radioactive particles formed in high altitude detonations are smaller (0.01 – 20 μm) than the particles formed via condensation on debris from the ground (up to a few mm) (Crocker et al., 1966; RADIAK, 1996).

The processes influencing the deposition of radioactive particles on the ground are dry deposition, i.e., gravitational settling and downward transport by turbulence and mixing by large weather systems, and wet deposition by rain or snow. Dry deposition is dominated by settling for larger dense particles (diameter >20 μm). Table 1 contains examples of calculated velocities and corresponding descending times for particles of different sizes.

The yield of the explosion influences the height and size of the fireball which in turn influences the descending time of the particles. For example, particles from a surface burst with a yield of a few kt will fall from a height of about 2 km, while the corresponding height for a Mt yield explosion will be about 20 km (RADIAK, 1996). Wet deposition is most efficient for relatively weak detonations since the thickness of the atmospheric compartment affected by wet deposition usually is less than 10 km. Hence, most of the debris from a high-yield detonation will be above this altitude (RADIAK, 1996).

The fallout from detonation of nuclear devices can, according to UNSCEAR (2000), be categorized as:

- local fall-out (mainly large particles from surface bursts deposited within hours)
- intermediate fallout (smaller particles from surface bursts deposited up to several thousand km downwind with a retention time of days)
- global fall-out (consists of small particles from high air-bursts or surface bursts injected into the stratosphere with retention times from 3-24 months)

Table 1. Calculated settling velocities for spherical particles with a density of 2.6 g/cm^3. The velocities used are typical for conditions at altitude 5 km. The presented deposition times are from an altitude of 10 km (RADIAK, 1996).

Radius (μm)	Settling velocity (cm/s)	Descending time
1000	1500	10 min
100	160	1.5 h
10	3.5	3 d
1	0.035	1 year[1]

In the formation of radioactive particles, fractionation, i.e., enrichment or depletion, of radioactive nuclides in relation to theoretical fission and activation yields can occur. Important parameters influencing the fractionation are the vapor pressure of the radioactive

[1] Please note that these calculations are based on gravitational settling only and that for smaller particle sizes other mechanisms are more important than gravitational settling.

nuclides, the vapor pressure of the major constituents of the gas-phase and the composition of the gas-phase in the fireball (Edvarson, 1965; Holmberg and Andersson, 1968). There are, for example, several isotopes of highly volatile elements such as I, Br, Xe and Kr with high fission yields and short half-lives that are an early part of decay chains producing more long-lived nuclides. Two examples of such decay-chains are presented in Fig. 2-3. Since e.g., the noble gases will not condense on particles, this will influence the condensation of the more long-lived daughter nuclides both with respect to the final composition of the particles as well as the distribution of the nuclides associated with the particles. Furthermore, the explosion yield and altitude will influence the temperature and cooling of the fireball and, as mentioned above, the formation and deposition of particles, which in turn also influences the fractionation. In for example a surface burst, the local fall-out that deposit early may be depleted in volatile elements while the more widely spread fall-out may be enriched (Crocker et al., 1966; Eisenbud and Gesell, 1997).

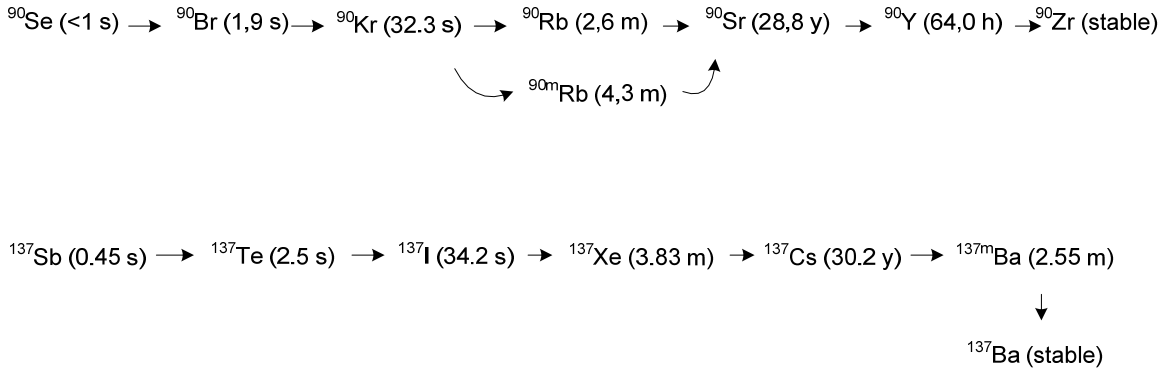

Figure 2-3. The decay-chain and half-lives of nuclides with mass number 90 and 137 ($T_{½}$ obtained from (National Nuclear Data Center (NNDC), 2007).

Air sampling by the Swedish Defence Research Agency

In 1955 the former Research Institute of National Defence (now Swedish Defence Research Agency) began collecting and examining debris from nuclear weapons tests. The sampling was done using air-filters both at ground level and by air-craft at high altitudes. The research initially done involved for example methods development for autoradiographic examination of individual particles (Sisefsky, 1961), studies of fractionation of fissions products in the debris (Edvarson et al., 1959) and age and origin determination of debris (Edvarson and Low, 1960). The origin of the debris could be identified to be, e.g., from the series of tests conducted by USSR in Novaya Zemlya as well as the Windscale accident.

With the PTBP entering into force, the massive testing by the US and USSR stopped, while China begun their period of testing in 1964. China performed 22 tests above ground over a time period of 16 years. Debris from all Chinese tests conducted above ground was collected and examined in Sweden. These tests were of different types and well separated in time, and hence provided a good opportunity to study debris from individual detonations. Parameters such as size and structure of particles, specific activity and fractionation were determined and used to draw conclusions on explosion type (De Geer, 1996).

The air sampling program at FOI is still on-going with two purposes; both as a part of the comprehensive nuclear test ban treaty (CTBT) verification regime and as a national program focussed on radiological protection. The national program consists of six air filter stations

located from Kiruna in the north to Ljungbyhed in the south of Sweden. Some of these are also equipped with charcoal cartridges for iodine sampling and/or funnels for collection of precipitated material for determinations of total deposition. Gamma-spectrometric analyses of, i.e., ^{7}Be, ^{22}Na, ^{210}Pb and ^{137}Cs are performed routinely on the filters (Söderström et al., 2006).

In other Nordic countries there are also stations in operation for monitoring airborne particulate radionuclides at low concentration levels. Digital autoradiography of archived air filter samples collected by Forsvarets Forskningsinstitutt (FFI) on various locations in Norway during the 1960s demonstrate numerous hot spots indicating radioactive particles (Salbu et al, unpublished).
In 1987, after a leakage from a Soviet underground nuclear weapons test at Novaya Zemlya, several countries could detect the event at their national stations (Bjurman et al., 1990).

3 Nuclear accidents

Although most nuclear accidents have had a local impact only, some events (fires, explosions, re-entry of satellites) have resulted in global or regional contamination. When nuclear accidents occur, releases of refractory radionuclides will indicate the presence of fuel particles (Salbu, 2000a).

Nuclear reactor accidents

Nuclear reactors with their considerable inventories of radioactive material are potential sources of environmental contamination. Several incidents and some accidents have occurred, but in three cases radioactive particle releases are known to have influenced the Nordic region.

Windscale[2]

Two air-cooled graphite moderated metal U reactors operated at Windscale from 1951 to 1957, when both were shut down due to the fire in Pile No1 (Salbu, 2001). Atmospheric particle releases were observed already in the mid 1950s. Due to low-temperature oxidation and corrosion of spent fuel elements misplaced in the air-cooled ducts leading to the discharge stack with inefficient filtering system, fuel particles containing actinides and fission products were continuously dispersed. During the years the total release is estimated to about 20 kg U as large particles, up to 700 μm in length (Jakeman, 1986). Based on electron microscopy, the particles had a flake-like structure, significantly different from those observed in the Chernobyl fallout (Salbu et al., 1994). The particles were inert towards leaching with 1 M HCl (Oughton et al., 1993).

Due to the graphite fire in 1957, about 10^{9} Bq of ^{239}Pu were released, probably associated with U oxide particles. These were observed as localised heterogeneities by autoradiography of samples from the stack filters, from contaminated surfaces from outside the reactor and from contaminated soils (Arnold, 1992; Chamberlain and Dunster, 1958). Particles in the size

[2] The description of the radioactive particle situation at this site has been taken from a recent PhD thesis (Lind, 2006).

range 20-500 μm were observed up to 4 km from the site, but were considered of little relevance to public health (Appleby and Luttrell, 1993).

Chernobyl[3]

The Chernobyl nuclear power plant accident in April 1986 is the most serious accidental release of radioactivity in the history of nuclear reactor operation. Severe violations of operating procedures in the course of an engineering test of the generator resulted in a prompt critical excursion of the 1 GW_e, graphite-moderated, light-water cooled reactor on April 26th, 1986.

The energy generated led to a series of explosions that destroyed the outer containment, exposing the core to the environment and injecting highly radioactive debris into the atmosphere. The releases continued in the following days due to the subsequent fire in the graphite moderator. However, the 'blanketing' of the core with boron, clay, lead, etc. led to increased temperatures and new releases from the 1st to the 5th of May. Emissions were terminated on May 6th upon cooling the reactor through tunnels constructed under the core (SCOPE, 1993).

As a result of the accident, about 6-8 tonnes of UO_2 fuel were released into the atmosphere (Victorova and Garger, 1990). Large fuel particles with variable radionuclide composition deposited within a 30 km zone with respect to the plant, while small-sized particles were identified up to 2000 km from the site (Devell et al., 1986; Kashparov et al., 1999; Kuriny et al., 1993; Salbu, 1988). During the initial explosion on April 26th, mechanical destruction of the UO_2 fuel occurred under high pressure and high temperatures (>2000ºC immediately prior to the explosion and reaching 2400-2600 ºC in local regions of the reactor), and deposition of fuel particles took place to the west of the reactor. In the period 26–30 April, volatile fission products and U fuel particles were released under moderate temperatures (330 - 930 ºC) and oxidising conditions due to the fire (Kashparov et al., 1999), and deposition of particles occurred to the north, northeast and south of the plant. From April 30th to May 6th the temperature was lowered and subsequently the emission of volatiles decreased (Kashparov et al., 1999). The particle weathering rate constant (k, $year^{-1}$) of U fuel particles estimated from a series of NH_4Ac extraction studies on Chernobyl soils collected over a 10 years period were found to range from 0.04 - 0.4y^{-1} (Kashparov et al., 1999). The results indicated that varying weathering rates could be related to different particle characteristics as well as varying soil pH. The weathering rate constants (k, $year^{-1}$) for U particles deposited to the west of the reactor were significantly lower than for particles deposited to the north and south of the reactor (Kashparov et al., 1999).

Dobrovolsky and Lyalko (1995) divided reported hot Chernobyl particles into 4 categories:

1. Fuel particles, i.e. more or less pure U oxides (UO_2 to U_3O_8/U_2O_5).
2. Fuel construction particles, i.e. particles with a matrix of nuclear fuel mixed with reactor construction material. A lot of (Zr, U)O_x particles have been found inside the

[3] The description of the radioactive particle situation following the Chernobyl accident has been taken from a recent PhD thesis (Lind, 2006).

30 km exclusion zone around the Chernobyl nuclear power plant, both to the west and to the north. Particles of U-Zr composition seem to be rather inert.

3. Construction particles are composed of reactor construction materials. They are carriers of some radionuclides.
4. Particles of hybrid type. Particles that were mainly derived from interaction of fuel and reactor construction material with fire distinguishing material etc.

This categorization seems valid for close-in fall-out around Chernobyl. However, a fifth category comprising single element (e.g. Ru, Cs) particles should be added. Such particles were found as far away as Norway, Sweden and Finland, 1500–2000 km from Chernobyl (Fig. 3-1).

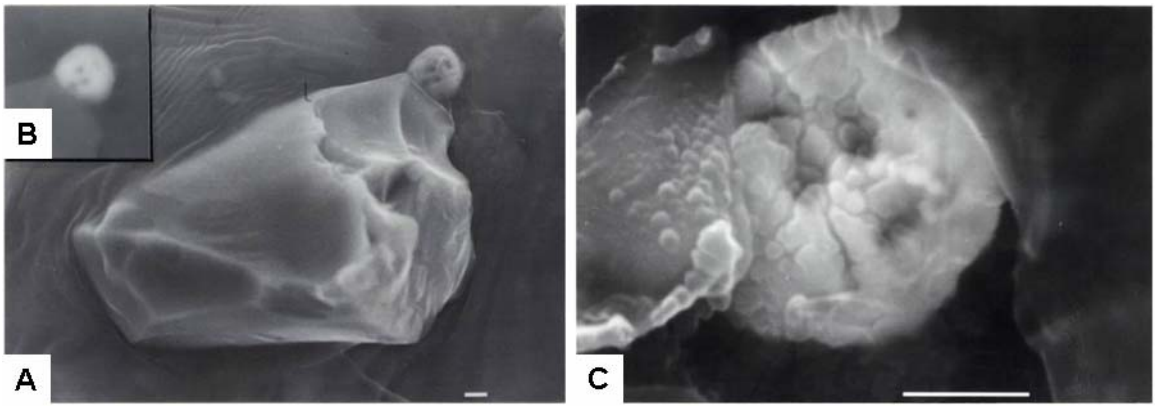

Fig. 3-1. Scanning electron micrograph of a Ruthenium particle found in 1987 in Valdres, Norway. A) Overview of the Ru particle attached to a larger silicon particle, SEI-mode; B) Close up of the Ru particle, BEI-mode. Bright areas show the presence of elements with high atomic numbers; C) Higher magnification of the hot particle, BEI-mode (Salbu, 1988).

The Sosnovyy Bor incident in 1992

One fuel channel of the RBMK reactor Unit 3 in Sosnovyy Bor, near St. Petersburg, Russia, broke down on 23-24 March 1992. Small amounts of noble gases, iodine and particulate radioactive materials were released and transported toward Finland (Toivonen et al., 1992). STUK - Radiation and Nuclear Safety Autority measured air concentrations along the coast of the Gulf of Finland, 12-20 h after the incidence occurred. Air samples collected in Kotka and Loviisa were analysed by autoradiography. About ten radioactive particles were identified in air filters and six of them were isolated and analysed using a gamma-ray spectrometer (Pöllänen, 1997). The Finnish Meteorological Institude detected also about ten radioactive particles in their air samples (Paatero and Hatakka, 1997).

Nuclear weapon accident at Thule

Since the beginning of the nuclear age there have been numerous accidents involving nuclear weapons. In January 1968 a B52 plane from the US strategic Air Command caught fire and crashed on the sea ice in Bylot Sound about 12 km west of the Thule Air Base, Northwest Greenland. The plane carried four nuclear weapons. Part of the weapons plutonium was distributed over some square kilometres of the ice in the explosive fire that followed. Plutonium-contaminated ice was recovered and shipped back to the US, as was the plutonium-contaminated wreck. The underlying sea sediments received a fraction of the weapons plutonium when the sea ice melted the following summer and probably also during the accident as the impact caused the ice to break up (Langham, 1970). A number of

expeditions (1968, 1970, 1974, 1979, 1984, 1991, 1997, 2003) has been conducted over the years in order to study the inventory and distribution of plutonium in the sediments as well as its uptake in biota. Terrestrial expeditions aimed to search for contaminated terrestrial sites have been conducted in 1968, 1997, 2003, 2006 and 2007.

It has been estimated that the pollution remaining in the seabed in Bylot Sound by 1968 amounted to approximately 1.4 TBq ^{239,240}Pu (about 0.5 kg), 0.025 TBq ^{238}Pu, 4.6 TBq ^{241}Pu and 0.07 TBq ^{241}Am. Results from a sample collection in 1997 indicate, however, that previous estimates of the plutonium inventory may not have included radioactive particles in the seabed sediments in sufficient detail (Eriksson, 2002) and that the inventory of $^{239+240}$Pu is somewhere between 1 and 10 TBq. The analyses of the sediment samples have shown that plutonium is very inhomogeneously distributed in the sediments due to the presence of hot particles. The systematic gamma screening of ^{241}Am in 5-15 g aliquots of the sediment samples have indicated a geometric mean ^{239,240}Pu activity of a few Bq with an approximate 95% confidence interval of from 0.1 to 50 Bq. The maximum ^{239,240}Pu activity determined in a hot particle in the sediments is 1500 Bq. The particles contain significant amounts of U in addition to Pu and these elements coexist throughout the particles (Eriksson et al., 2005), although varying Pu-L_α/U-L_α ratios obtained in environmental scanning electron microscopy with x-ray microanalysis (ESEM-XRMA) line scans reflected heterogeneities on the particle surfaces (Lind et al., 2007). Furthermore, enriched U constitutes the major part of the fissile material as determined for single particles (Lind et al., 2007). Synchrotron radiation based micrometer-scale X-ray Absorption Near Edge Structure Spectroscopy (μ-XANES) showed that U and Pu in the particles were present as a mixture of oxides of U (predominately UO_2 with the presence of U_3O_8) and Pu ((III)/(IV), (IV)/(V) or (III), (IV) and (V)). According to Lind et al. (2007), neither U metal, Pu metal, uranyl nor Pu(VI) could be observed.

At present, focus is directed towards the contaminated land areas south of the crash-site and an investigation concerning magnitude and spatial distribution in the area is ongoing.

4 Technologically Enhanced Naturally Occurring Radioactive Material (TENORM)

Radioactive particles are not exclusively of anthropogenic origin. For example, small grains of Th-bearing and U-bearing minerals, which occur naturally in soil and sediments, may also be considered as radioactive particles (Entwistle et al., 2003). Moreover, Th, U and daughter nuclides such as ^{210}Po/^{210}Pb (Landa et al., 1994) may be heterogeneously distributed in minerals, thus occurring as hot spots in U mining and tailing sites. Recently, single grains of U minerals have been isolated from soil samples originating from former U mining sites in Kazakhstan and Kyrgyzstan (Lind et al., 2008). Apparently, no reports on radioative TENORM particles are to be found in the open literature, However, in ESEM-XRMA, heterogeneities (i.e. U hot spots) have been observed on surfaces of Norwegian mineral specimens (Salbu et al., unpublished).

5 Reprocessing and Nuclear waste sites

Siberian Nuclear sites

Nuclear weapon materials were produced at three main sites in the former Soviet Union (FSU): Mayak PA (Chelyabinsk-45 or 65), Krasnoyarsk Mining and Chemical Industrial Complex (Krasnoyarsk-26), and Siberian Chemical Combine (Tomsk-7). The following descriptions of the radioactive particle situation at these site have been taken from a recent

PhD thesis (Lind, 2006). Relatively large routine releases occurred during the early years of operation of these facilities (UNSCEAR, 1993). In addition, accidents have contributed to radioactive particle contamination in the vicinity of the sites. Although these sites are located several thousand km from the Kara Sea, remobilisation of radionuclides from radioactive particles will occur over time and subsequent river transport of e.g. Pu into the Arctic Ocean needs to be included in impact assessments.

.Mayak PA (Chelyabinsk-65)

Apparently fashioned after the US Hanford site (Cochran et al., 1993), the Mayak Production Association (PA) was the first Pu production complex in the FSU (JRNEG, 1997) and is situated in the southern Urals within the catchment of the Techa river, a tributary to the Iset-Tobol-Irtysh-Ob river system, about 2800 km from the Kara Sea. Since 1948/49, totally 7 nuclear reactors and 2 reprocessing plants have been in operation at the site. The radionuclide contamination is mainly due to discharges from reactors and reprocessing plants to the Techa River, during 1949-1956, to Lake Karachay from 1951 and to the reservoirs from 1956 until present. Since 1987, civil nuclear waste has been reprocessed.

About 100 PBq of liquid waste including alpha emitters was released directly to the River Techa during 1949-1951 (Vorobiova et al., 1999). A fraction of the radionuclides released to the Techa River was soluble and another fraction was associated with particles (Degteva et al., 2000). Since 1951, about 20 EBq of liquid waste has been discharged into Lake Karachay, a small lake without outlet (JRNEG, 1997). Additional minor sources are fallout from the waste tank explosion in 1957 contaminating a terrestrial area up to 300 km from the site, wind dispersion of Lake Karachay sediments in 1967 and airborne releases from the plant stacks since installation. Based on recent investigations, the Pu inventory in the reservoirs is at least 40 TBq (JRNEG, 1997). Pu isotope ratios in sediments reflected a change in the discharge composition with time. In the deep sediment profiles Pu originating from the early discharges was present, while in the upper sediment profiles Pu was derived from recent reprocessing of civil nuclear fuel (Oughton et al., 2000).

Hot spots have been identified by digital phosphor imaging (autoradiography), γ-spectrometry and SEM analysis have been performed on samples from Reservoir 10 and 11 as well as from the Asanov swamp.

Siberian Chemical Combine (Tomsk-7)

The Siberian Chemical Combine (SCC), formerly known as Tomsk-7 is located 20 km from Tomsk city on the Romashka River, a tributary to the Tom-Ob river system, and 2700 km from the Kara Sea outlet (AMAP, 2004). The SCC has been in operation since 1952 (Alexakhin et al., 2004) and is the largest complex for the production of weapon grade material in the Russian Federation (UNSCEAR, 2000). At SCC, the production of Pu, U and transuranic elements has generated large amounts of radioactive waste that is stored in underground disposal sites and in storage ponds being severely contaminated (AMAP, 2004). Since commissioning of the first reactor in 1955 the plant has released contaminated cooling water from a total of 5 U-graphite reactors (one with a single pass core coolant system) into the Romashka-Tom-Ob river system (Waters et al., 1999). No information on the physico-chemical forms of effluent discharges appears to be available in the open literature.

An accidental explosion occurred in the radiochemical plant at the site on April 6th, 1993 (AMAP, 2004), due to improper procedures applied for mixing solutions prior to U extraction. Prior to the accident, the apparatus contained 25 m^3 of solution containing 8773 kg U, 310 g Pu and 248 g Np. Approximately 37 TBq of radionuclides were released by the explosion, of which about 19–22 TBq were released outside the facility. As a result of the accident, the snow-covered region close to the facility was contaminated and a radioactive trace including radioactive particles spread 20 km in a North-Eastern direction. Total γ-activity of some of the hottest particles reached 12 kBq (Tcherkezian et al., 1995).

Krasnoyarsk Mining and Chemical Industrial Complex (Krasnoyarsk-26)

The Krasnoyarsk Mining and Chemical Industrial Complex (KMCIC), formerly known as Krasnoyarsk-26 and now renamed Zheleznogorsk was established in 1958 and is located on the Yenisey river 9 km from the city of Zheleznogorsk and 2400 km from the Kara Sea outlet (AMAP, 2004). The primary activities of the KMCIC have been Pu production based on three RBMK-type graphite-moderated reactors, Pu reprocessing, and storage of radioactive wastes. Commissioned in 1958 and 1961, respectively, the first two reactors used open-loop core cooling. Coolant entered into the reactors from the Yenisey River and was discharged into the river. Therefore, cooling-water activation products, corrosion products from the fuel cladding and other parts of the reactor as well as fission products from "tramp" U and leakage from faulty fuel rods entered the river with the cooling water (Waters et al., 1999). These past releases have resulted in radioactive contamination of river water and sediments downstream of the complex. The two oldest reactors were shut down in 1992. The third reactor, which is still in use, has a closed primary cooling cycle (Linnik et al., 2005). However, the control rods are cooled in a once-through coolant loop and thus, together with migration of radionuclides from a liquid radioactive waste disposal site (Severny test site), represent sources of continuing discharge of radioactivity to the Yenisey (Bolsunovsky, 2004; Bolsunovsky and Bondareva, 2005; Bolsunovsky et al., 2002). About 20 and 30 years ago, respectively, two reactor accidents occurred, causing U fuel particles to be released into the Yenisey along with the cooling water (Bolsunovsky and Tcherkezian, 2001). According to Sukhorukov et al. (2004), a third event occurred in 1966 when nuclear waste storage ponds were affected by a large flooding event in the river Yenisey. No information appears to be available either on the magnitude of released radionuclides or on the environmental impact caused by the accident.

Niteelva

No reports on the existence of radioactive particles in Niteelva sediments (repository for historical releases from Institue of Energy Technique, Kjeller, Norway) appear to have been published in the open literature and preliminary results from screening of sediment samples performed at IFE have not indicated any heterogeneities.

Andreyeva Bay

No reports on the existence of radioactive particles at the spent fuel site at Andreyeva Bay appear to have been published in the open literature.

Dumping of nuclear waste in the Russian Arctic: Kola Bay (STUK)

The naval shipyards and military nuclear bases located in Severomorsk and Polarnyy have facilities for nuclear fuel storage and radioactive waste (Ikaheimonen et al., 1997; Rissanen et al., 1997; Rissanen et al., 1998). Storage vessels are also located around the city of Murmansk. The Atomflot civilian nuclear icebreaker base has a liquid-waste purification plant.

Samples of sediment, algae and lichen, as well as benthic samples were collected during the expedition in the Kola Bay, NW Russia, in 1995. Laboratory analyses revealed that the radioactive materials were not homogeneously distributed in some of the sediment and lichen samples. Two types of radioactive particle were identified in marine sediment and lichen samples (Pöllänen et al., 2001). The particles were identified by means of gamma-ray spectrometry and autoradiography, separated and subjected to various analysis techniques. Several complementary techniques were used to characterise particle properties thoroughly. ^{137}Cs was present in the sediment matrix in large (~ 100 μm) greenish particles that were most probably pieces of paint. Although their element composition was heterogeneous, ^{137}Cs was found to be evenly distributed. ^{60}Co in the lichen matrix was present in small (~ 1 μm) particles. Neither U nor transuranium elements were detected in either type of particle.

Dumping of nuclear waste in the Russian Arctic: Novaya Zemlya and the Kara Sea
During 1959-1991, radioactive waste including 6 reactors with fuel, 11 reactors without fuel, vessels, barges and more than 6 000 containers were dumped in the Abrosimov, Stepovogo and Tsivolky bays and in the Kara Sea Trough (AMAP, 1997). Furthermore, the fuel assembly from the Lenin reactor, assumed dumped along the east coast of Novaya Zemlya, has not yet been localised. The re-estimated inventory is 37 PBq at the time of dumping, and about 4.7 PBq at present (AMAP, 1997). In the close vicinities of dumped objects, especially containers, enhanced levels of Pu-isotopes and fission products in sediments were observed. Based on autoradiography, heterogeneities in the sediment samples reflected the presence of particles, and strong oxidising solutions (H_2O_2 in HNO_3) were needed to leach the Pu-isotopes from the sediments. Crud particles containing ^{60}Co were also identified using SEM (Salbu et al., 1997).

6 Characterization of particles

Alpha, Beta and Gamma Spectrometry
Spectrometry of the radiation from the particle gives information of its radionuclide composition. Direct spectrometry without any sample preparation is possible but radiochemical separation needs to be performed in order to get more detailed results. Methods have been developed in order to receive more information from the direct spectrometry, e.g., activity per isotope and density of the particles, which has the advantage that it avoids the destruction of the sample and data is provided faster (see e.g., (Pöllänen et al., 2007)). However, sometimes alpha particle energies are very close as for the cases of ^{239}Pu-^{240}Pu and ^{243}Cm-^{244}Cm. It is possible to quantify the isotopes by spectral deconvolution if activity concentrations are sufficiently high (Mitchell *et al.*1998.) The curium isotopes have too short half lives to be measured by mass spectrometric techniques. According to Eriksson (2005), X-ray spectrometry allows to determine the ^{241}Am and ^{238,239,240}Pu activities by analyzing the X-ray spectra collected during radioactive desintegration of the isotopes.

Scanning Electron Microscopy
SEM is a non-destructive, surface and sub-surface analysis technique providing information about the elemental composition, the morphology and the sizes of the specimen (Salbu, 1988). The SEM image has an excellent high resolution quality which is of good use when studying particles in the size range of < 1 μm to 300 μm (see Fig 3-1). However, the samples (i.e. the particles) must be conductive to avoid charging effects which lower the image quality although coating is usually avoided.

In scanning electron microscopes the area or micro-volume to be examined is irradiated with a finely focused electron beam (electrons are emitted from a filament and focussed through electrostatic lenses), which is swept in a raster across the surface of the beam. When the electron beam impinges on a specimen surface, elastic and inelastic electron-specimen interactions produce a variety of signals, including secondary electrons, backscattered electrons and x-rays. To measure the sample's elemental composition a detector is coupled to the system, either an EDX (Energy dispersive X-ray detector) or a WDX (Wavelength Dispersive X-ray detector) can be used. The EDX is much faster in performing the measurements but the WDX has a better "peak to background"-ratio, better counting efficiency and better resolution.

The microscope can be operated in two different modes, in secondary electron (SE) detection or in the back-scattered electron mode. The latter is a useful tool while searching for particles consisting of actinides because they appear brighter than other material with lower atomic number. This is due to that for material with higher atomic number more electrons are scattered compared to lighter elements, resulting in a stronger signal.

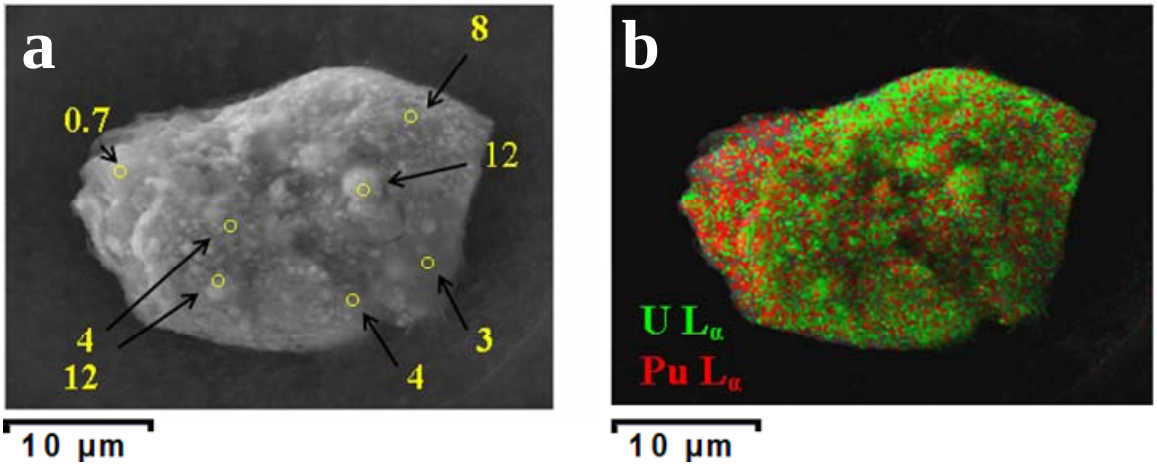

Figure 6-1. a: SEM image (SE mode) of a hot particle and U:Pu L_α intensity ratios for different EDX spot measurements. **b:** EDX mapping of the characteristic L_α intensity lines of U (green) and Pu (red) superimposed on the SEM image (Ranebo et al., 2007).

In addition to being a powerful tool for characterisation of particles on its own, SEM/ESEM is also utilised to localise the samples prior to further detailed studies using Synchrotron Radiation (SR) based X-ray microbeam techniques (Salbu et al., 1999). Images (both BEI and SEI) recorded at a wide range of magnifications are usually needed to facilitate the re-localisation of the microscopic samples on the carbon sticky tape during synchrotron experiments. The carbon sticky tape with the sample attached is fixed onto a slide frame with adhesive tape. Then, the samples are usually encapsulated (sandwiched) between plastic film foils (preferentially Mylar or Kapton) in such a way that contact with the particles is avoided.

X-ray micro techniques

Micro-analytical techniques refer to sensitive methods that provide information on the two-dimensional and/or three-dimensional distribution of elemental or molecular species within or at the surface of solid samples (Adams et al., 1998). They are usually based on microscopic beams of charged particles (e.g. electrons) or photons (e.g. x-rays) that are used to excite the sample locally. Typically, SEM/ESEM (electron beam) with XRMA (x-ray signals) and SR based micro x-ray techniques (x-ray beams and signals) can be used to study radioactive particle characteristics (Eriksson et al., 2005; Jernström et al., 2004; Lind, 2006; Lind et al.,

2007; Salbu et al., 2001). Both XRMA and SR based micro x-ray techniques are based upon measurement of x-ray signals arising from x-ray fluorescence and absorption.

The high intensity, tunability, high collimation, coherence and linear polarization of synchrotron radiation of the SR x-ray beams make them ideally suited for the generation of x-ray beams of microscopic dimensions (i.e. x-ray micro beams). The capabilities of microscopic XRF based on conventional x-ray sources in laboratories are much more limited in comparison (Adams et al., 1998). Scattering of the x-ray beam, which is the main culprit of background in x-ray fluorescence spectra, is dependent on polarization of the incident radiation whereas the photo absorption and hence the fluorescence production is not. Thus, the peak-to-background ratios of SR based x-ray fluorescence (SRXRF) analysis is superior to conventional XRF (Adams et al., 1998). Furthermore, the high coherence makes it possible to generate quasi-monochromatic x-ray microbeams from the white spectrum using x-ray monochromators. By tuning the energy of the source with a monochromator, it is possible to utilise the strong energy dependence of the photo absorption to increase the sensitivity of the elements of interest selectively or to obtain information on the speciation of major, minor and trace constituents, e.g. μ-x-ray absorption near edge spectroscopy (μ-XANES). Some micro beams can also be utilised to obtain diffraction patterns from micrometer spots on samples, i.e. micro x-ray diffraction (μ-XRD) (Catalano et al., 2004; Salbu et al., 2001; Salbu et al., 2004). Other micro analytical techniques that have proved or may prove useful for particle characterisation include μ-PIXE (Lopez et al., 2007), μ-RAMAN (Allen et al., 1987), TEM (transmission electron microscopy) with XRMA (Ormstad et al., 1997), electron diffraction (Wolf et al., 1997) or Electron Energy Loss Spectroscopy (EELS) (Buck et al., 2004).

Mass spectrometry

Mass spectrometry is a well-known analytical technique for the isotopic and elemental composition. In principle it is based on three parts: An ion source, where ions from the sample are produced, a mass analyzer, where the ions are separated according to their mass-to-charge ratio, and finally, a detector system for the detection of the ions. There exist a wide range of different mass spectrometers. The main difference between different mass spectrometers is their ion source, the principle of how to produce ions from the sample.

Inductively Coupled Plasma - Mass Spectrometry (ICP-MS)

ICP-MS is a highly sensitive technique for quantitative elemental and isotopic determination of a specimen. The ions are produced in a high-temperature plasma where the sample is decomposed and then analyzed regarding their mass-to-charge ratio. A radiochemical separation of the sample is required prior the analyses in order to remove isobaric interferences.

Secondary Ion Mass Spectrometry (SIMS)

Secondary ion mass spectrometry (SIMS) is one of the most surface sensitive techniques giving isotopic information down to the level of atomic layers. However, it is more difficult to obtain quantitative results compared to other techniques because of different ionization behaviour between elements. An outstanding advantage with SIMS for studies of hot particles containing U is that it removes the problem of the need of ultra-clean sample preparations (radiochemistry, laboratory facilities and analytical instruments) for avoiding cross-contaminations with natural uranium.

The principle of the SIMS is based on an ion source producing ions which are accelerated towards the sample and secondary ions are sputtered from the sample. The sputtering is similar to a knock-out process where the ions break the sample ions bounding to the atom lattice. Secondary ions are produced representing the elemental and spatial composition at the

surface and thereby the name of the instrument. The secondary ions are further accelerated into the secondary part, the mass analyzer, where they first are filtered according to their energy in the electrostatic analyser and then according to their mass in the magnetic sector. The SIMS measurements provide the possibility to make isotopic images, depth profile measurements, mass spectra analyses, isotopic ratio measurements and line scans of the sample.

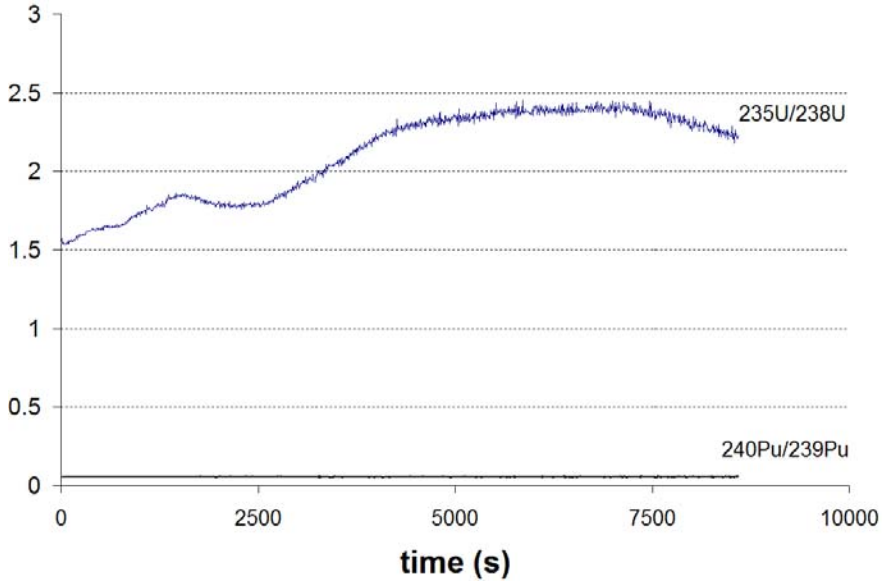

Fig. 6-2. An example of a SIMS ^{235}U:^{238}U and ^{240}Pu:^{239}Pu intensity profiles as a function of measurement time (correlated to the particle depth) showing an increasing U ratio and a constant Pu ratio inside the particle.

Accelerator Mass Spectrometry (AMS)[4]

Accelerator Mass Spectrometry (AMS) offers quantitative measurement of low levels of long-lived U and Pu isotopes with detection limits and freedom of isobaric mass interferences that are superior to α-spectrometry and ICP-MS (Fifield et al., 1996). The sensitivity of the AMS technique for determination of Pu (10^6 atoms) is at least two orders of magnitude higher than for α-spectrometry and one order of magnitude higher than what can be achieved with conventional ICP-MS (Skipperud, 2005). Precision of AMS, however, is not high for heavy nuclides compared to most mass spectrometric techniques.

The AMS technique requires the preparation of homogeneous solid metallic or metal oxide samples containing the radionuclide of interest together with a suitable isotopic tracer. The ion source is produced by sputtering the solid sample with Cs^+ ions and injecting the negative ions into the accelerator (Fig. 6-).

[4] This description of the AMS method has been taken from a recent PhD thesis (Lind, 2006).

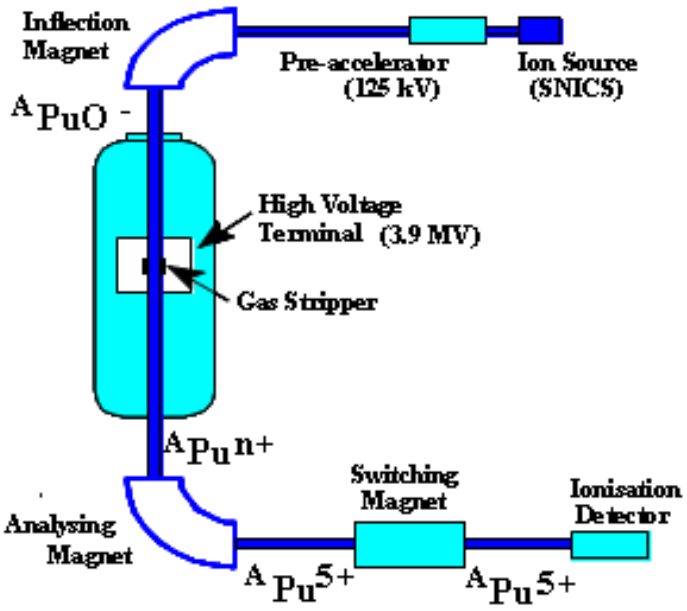

Fig. 6-3. Schematic presentation of the 14 UD tandem AMS at ANU, Canberra, Australia.

The beam is then accelerated towards a high-voltage positive potential, stripped of outer electrons and the positively charged ions formed (up to 13+) are accelerated. Then, the isotopes are separated according to their mass/charge ratios in a high magnetic field before counting. Methods for measuring heavy radionuclides such as U, Pu and Np have been developed and applied successfully to measurement of activity concentrations and isotope ratios in low-level environmental samples (Fifield et al., 1996; Fifield et al., 1997; Marsden et al., 2001).

7 Production of artificial particles

Standard particles would allow an optimization of the accuracy and precision of analytical techniques used for characterization of hot particles. At Institute for Transuranium Elements (ITU), Karlsruhe, mono-dispersed micro-meter sized uranium particles have been produced from reference material (Erdmann et al., 2000). As many hot particles also contain plutonium, a new project was started in 2004 to develop a method for the production of standard particles containing plutonium and uranium/plutonium mixtures. The already existing set-up were to be used, a vibrating orifice aerosol generator system (Model 3450,TSI Inc.) with an additional furnace system for calcinations of the produced uranyl nitrate aerosol particles. However, modified and implemented into a glove-box for the new purpose. Both pure Pu particles have been produced, as well as, different mixtures with U/Pu.

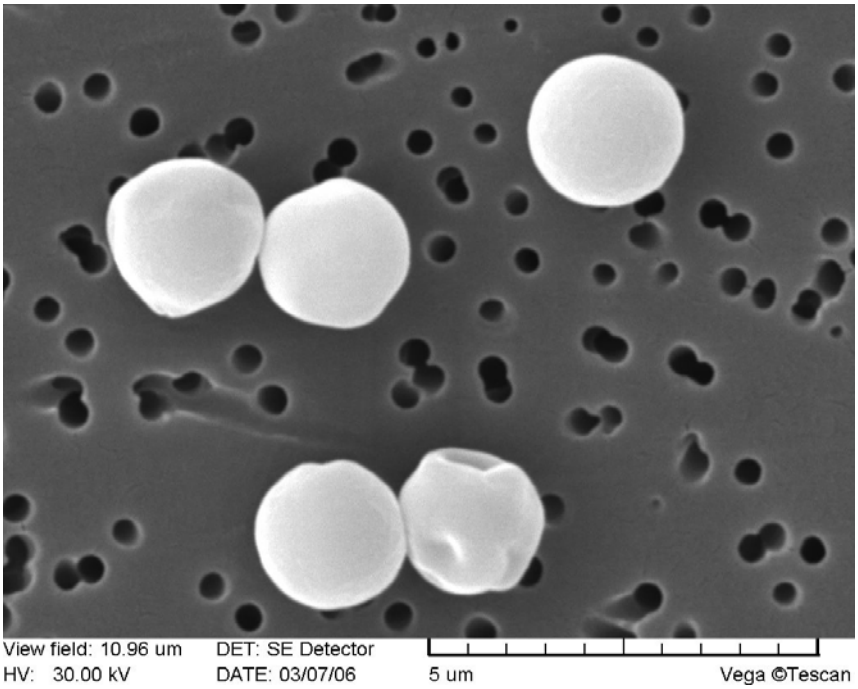

Fig. 7-1. SEM image of mono dispersed uranium particles.

8 Conclusions

The present overview report show that there are many existing and potential sources of radioactive particle contamination of relevance to the Nordic countries. Following their release, radioactive particles represent point sources of short- and long-term radioecological significance, and the failure to recognise their presence may lead to significant errors in the short- and long-term impact assessments related to radioactive contamination at a particular site. Thus, there is a need of knowledge with respect to the probability, quantity and expected impact of radioactive particle formation and release in case of specified potential nuclear events (e.g. reactor accident or nuclear terrorism). Furthermore, knowledge with respect to the particle characteristics influencing transport, ecosystem transfer and biological effects is important. In this respect, it should be noted that an IAEA coordinated research project was running from 2000-2006 (IAEA CRP, 2001) focussing on characterisation and environmental impact of radioactive particles, while a new IAEA CRP focussing on the biological effects of radioactive particles will be launched in 2008.

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Bibliographic Data Sheet **NKS-180**

Title	Overview of sources of radioactive particles of Nordic relevance as well as a short description of available particle characterisation techniques
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Abstract	The present overview report show that there are many existing and potential sources of radioactive particle contamination of relevance to the Nordic countries. Following their release, radioactive particles represent point sources of short- and long-term radioecological significance, and the failure to recognise their presence may lead to significant errors in the short- and long-term impact assessments related to radioactive contamination at a particular site. Thus, there is a need of knowledge with respect to the probability, quantity and expected impact of radioactive particle formation and release in case of specified potential nuclear events (e.g. reactor accident or nuclear terrorism). Furthermore, knowledge with respect to the particle characteristics influencing transport, ecosystem transfer and biological effects is important. In this respect, it should be noted that an IAEA coordinated research project was running from 2000-2006 (IAEA CRP, 2001) focussing on characterisation and environmental impact of radioactive particles, while a new IAEA CRP focussing on the biological effects of radioactive particles will be launched in 2008.
Key words	radioactive particles, sources, characteristics

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