

# Source identification of Pu and $^{236}\text{U}$ deposited on Norwegian territories

Identifisering av kilder til radioaktiv forurensing på norsk  
territorium

Philosophiae Doctor (PhD) Thesis

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## **Preface**

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## Abstract

Plutonium (Pu) is a predominately anthropogenic element produced during neutron irradiation of U in reactors and nuclear weapon detonations. Pu has been released to the environment during nuclear weapon detonations, nuclear reactor accidents, and in association with reprocessing of spent nuclear fuel. The most important source of Pu in the environment were the 543 atmospheric nuclear detonations conducted worldwide in the period 1945 – 1980 by the former Soviet Union (FSU), USA, United Kingdom (UK), China and France. The most intensive period of atmospheric nuclear testing was 1945 – 1962, interrupted by a moratorium during November 1958 to September 1961. This period was dominated by US testing during the pre-moratorium period and FSU testing during the post-moratorium period. Debris from the atmospheric nuclear detonations was deposited on a local, regional and global scale depending on detonation height, yield and meteorological conditions.

The isotopic composition of Pu is source dependent. Pu is produced by neutron capture in  $^{238}\text{U}$ , generating  $^{239}\text{U}$  which rapidly ( $t_{1/2}=23.5$  min) decay to  $^{239}\text{Pu}$ . Heavier isotopes are produced due to neutron capture in this  $^{239}\text{Pu}$ . Heavier isotopes of Pu are successively produced during continued neutron irradiation. Reactor derived Pu would have  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios reflecting the irradiation history of the fuel (burn up). For technical reasons, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in weapon purpose Pu has to be below 0.07. The burn up in weapon production reactors kept low by frequent exchange of the fuel in order to ensure this. For power production reactors on the other hand, frequent exchange of the fuel is impracticable and uneconomical. The fuel is exchanged after a prolonged irradiation and high burn up. As the proportion of heavier Pu isotopes increase in relation to  $^{239}\text{Pu}$ , all Pu atom ratios, and in particular the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios increase;  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios up to 0.67 have been reported. Debris from undetonated weapons and low yield detonations would also have low  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios, similar to the original weapon material. Post detonation Pu atom ratios depend on the yield and design of the device with high yield detonations generally producing Pu with higher  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios than low yield detonations. Global fallout was the mixed debris injected into the stratosphere by high yield detonations. After a considerable retention time in the stratosphere, this debris was deposited to form a general Pu background with an easily recognisable  $^{240}\text{Pu}/^{239}\text{Pu}$  signature of  $\sim 0.18$ . Pu isotopic signatures substantially different from global fallout indicates significant influence from local or regionally deposited Pu from nuclear weapon test sites, reactor accidents or releases from reprocessing.

In the present thesis U and Pu isotopic signatures has been utilised for source identification and quantification purposes. The samples employed have been humic surface soils from 45 geographically well distributed sites in Norway (*Paper I*); an ice core from Austfonna (Svalbard) covering the period 1949 – 1999 (*Paper II*); and air filter samples from Norway during 1957 - 1963(*Paper III*). Radioactive particles have been identified in the air filter samples the ice core samples (*Paper II*) and (*Paper III*), while atmospheric dispersion modelling (NOAA HYSPLIT\_4) has been used to corroborate the source identification in (*Paper III*).

Debris from the Chernobyl accident in 1986 was distributed over vast areas of Europe, including Fenno-Scandinavia. Whereas the deposition of caesium from the Chernobyl accident is known to have been strong in Norway, knowledge of the Pu deposition is scarce. In order to assess the relative importance of sources to Pu deposition in mainland Norway, Pu concentrations and atom ratios in humic surface soil samples collected from 45 geographically well distributed sites were determined by SF-ICP-MS (sector field inductively coupled plasma – mass spectrometry) and AMS (Accelerator mass spectrometry). Pu concentrations in samples collected from the same sites in 1990 and 2005 were compared in order to assess the relative migration of Pu from the surface layers. Pu concentrations ( $0.7 - 149 \text{ Bq m}^{-2}$ ) were found to be higher at coastal sites with high annual precipitation than at inland sites with lower annual precipitation, this is in accordance with the global fallout theory. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios were found to vary within 0.161 – 0.211, mainly in accordance with the global fallout signature. However, at some inland sites Pu atom ratios were found to exceed global fallout Pu signatures, indicating a slight Chernobyl influence.

Time resolved samples permit the investigation of variations of deposition and isotopic signatures in deposited debris. Pu and  $^{236}\text{U}$  concentrations and atom ratios were determined by AMS in an Arctic ice core from Austfonna (Svalbard) covering the period 1951 – 1999, and in air filter samples collected during 1957 – 1963.

The Austfonna represents an interesting sampling location in its proximity to FSU test site Novaya Zemlya (NZ). In addition tropospheric transport from Eurasia, and possibly the FSU test site in Semipalatinsk (STS) is facilitated by a semi-permanent high pressure area above Siberia during winter. Digital autoradiography indicated the presence of radioactive particles at depths corresponding to atmospheric nuclear testing (1949 – 1962) and the Chernobyl accident or a vented NZ underground detonation (1985 – 1989). Concentrations of Pu and  $^{236}\text{U}$  in the Austfonna ice core ( $0.008$  to  $0.254 \text{ mBq cm}^{-2}$  and  $0.0039$  to  $0.053 \text{ } \mu\text{Bq cm}^{-2}$  respectively) were found to be higher at depths corresponding to the pre-moratorium period (1956 – 1959) than at

depths corresponding to the post- moratorium period (1959 – 1962). This observation contrasts with observations at other sites where the deposition of anthropogenic radionuclides was found to be higher in the post-moratorium period. The discordance either indicates post depositional redistribution or a different depositional regime at the Austfonna glacier than at other sites.

The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio measured in air filters (0.0517 – 0.237) showed a strong dependence on northern hemisphere atmospheric test activities, tending towards a value reflecting stratospheric fallout during periods without atmospheric nuclear testing, and substantially lower ratios in periods associated with atmospheric nuclear testing activities. The lower Pu atom ratios observed during periods of atmospheric nuclear testing indicates significant influence of tropospherically transported Pu from FSU atmospheric nuclear testing. A further indication is provided by the presence of radioactive particles in filters from the autumn and winter seasons of 1961 and 1962, i.e. periods associated with atmospheric testing at FSU test sites NZ and STS. Direct tropospheric transport from FSU test site STS was indicated by high Pu and  $^{236}\text{U}$  concentrations and low  $^{240}\text{Pu}/^{239}\text{Pu}$ ,  $^{241}\text{Pu}/^{239}\text{Pu}$  and  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios during November 1962. Atmospheric dispersion modelling (HYSPLIT) using real time meteorological data confirmed the plausibility of this transport, and limited the potential source to three low yield STS test detonations during 30 October – 1 November 1962.

## Sammendrag

Plutonium (Pu) er et hovedsakelig menneskeskapt grunnstoff, dannet ved nøytronbestråling av uran (U) i reaktorer og atomvåpendetonasjoner. Plutonium har blitt sluppet ut i naturmiljøet som en følge av atomprøvesprengninger, reaktorulykker, og i forbindelse med gjenvinning av brukt reaktorbrensel. Den viktigste kilden til Pu i miljøet var 543 atmosfæriske atomvåpensprengninger, foretatt på begge halvkuler i perioden 1945 - 1990. Atmosfæriske atomprøvesprengninger ble foretatt av den tidligere Sovjetunionen (FSU), USA, Storbritannia (UK), Kina og Frankrike. Den mest intense prøvesprengningsperioden var 1945 – 1962, avbrutt av et moratorium fra november 1958 til september 1961. Denne perioden var dominert av detonasjoner foretatt av USA før moratoriet og FSU i perioden etter moratoriet. Nedfall fra de atmosfæriske prøvesprengningene ble deponert på et lokalt, regionalt og globalt plan hovedsakelig avhengig av prøvesprengningens sprengkraft, detonasjonshøyde samt meteorologiske forhold.

Isotopsammensetningen av Pu er kildeavhengig. I reaktorer blir Pu produsert ved nøytroninnfangning hovedsakelig i  $^{238}\text{U}$ , som gir dannelse av  $^{239}\text{U}$  som raskt ( $t_{1/2}=23,5$  min) henfaller til  $^{239}\text{Pu}$ . Tyngre Pu isotoper dannes ved fortsatt nøytronbestråling av brenselet. Pu i reaktorer vil ha et  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforhold som avhenger av brenselets bestrålingshistorie (burn up). Våpenteknisk Pu vil ha en høy andel  $^{239}\text{Pu}$  og et lavt  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforhold. Typiske  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforhold i våpenmateriale er under 0,07,  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforholdet i reaktorer tiltenkt våpenproduksjon er tilsvarende lavt. Pu med lavt  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforhold produseres ved å la reaktoren være i drift i en kort periode før brenselet byttes (lav burn up). Dette er imidlertid en lite økonomisk måte å drive energiproduserende atomreaktorer på, her tilstrebes god utnyttelse av brenselet og få avbrudd i driften. Ved forlenget drift øker dannelse av tyngre Pu-isotoper på bekostning av  $^{239}\text{Pu}$ ; derfor øker alle Pu atomforhold i reaktoren, og spesielt  $^{240}\text{Pu}/^{239}\text{Pu}$ . Det har i litteraturen vært referert til atomforhold opp til 0,67 for energiproduserende reaktorer. Pu atomforhold i materiale fra udetonerte atomvåpen og fra detonasjoner med lav sprengkraft vil også være lavt, ofte tilsvarende det opprinnelige våpenmaterialet. Pu atomforhold i nedfall fra et detonert atomvåpen avhenger av bombens sprengkraft og design, hvor detonasjoner med høy sprengkraft som oftest produserer nedfall med høyere  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforhold enn detonasjoner med lav sprengkraft. Globalt nedfall (GFO) ble dannet av de detonasjonene som hadde tilstrekkelig kraft til å injisere store deler materiale i stratosfæren. Dette nedfallet hadde en betydelig oppholdstid i stratosfæren for det ble

deponert på bakken. Globalt nedfall er relativt jevnt fordelt, og har dannet et gjenkjennelig Pu bakgrunnssignal med et  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforhold på omtrent 0,18 på begge halvkluler. Pu med atomforhold vesentlig forskjellig fra GFO antyder signifikant lokalt eller regionalt nedfall fra atomprøvesprengninger, materiale fra reaktorulykker eller utslipp forbundet med gjenvinning av brukt reaktorbrensel.

Dette arbeidet har omfattet kildeidentifisering og kildekvantifisering basert på Pu og U atom og aktivitetsforhold. Prøver av overflatejord fra 45 steder i Norge (artikkel I); en iskerne fra Austfonna, Svalbard som dekker perioden 1949 – 1999 (artikkel II); samt luftfiltre samlet inn i Norge i perioden 1957 – 1963 (artikkel III). Radioaktive partikler har blitt gjenfunnet i iskerneprøvene og luftfilterprøvene (artikkel II og III), og en atmosfærisk spredningsmodell (NOAA HYSPLIT\_4) har vært brukt for å forsterke kildeidentifisering i artikkel III.

Tsjernobyl-ulykken i 1986 forårsaket spredning av radioaktivt materiale over store deler av Europa, inkludert Fennoskandia. Det er kjent at deponeringen av cesium fra Tsjernobyl-ulykken var kraftig i Norge, imidlertid er deponeringen av Pu dårlig beskrevet. For å anslå påvirkningen av Pu fra Tsjernobyl-ulykken ble konsentrasjoner og atomforhold av Pu bestemt ved bruk av SF-ICP-MS (sector field-inductively coupled plasma-mass spectrometry) og AMS (accelerator mass spectrometry) i prøver av overflatejord samlet inn fra til sammen 45 steder i 1990 og 2005. Prøvestedene var geografisk jevnt fordelt i hele Norge, og det ble tilstrebet å samle inn prøver fra de samme stedene i 1990 og 2005. Videre ble Pu konsentrasjonene påvist i prøvene fra 1990 og 2005 sammenliknet for å anslå den relative retensjonen i overflatejorden. Pu-konsentrasjonene i begge datasettene ble funnet å variere mellom 0,7 og 149 mBq m<sup>-2</sup>. Konsentrasjonene av Pu i jordprøvene ble funnet å være høyere ved kysten, i samsvar med høyere årlig nedbør, tilsvarende ble lavere konsentrasjoner funnet ved innlandslokasjoner. Dette er i samsvar med generelle teorier om at globalt nedfall i stor grad følger nedbørmønsteret. Pu-240/Pu-239 atomforholdene ble funnet å ligge mellom 0,161 og 0,211, hovedsakelig i samsvar med globalt nedfall. Noen av prøvestedene i innlandet ble funnet å inneholde Pu med atomforhold vesentlig høyere enn globalt nedfall, noe som indikerte en viss påvirkning av Pu fra Tsjernobyl-ulykken.

Variasjoner og trender i deponeringen kan undersøkes ved hjelp av prøvetyper med tidsoppløsning. Pu og U atomforhold ble bestemt ved hjelp av AMS i en iskerne fra Austfonna (Svalbard) og i luftfilterprøver fra årene før og etter moratoriet.

Austfonna er et spesielt interessant prøvetaksingssted grunnet nærheten til de tidligere Sovjetiske prøvesprengningsfeltene ved Novaja Semlja (NZ). I tillegg fører et semi-

permanent høytrykksområde over sentrale Sibir vinterstid til økt transport av troposfærisk luft fra Eurasia mot Arktis. Dette vil potensielt kunne øke transport av materiale fra atomprøvesprengninger over prøvesprengningsfeltet Semipalatinsk (STS) til Arktis. Digital autoradiografi indikerte radioaktive partikler på dybder som svarer til periodene med atomprøvesprengninger ved STS og NZ (1949 – 1962) samt på dybder som svarer til 1985 – 1989 (Tsjernobyl-ulykken og en underjordisk prøvesprengning med lekkasje av radioaktive gasser til atmosfæren ved NZ). Pu og  $^{236}\text{U}$  konsentrasjonene (henholdsvis 0,008 til 0,254 mBq cm<sup>-2</sup> og 0,0039 til 0,053 μBq cm<sup>-2</sup>) ble funnet å være høyere i perioden før moratoriet (1956 - 1959) enn i dybder svarende til perioden etter moratoriet (1959 – 1962). Dette er i motsetning til observasjoner fra andre prøvetakingssteder, hvor deponeringen av radionuklider har vært høyere i perioden etter moratoriet. Forskjellen mellom resultatene oppnådd for iskjernen fra Austfonna og andre arbeider antyder enten at prosesser i isbreen etter deponering har forstyrret kronologien, eller at deponeringen av radioaktivt nedfall har vært annerledes ved Austfonna.

I luftfiltrene fra perioden 1957 – 1963 ble det funnet en sterk sammenheng mellom  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforhold (0,0517 – 0,237) og perioder med atomprøvesprengninger på den nordlige halvkule. I de perioder uten atomprøvesprengninger på den nordlige halvkule antok  $^{240}\text{Pu}/^{239}\text{Pu}$  atomforholdet målt i filtrene en verdi som gjenspeilte globalt nedfall, mens i perioder hvor det ble foretatt atomprøvesprengninger ved FSU prøvesprengningssteder NZ og STS lå dette atomforholdet betydelig lavere. Dette antyder at materiale fra FSU prøvesprengninger har blitt transportert i betydelig grad til Norge via troposfæren. Denne indikasjonen ble forsterket av resultatene fra autoradiografi. Her ble det funnet betydelig flere radioaktive partikler i perioder med atomprøvesprengninger på den nordlige halvkule enn i perioder uten. En episode med direkte troposfærisk transport av materiale fra atomvåpensprengninger ved STS prøvesprengningsområde til Norge ble indikert av høye Pu og  $^{236}\text{U}$  konsentrasjoner, og svært lave  $^{240}\text{Pu}/^{239}\text{Pu}$ ,  $^{241}\text{Pu}/^{239}\text{Pu}$  and  $^{236}\text{U}/^{239}\text{Pu}$  atomforhold i november 1962. Beregninger foretatt på basis av historiske sanntids meteorologiske data med en atmosfærisk spredningsmodell (HYSPLIT) forsterket plausibiliteten for at det radioaktive materialet påvist i disse filtrene stammet fra prøvesprengninger over ved Semipalatinsk. Modellen, og værforholdene i Eurasia på dette tidspunktet avgrenset den sannsynlige kilden til tre mindre atomprøvesprengninger ved STS i perioden 30. oktober til 1. november 1962.

## List of papers

### Paper I

C.C. Wendel, L. Skipperud, O.C. Lind, E. Steinnes, S. Lierhagen, B. Salbu. Levels and trends of Pu deposited on humic surface soils. Submitted, Journal of environmental radioactivity.

### Paper II

C.C. Wendel, D.H. Oughton, O.C. Lind, L. Skipperud, L.K. Fifield, E. Isaksson, S.G. Tims, B. Salbu. Chronology of Pu isotopes and  $^{236}\text{U}$  in an arctic ice core. *Science of the total environment*. **461 – 462**, 734 - 741

### Paper III

Cato Christian Wendel, Deborah H. Oughton, Ole Christian Lind, Lindis Skipperud, L. Keith Fifield, Jerzy Bartnicki, Stephen G. Tims, Steinar Høibråten, Brit Salbu. Long-range tropospheric transport of uranium and plutonium weapons fallout from Semipalatinsk nuclear test site to Norway. *Environment international*. **59**, 92 - 102

## Abbreviations and definitions

AMS	Accelerator Mass Spectrometry
SF-ICP-MS	Sector field Inductively Coupled Plasma –Mass Spectrometry
SEM	Scanning electron microscope
ESEM-XRMA	Environmental Scanning Electron Microscope with X-ray Microanalysis
HYSPLIT	Hybrid single particle Lagrangian integrated trajectory
CCN	Cloud Condensation Nuclei
GFO	Global fallout
NAO	North Atlantic Oscillation
FSU	Former Soviet Union
NZ	Novaya Zemlya test sites
STS	Semipalatinsk test site
PPG	Pacific proving grounds
NTS	Nevada test site (continental USA)
LN	Lop Nor test site (China)
GCR	Magnox reactor
PHWR	Pressurised heavy water reactor
AGR	Advanced gas cooled reactor
RMBK	Pressure tube boiling water reactor
PWR	Pressurised water reactor
LWR	Light water reactor
RTG	Radioisotope thermoelectric generator
HEU	Highly enriched uranium, >90% <sup>235</sup> U
DU	Depleted uranium, < 0.72% <sup>235</sup> U
Fission detonations	The energy released during the detonation originates from fission of (usually) <sup>235</sup> U and / or <sup>239</sup> Pu. Yield <~ 50 kt

Boosted fission detonations	Fission weapons wherein a fusion reaction during detonation contributes with neutrons boosting the fission. Yield range 50 – 500 kt.
Thermonuclear detonations	Fission device providing sufficient energy and compression to yield fusion in one or more fusion devices arranged close to the fission device. Additional secondary fission devices triggered by escape neutrons from the fusion processes possible. Historical yield range 0.5 Mt – 58 Mt
kt	$10^6$ kg TNT equivalents, corresponds to $\sim 1 \times 10^{12}$ MJ
Mt	$10^9$ kg TNT equivalents, corresponds to $\sim 1 \times 10^{15}$ MJ
ng	Nanograms ( $10^{-9}$ g)
pg	Picograms ( $10^{-12}$ g)
fg	Femtograms ( $10^{-15}$ g)
$H_{\text{det}}$	Detonation height

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# 1 Introduction

Anthropogenic (man-made) radionuclides have been introduced to the environment in significant quantities since the start of the nuclear era. The most important sources were nuclear detonations, carried out by eight nations (USA, FSU, UK, China, France, India, Pakistan and North Korea). Altogether more than 2422 atmospheric, underground and underwater tests were conducted in the period 1945 – 2013, with a total yield equivalent to 530 Mt TNT (UNSCEAR, 2000a; Björklund and Goliath, 2009). A number of safety tests without fission yield have been included in the total number of atmospheric tests.

The largest proportion of the yield (440 Mt) is attributed to atmospheric tests carried out in the period 1945 – 1980, with the most intensive period 1951 – 1962 (409 Mt) (UNSCEAR, 2000a; Björklund and Goliath, 2009). Debris from these tests was distributed between the local (~15 %), tropospheric (~8.5 %) and stratospheric (~76%) compartments (UNSCEAR, 2000a). These releases were not only the largest releases, but due to their nature, also caused the most widespread contamination.

Releases of radionuclides from the nuclear fuel cycle (mining, milling, fuel fabrication, reactor operation and reprocessing) have also been a major source of radioactive contamination (Jones *et al.*, 1996; Beasley *et al.*, 1998a; Cooper *et al.*, 2000; Lind, 2006). A series of nuclear reactor accidents have occurred, however, most of them have only had a local impact. The consequences of the Chernobyl accident in 1986 and the Fukushima accident in 2011 have been more severe; these have caused widespread contamination (UNSCEAR, 2000b; Stohl *et al.*, 2012). Finally, discharges to the aquatic environment from reprocessing installations have been distributed regionally.

Releases of radionuclides have also occurred in connection with satellite accidents and during transport of nuclear weapons. Three satellite accidents with global and regional importance have taken place; the disintegration of a transit navigational satellite containing a SNAP-9A radioisotope thermoelectric generator (RTG) in the high stratosphere of the southern hemisphere; the Cosmos-954 satellite disintegrating above Canada in 1978; and the Cosmos-1402 satellite disintegrating over the south Atlantic ocean in 1983 (Krey, 1967; Bakhtiar *et al.*, 1985; Leifer *et al.*, 1987; UNSCEAR, 1993; Jia *et al.*, 1999).

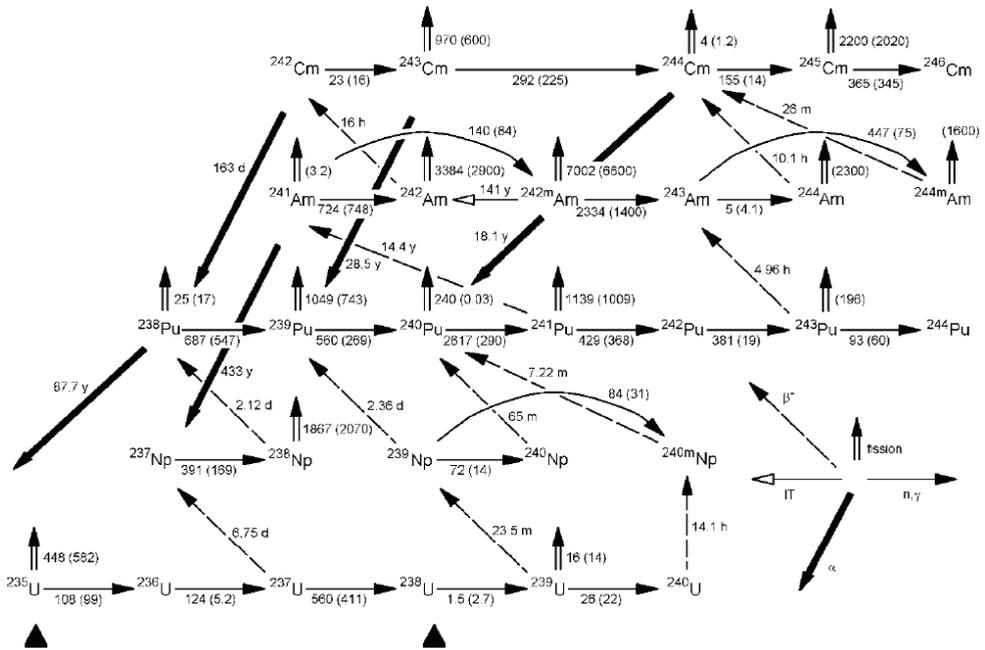
Local contamination has been caused by aircraft accidents involving nuclear weapons in Palomares, Spain and at Thule, Greenland in 1966 and 1968,

respectively, (e.g. Mitchell *et al.* (1997), Pöllänen *et al.* (2006), Lind *et al.* (2007)). Likewise, dumping of radioactive waste at sea has had local environmental influence due to a poor state of the containers and leakage (Salbu *et al.*, 1997).

All major nuclear sources of release such as nuclear weapon tests and nuclear reactor accidents have contributed to the presence of Pu and anthropogenic U in the environment. These radionuclides are of particular interest, as the isotopic signature can be used for source identification. U is a natural radionuclide with the isotopes  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  occurring with isotopic abundances of 0.005 %, 0.72 % and 99.27 %, respectively. Pu isotopes and  $^{236}\text{U}$  are, however, almost exclusively anthropogenic, and formed during reactor operation and nuclear weapon detonations.

Nuclear weapon material is composed of Pu (more than 93 %  $^{239}\text{Pu}$ ), enriched U (more than 90 %  $^{235}\text{U}$ ) or a combination of the two. Enriched U is normally produced from natural U through ultracentrifugation, while  $^{239}\text{Pu}$  is produced through neutron irradiation of  $^{238}\text{U}$  in reactors. The production of Pu with a high concentration of  $^{239}\text{Pu}$  is accomplished through short irradiation times within the reactor. In power production reactors, irradiation times are longer, and heavier Pu isotopes ( $^{240}\text{Pu}$  and heavier) are produced throughout fuel utilisation. Thus, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in particular, becomes successively higher the longer the fuel is irradiated.

Reactor irradiation of fuel is associated with low neutron fluxes and long irradiation times. In this process the short lived heavier U isotopes ( $^{239}\text{U}$ ,  $t_{1/2}=23.5$  min) transmute to Pu isotopes prior to further neutron capture. Therefore, in reactors  $^{239}\text{Pu}$  is formed, and heavier Pu isotopes originates from neutron activation in this  $^{239}\text{Pu}$ . Heavier isotopes up to  $^{243}\text{Pu}$  are formed as shown in Figure 1, however, due to the short half live of  $^{243}\text{Pu}$  (4.96 h), this isotope decays to  $^{243}\text{Am}$  prior to further neutron capture, and the production of  $^{244}\text{Pu}$  is insignificant in reactors in comparison with thermonuclear devices (Winkler, 2007).



**Figure 1. Neutron irradiation and decay during reactor operation of a typical power production LW reactor. Numbers along axis refer to half-lives or effective cross sections. From (Choppin *et al.*, 2002).**

In the high neutron fluxes and short irradiation times associated with thermonuclear detonations (fission – fusion and fission-fusion-fission detonations), heavier Pu isotopes ( $^{240}\text{Pu}$  and heavier) are produced according to the reactions shown in Figure 2. Low yield detonations release Pu with an isotopic composition close to that of the original weapon material, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio is usually within the range 0.01 – 0.07. In general, higher yield detonations, in particular boosted detonations and thermonuclear detonations, produce Pu isotopes heavier than  $^{240}\text{Pu}$  during the detonation, increasing the  $^{240}\text{Pu}/^{239}\text{Pu}$ ,  $^{241}\text{Pu}/^{239}\text{Pu}$ ,  $^{242}\text{Pu}/^{239}\text{Pu}$  and  $^{244}\text{Pu}/^{239}\text{Pu}$  atom ratios. However, this is device dependent, as Pu is mainly produced from  $^{238}\text{U}$  present as a tamper or secondary fission material (fission – fusion – fission devices).

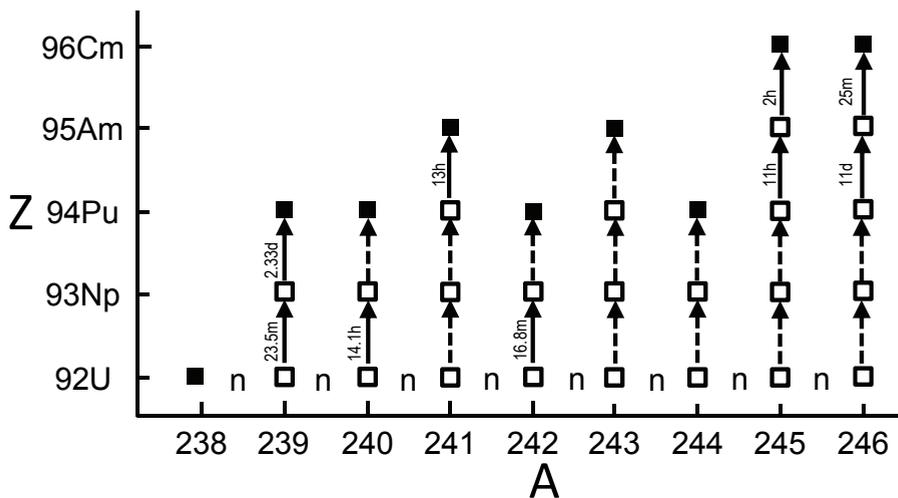


Figure 2. Neutron activation and decay to beta stable nuclides in thermonuclear detonations.  $\square$  - Beta emitting nuclide,  $\blacksquare$  - alpha emitting nuclide. Half-lives indicated along the arrows. Modified from Diamond *et al.* (1960).

Key sources of Pu isotopes and  $^{236}\text{U}$  are summarised in chapter 2. Typical  $^{240}\text{Pu}/^{239}\text{Pu}$ ,  $^{241}\text{Pu}/^{239}\text{Pu}$ ,  $^{242}\text{Pu}/^{239}\text{Pu}$  and  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios for a range of different sources are presented in Table 1.

**Table 1. Atom ratios characteristic for debris from nuclear detonations and various reactor types, ref. date (01.12.2011)**

Source	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{241}\text{Pu}/^{239}\text{Pu}$	$^{242}\text{Pu}/^{239}\text{Pu}$	$^{236}\text{U}/^{239}\text{Pu}$	Ref.
Undetonated weapons plutonium	0.01 – 0.07	-	-	-	a
Low yield detonations, U-based	0.00015 – 0.053	$(0.2 - 2.3) \times 10^{-4}$	-	-	b
Low yield detonations, Pu based	0.01 – 0.08	$(0.2 - 6.7) \times 10^{-4}$	-	-	b
Low yield detonations, GZ Semipalatinsk	$0.0438 \pm 0.0001$	$(2.21 \pm 0.035) \times 10^{-4}$	$(7.89 \pm 0.26) \times 10^{-5}$	$0.0244 \pm 0.001$	c
Global fallout, N. hemisphere	$0.182 \pm 0.005$	$(1.12 \pm 0.85) \times 10^{-3}$	$(3.71 \pm 0.3) \times 10^{-3}$	$0.235 \pm 0.014$	d, e
Bikini atoll, Ivy Mike	$0.46 \pm 0.01$	-	$0.019 \pm 0.003$	-	f
Reactor debris, Chernobyl	$0.13 - 0.53$	$0.12 - 0.13$	$0.034 - 0.048$	$5.43 - 8.14$	g
GCR-reactor, fuel burn up 3.6 GWd t <sup>-1</sup>	0.23	0.045	0.006	-	h
PHWR-reactor, fuel burn up 7.5 GWd t <sup>-1</sup>	0.41	0.077	0.023	-	h
AGR-reactor, fuel burn up 18 GWd t <sup>-1</sup>	0.57	0.184	0.093	-	h
RMBK-reactor, fuel burn up 27.5 GWd t <sup>-1</sup>	0.67	0.203	0.108	-	h
PWR-reactor, fuel burn up 33 GWd t <sup>-1</sup>	0.43	0.229	0.096	-	h

- a Warneke *et al.* (2002), Rokop *et al.* (1995), Eriksson *et al.* (2008)  
b Hicks and Barr (1984), Hansen (1995), Oughton *et al.* (2000), Smith *et al.* (2000)  
c Beasley *et al.* (1998b)  
d Kelley *et al.* (1999)  
e Sakaguchi *et al.* (2009)  
f Lindahl *et al.* (2012), Diamond *et al.* (1960)  
g Oughton *et al.* (2001), Entwistle *et al.* (2003), Salminen-Paatero *et al.* (2012)  
h Carlson *et al.* (1988)

### *Particles containing Pu and U*

Refractory (high melting point) radionuclides (e.g. Pu, U) tend to be in a particulate form when released to the environment (Salbu, 2000a). Radioactive particles released during nuclear events, are formed due to mechanical destruction of the original material, evaporation and later condensation of radioactive material, or formed during transport or residence in the environment (Salbu, 2001; Lind *et al.*, 2008; IAEA, 2011).

In a report from 2011 (IAEA, 2011), the international Atomic Energy Agency (IAEA) defines radioactive particles as:

*“Localized aggregation of radioactive atoms that give rise to an inhomogeneous distribution of radionuclides significantly different from that of the matrix background. In water, particles are defined as entities having diameters larger than 0.45  $\mu\text{m}$ , i.e. that will settle due to gravity. Radionuclide species within the molecular mass range 0.001  $\mu\text{m}$  - 0.45  $\mu\text{m}$  are referred to as radioactive colloids or pseudo-colloids. Using the grain size categories for sand, silt and clays, particles larger than 2 mm should be referred to as fragments. In air, radioactive particles ranging from submicron in aerosols to fragments are classified according to the aerodynamic diameters, where particles less than 10  $\mu\text{m}$  are considered respiratory.”*

Whereas the elemental and isotopic composition of the particles are source dependent, the shape, size, density and aerodynamic properties affecting both transport conditions and biological significance depend heavily on the release conditions such as temperature, pressure and redox-conditions (Salbu, 2000a). Any source may generate several different classes of particles during the different phases of a release. Using the Chernobyl accident as an example, large particles and fragments deposited close to the destroyed reactor were found to have different chemical and physical properties depending on the scenario under which they were released. Particles released during the initial explosion were found to be less soluble than particles released during the following graphite fire (Oughton *et al.*, 1993; Salbu *et al.*, 1994; Salbu *et al.*, 2001). Similarly, particles released during surface and low altitude atmospheric nuclear detonations exhibit different physical and chemical properties than particles from high altitude detonations due to the incorporation of environmental materials (e.g. Kemmochi (1966), Crocker *et al.* (1966), Yamamoto *et al.* (1996)).

### *Particle transport*

The extents to which radioactive particles released to the atmosphere are subjected to transport prior to deposition depend heavily on particle size, density, release height and meteorological conditions. Particles larger than 20  $\mu\text{m}$  tend to separate from the main aerosol stream relatively rapidly due to gravitational settling, and may be subjected to a different transport regime than smaller particles (Pöllänen, 2002; Bartnicki *et al.*, 2003). Smaller particles may be subjected to considerable atmospheric transport and deposit far away from the release site. Consequently large particles, smaller particles and gas phase radionuclides may deposit onto vastly different areas.

The upwards transport of debris depends on release and meteorological conditions. Materials released during fires, explosions and nuclear detonations experience significant upward transport due to convection. The extent of this transport depends on the energy released during the event, and may vary from a few hundred meters from fires to several tens of kilometres for the largest detonations (e.g. Peterson (1970), Bartnicki *et al.* (2001), Garland and Wakeford (2007)). The settling height of the debris has a large impact on transport and deposition. Debris injected into the troposphere (0 – 9 km above ground) is deposited locally and regionally, while debris injected into the lower and upper stratosphere (~9 – 17 and ~17 – 50 km above ground respectively) is deposited globally (e.g. Peterson (1970), UNSCEAR (1993)). This is further discussed in chapter 3.

### *Impact of particles containing U and Pu*

Radioactive particles may be highly radioactive due to the presence of fuel and device remnants as well as fission and activation products. These entities represent point sources of radioactivity in the environment. If taken up by living organisms, they can deliver doses to tissues. Particles smaller than 10  $\mu\text{m}$  may enter the lungs, and particles smaller than 2.5  $\mu\text{m}$  may deposit in the alveoli of the lungs, representing a point source of radioactivity in the lung tissue (Yassi, 2001; IAEA, 2011). The deposited particle may cause a zone of dead tissue extending several mm, at further distance the irradiation may be sub-lethal but with an increased carcinogenic risk (Entwistle *et al.*, 2003).

Depending on the particle characteristics, weathering and remobilisation of particles can take place over time, as illustrated in Figure 3, allowing the entrance into the food chain (Salbu, 2001). Radioactive particles also represent analytical challenges due to inhomogeneous distribution in the environment and incomplete dissolution of

samples (partial leaching), possibly resulting in an underestimation of the inventory (IAEA, 2011).

The retrieval and analysis of radioactive particles originating from nuclear accidents or atmospheric nuclear detonations could release considerable information regarding source and release conditions, atmospheric behaviour and transport as well as post depositional processes influencing mobility and bioavailability.

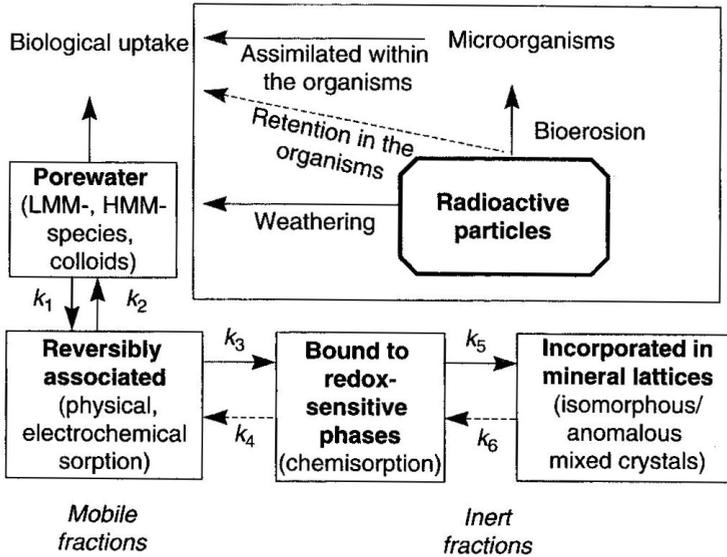


Figure 3. Processes affecting the speciation of radioactive debris deposited in soil-water and sediment-water systems. From Salbu (2000b).

### 1.1 Hypotheses and objectives of the current work

Following releases of radionuclides from nuclear weapons tests into the atmosphere of the Northern hemisphere, deposition of radionuclides have occurred in all European countries. Little information on Pu and  $^{236}\text{U}$  deposition in the Norwegian environment is available. The working hypotheses (denoted H1 – H5) of the present work are presented below.

Based on the available information on the deposition of short lived radionuclides associated with global fallout during the 1950-1960ies, fallout should be relatively uniformly distributed, and follow the precipitation pathway.

*H1: Pu originating from nuclear weapons tests should follow the precipitation pattern, and be enriched at the western coast of Norway.*

Based on the available information on the deposition of short lived radionuclides associated with nuclear reactor accidents (e.g., Windscale, Chernobyl), Norwegian territories have been affected by the fallout. Deposition of refractory radionuclides (lanthanides, Zr etc.) from the Chernobyl accident has also been observed in Norway.

*H2: Traces of Pu associated with reactor fallout should be more unevenly distributed than debris from weapons tests, and it should be possible to identify the signal at certain inland sites in Norway.*

Following historical severe nuclear events, radioactive particles have been emitted into the atmosphere. Radioactive particles from the Chernobyl accident have been deposited in many countries in Europe, including Norway.

*H3: Inert radioactive particles are expected to be present in samples affected by fallout from the nuclear weapon tests and from nuclear reactor accidents.*

Following deposition of Pu and U, several processes will influence the concentrations presently observed in top soils, such as vertical transport and production of increasing humic substance layer.

*H4: Over time the Pu and U in top soils will decrease.*

Due to the fact that Pu isotopes and  $^{236}\text{U}$  are almost exclusively anthropogenic, and formed by single and multiple neutron capture during nuclear weapon detonations (military sources) and reactor operations (civil sources), the isotopic signatures can be utilised to differentiate among military or civil sources.

*H5: Pu and U isotopic composition or atom ratios can be utilized for source identification in environmental samples. If the time resolution is sufficiently good, single events contributing to the deposition of Pu and  $^{236}\text{U}$  can be identified.*

*The objective of the current work is to quantify the activity concentrations and atom ratios of Pu isotopes and  $^{236}\text{U}$  in available archive samples in order to:*

- Estimate the inventory of Pu ( $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{242}\text{Pu}$ ) and  $^{236}\text{U}$  in samples from Norwegian territories; identify if the deposition followed the precipitation load and assess whether the top soils have been depleted with respect to these radionuclides over time (Papers I-III).
- Identify if radioactive particles are still present in fallout affected samples (Paper II, III).

- Utilise atom ratios of Pu ( $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{242}\text{Pu}$  relative to  $^{239}\text{Pu}$ ) as well as the  $^{137}\text{Cs}$  distribution pattern to distinguish between areas affected by military (global fallout) and civil (Chernobyl) sources (Paper I).
- Utilise atom ratios of Pu ( $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{242}\text{Pu}$  relative to  $^{239}\text{Pu}$ ) and  $^{236}\text{U}/^{239}\text{Pu}$  in combination with atmospheric dispersion modelling to obtain a plausible link between deposition and events such as single detonations or series of detonations. (Papers II and III).

The archive samples include soil surface samples collected at sites distributed across the Norwegian mainland in 1990 as well as in 2005 (same sites), an 28.6 m deep ice core from Nordaustlandet, Svalbard covering the period ~1950 – 2000 and selected air filter samples from 11 stations collected daily during 1957 until 1980.

To obtain qualitative (heterogeneous distributions and radioactive particles) and quantitative (activity concentrations) information on Pu isotopes and  $^{236}\text{U}$  in environmental samples, advanced techniques such as digital autoradiography, ESEM-XRMA, ICP-MS and AMS have been utilized.

## 2 Sources of radioactive contamination to the atmosphere

The input from atmospheric nuclear weapons tests and major nuclear accidents has been of local, regional and global importance, albeit mainly restricted to the hemisphere where the events occurred. Most other sources contributing to the release of anthropogenic radionuclides to the environment have had local or regional influence. The deposition of Pu and U in Norwegian terrestrial environments are therefore attributed to severe events such as atmospheric nuclear weapons tests during 1950-1960ies and major nuclear accidents occurring within the Northern hemisphere, such as the Windscale and Chernobyl accidents. To distinguish between the military (nuclear weapon) and civil (nuclear energy) sources, Pu and  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios are considered useful tools.

The process of source identification requires knowledge of the different sources, and characteristics of these. This chapter summarises some of the available information on sources contributing to radioactive contamination of the environment of the northern hemisphere.

### 2.1 Characteristics of nuclear weapon devices relevant to source identification

Following the classifications of Barnaby (1992), nuclear weapons may be characterised as pure fission weapons ( $\leq 0.05$  Mt), boosted fission weapons (0.05 – 0.5 Mt) and thermonuclear weapons ( $> 0.5$  Mt). These classes will briefly be discussed below. Pure fusion weapons and the fission cores of boosted and thermonuclear weapons are usually composed of a hollow sphere of highly enriched U (HEU, more than 90 %  $^{235}\text{U}$ ), Pu with a high abundance of  $^{239}\text{Pu}$  (93 – 99 %) or a combination of the two (Hicks and Barr, 1984; Barnaby, 1992; Hansen, 1995; Grønhaug, 2001; Lind, 2006; Ranebo *et al.*, 2007). These U and Pu isotopes have a large fission cross section for both low and high energy neutrons and are suitable as weapons material. Additionally, the fission core is often surrounded by a tamper, i.e., an outer heavy metal sphere. The tamper has the dual purpose of reflecting stray neutrons from the fission back into the weapon material and keeping the fissionable mass together for a longer time, thus increasing the efficiency of the detonation (Glasstone and Dolan, 1980; Barnaby, 1992). Tamper materials can be made from natural or depleted U, W, Be, Pb or steel (Maxwell *et al.*, 1955; Lapp, 1970; Rhodes, 1995; DOE, 1996). Natural or depleted U provides  $^{238}\text{U}$  as a secondary fissionable material when bombarded with high energy neutrons released in boosted or thermonuclear weapons, thus causing increased yield (Barnaby, 1992; Choppin *et al.*, 2002).

Boosting of nuclear weapons is accomplished through the presence of fusion material in the centre of the fission core (Barnaby, 1992). The high temperatures and pressures obtained during fission causes fusion to occur in this material and increase the yield by providing additional neutrons augmenting the on-going fission chain reactions.

Thermonuclear weapons can be classified as two stage (fission – fusion) devices or three stage (fission – fusion – fission) devices. In both categories the energy from the detonation of the first fission stage is released as high energy x-rays. This energy compresses and heats the fusion stage sufficiently to reach the very high temperatures and pressures necessary for initiating fusion. During fusion, heat and additional high energy neutrons are released. In three stage devices, these neutrons are utilised for fission of a natural or depleted U third stage / tamper (Izrael and Baxter (2002), and references therein).

### **2.1.1 Low yield weapons (<~0.05 Mt)**

Pu in debris from low yield detonations has been reported in a number of publications. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios are most often found in the range of weapon material Pu (0.01 – 0.07), indicating that little neutron capture has taken place during the detonation. Similarly, the production of  $^{236}\text{U}$  was modest and  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios in debris from low yield detonations are low, e.g. Hicks and Barr (1984), Beasley *et al.* (1998b), Lind *et al.* (2005), Lind *et al.* (2007).

### **2.1.2 High and very high yield weapons (0.05 – 58 Mt)**

Literature data (e.g. Diamond *et al.* (1960), Yamamoto *et al.* (1996), Lindahl *et al.* (2011a)) clearly shows that the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in debris from three stage (fission – fusion – fission) devices approaches that of power producing reactors with long fuel irradiation time. Information on  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in debris from two stage thermonuclear devices (fission – fusion) is scarce; however, the lack of a U tamper would imply lower production of Pu isotopes and lower  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios.

Debris from individual high yield tests at the US Eniwetak test site has shown a remarkable lack of correlation between yield and  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios. Recent work has been carried out on Eniwetak corals with a high and accurate time resolution (K. Fifield, pers. comm.). The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios (~0.4) in debris from the very high yield Ivy Mike test (10 Mt) were found to be in agreement with published values (e.g. Diamond *et al.* (1960), Lindahl *et al.* (2011a)), while two high yield tests (~2 Mt) in 1956 and 1958 produced debris with  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios of ~0.18 and 0.09 respectively. It is likely that the relatively low Eniwetak atom ratios are due to design

differences between the different devices, more specifically differences in the content of  $^{238}\text{U}$  from which heavier Pu isotopes are formed (K. Fifield, pers. comm.). Similarly, the lack of a pronounced  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio peak after the large FSU testing series in 1961 and 1962 seems to imply a lower production of heavier Pu isotopes in these detonations. Several of the very high yield atmospheric tests conducted by the FSU in 1961 and 1962 have been proposed to be two stage thermonuclear devices, thus without a fissionable uranium tamper, e.g. Lapp (1970), Grønhaug (2001), Barnaby (1992). If this was representative of FSU post moratorium testing it may explain the lack of a peak in  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios after the very high yield FSU testing series in 1961 and 1962.

Data on the production of  $^{236}\text{U}$  in high yield detonations are scarce at best. However, the generation of  $^{236}\text{U}$  should be enhanced by the high neutron fluxes and presence of  $^{235}\text{U}$  and  $^{238}\text{U}$  seen in high yield fission – fusion – fission devices.

### 2.1.3 Global (stratospheric) fallout

The term global fallout describes fallout of debris injected into the stratosphere by atmospheric detonations. This debris later deposited relatively homogeneously, mainly within the hemisphere of the injection. Global fallout is recognisable by a characteristic  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio,  $0.18 \pm 0.01$  and  $0.17 \pm 0.02$  within the northern and southern hemisphere respectively (Kelley *et al.*, 1999). The difference in the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios of fallout observed between the northern and southern hemisphere likely reflect the relative influence of the detonations carried out. While northern hemisphere global fallout carried influence from FSU high yield polar detonations and US equatorial detonations, southern hemisphere global fallout originated mainly from US and UK equatorial detonations (initially high  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios, later lower (K. Fifield, pers. comm.).

Significant perturbations of the  $^{240}\text{Pu}/^{239}\text{Pu}$  signal are found in areas affected by local fallout at testing sites (0.039 – 0.0435, Lind (2006), Beasley *et al.* (1998b)), in debris from accidents (0.055 – 0.53, e.g., Lind *et al.* (2007), Srncik *et al.* (2008), Salminen-Paatero *et al.* (2012)) and operational releases from reprocessing operations (0.03 – 0.24, Lindahl *et al.* (2011b)).

The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in stratospheric debris varies with the contemporary input. The dominant sources of stratospheric Pu in 1952 and 1954 were the very large US detonations at equatorial testing sites Enewetak and Bikini (Björklund and Goliath, 2009). These tests were conducted as surface detonations, spreading massive amounts of debris locally, tropospherically and stratospherically. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in

debris from several of these tests have been determined, (e.g. Diamond *et al.* (1960), Yamamoto *et al.* (1996), Lindahl *et al.* (2011a)),  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios in the range 0.32 – 0.46 were been reported. High  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio debris from detonations at equatorial sites was also apparent in herbage samples from England in the years 1954 – 1956 (Warneke *et al.*, 2002), and in an ice core from Greenland (Koide *et al.*, 1985) in the estimated years 1956 and 1957. These findings indicate stratospheric transport of debris from equatorial latitudes.

While the isotopic signatures of Pu from different sources are well described, concentrations of  $^{236}\text{U}$  in global fallout are underreported due to analytical challenges. Some publications on isotopic signatures of  $^{236}\text{U}/^{239}\text{Pu}$  (or  $^{236}\text{U}/^{239+240}\text{Pu}$ ) have emerged during the last few years. The  $^{236}\text{U}/^{239}\text{Pu}$  atom ratio in integrated global fallout ranges between 0.05 and 0.5 (Ketterer *et al.*, 2007; Sakaguchi *et al.*, 2009), however, significantly higher ratios are indicated in more recent samples (Srncik *et al.*, 2011; Eigl *et al.*, 2013) (Paper II).

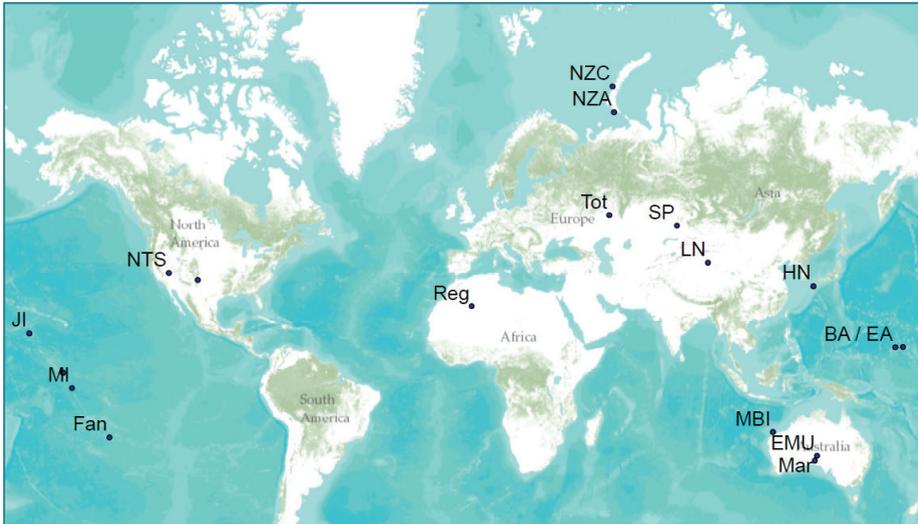
## 2.2 A brief history of atmospheric nuclear weapons testing

Atmospheric nuclear tests have been conducted by five nations: USA, FSU (FSU), UK, France and China (UNSCEAR, 2000a). Detonation sites of all known atmospheric nuclear tests, including safety tests are illustrated in Figure 4.

During 1945 – 1980, a total of 543 atmospheric nuclear tests with a total yield equivalent to 440 Mt TNT were conducted worldwide, causing local, regional and global contamination. Three main periods of atmospheric nuclear testing can be identified. The first period lasted from 1945 – 1958, and comprised US, UK and FSU testing at both hemispheres. In the period 1959 to September 1961, a moratorium was imposed during which no tests were conducted except for four French detonations in Algeria. During the second period (September 1961 through December 1962) tests were conducted by the FSU and US at north hemisphere test sites. The partial test ban treaty, signed by the Soviet Union, USA and the United Kingdom prohibited atmospheric nuclear testing by the signatory states from 1963 onwards (CTBTO, 2012). During the third period (1964 – 1980) atmospheric testing was conducted by China (north hemisphere test site Lop Nor) and France (south hemisphere test sites Mururoa and Fangataufa).

In addition, a number of peaceful nuclear detonations (PNEs) have been performed, primarily by the USA, 27 PNE detonations and the FSU, 124 PNEs and 32 tests for PNE devices (Carter and Moghissi, 1977; Cochran and Norris, 1996). These detonations were relatively low yield (<0.180 Mt) surface or underground tests

(Björklund and Goliath, 2009), and contamination should be insignificant beyond the local scale.



**Figure 4. Detonation sites of atmospheric nuclear detonations worldwide in the period 1945 – 1980. NZ – Novaya Zemlya (NZ A – Chernaya Guba, NZ B- Sukhoy Nos); Tot – Totsk; SP – Semipalatinsk; HN – Hiroshima and Nagasaki; NTS – Nevada test site (and New Mexico); JI Johnston Island; BA / EA – Bikini and Eniwetak atolls; MI – Malden Island; REG – Reggane (Algeria); FAN – Fangataufa / Mururoa; MBI - Monte Bello Island; EMU – Emu desert; Mar – Maralinga.**

#### *Northern hemisphere test sites*

According to Carter and Moghissi (1977) atmospheric nuclear detonations were conducted at six northern hemisphere test areas during 1945 – 1980. Detonations took place at Nevada test site and Pacific Proving Grounds (USA), Semipalatinsk and Novaya Zemlya (FSU) and Algeria (France) until the end of 1962. After 1962, northern hemisphere atmospheric testing took place at the Lop Nor test site (China) until 1980. A total of 445 atmospheric nuclear detonations with an accumulated yield of 428 Mt TNT equivalents were detonated at these sites (UNSCEAR, 2000a). During the most intensive period (1961 and 1962) 176 tests with a total yield of 274 Mt were conducted, mainly at the FSU test areas in Semipalatinsk and Novaya Zemlya (Björklund and Goliath, 2009). A summary of northern hemisphere atmospheric nuclear testing and gross beta activity concentrations measured in Norwegian ground level air is given in Figure 5.

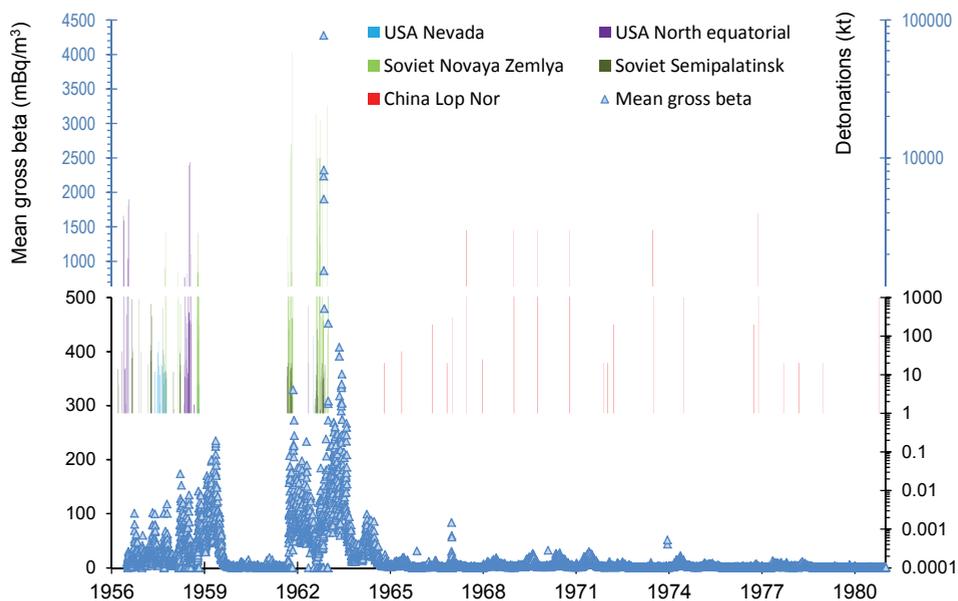


Figure 5. Northern hemisphere atmospheric nuclear detonations in the period 1959 – 1980 and mean gross beta activity concentrations measured in ground level air in Norway in the same period. Data from Björklund and Goliath (2009) and T. Bergan (pers. comm.).

### 2.2.1 Former Soviet Union test sites

#### *Semipalatinsk Testing Site (STS)*

The Semipalatinsk test site is situated in the North eastern part of Kazakhstan covering an area of 18500 km<sup>2</sup>. Two main testing areas were in operation; Ground zero where atmospheric tests were conducted, and the Degelen Mountains where underground tests were conducted (Grosche, 2002).

The first soviet nuclear detonation took place at STS in August 1949, and the site was in operation until 1989. In the period 1949 – 1962, 116 surface and air detonations with an accumulated yield of 6.6 – 6.9 Mt were carried out (Björklund and Goliath, 2009). Several tests are known to have caused considerable local and regional contamination (e.g. Gusev *et al.* (1997), Khalturin *et al.* (2005) and Gordeev *et al.* (2006)). Pu and U isotopes in local fallout have been well characterized (e.g. Beasley *et al.* (1998b), Yamamoto *et al.* (2004) and Lind (2006)). UNSCEAR (2000a) estimates that 8 of the atmospheric tests carried out at STS were thermonuclear. Nevertheless, the <sup>240</sup>Pu/<sup>239</sup>Pu atom ratios in debris associated with tests at the site are generally low. Atom ratios of <sup>240</sup>Pu/<sup>239</sup>Pu in a particle and soil samples collected at

STS ground zero were  $0.039 \pm 0.009$  and  $0.0438 \pm 0.0001$  respectively (Beasley *et al.*, 1998b; Lind, 2006).

#### *Novaya Zemlya Testing Site (NZ)*

Following concerns that fallout from high yield tests at STS could be severely detrimental to public health, decisions were made to conduct further large tests at the remote Novaya Zemlya testing site (e.g. Khalturin *et al.* (2005)). The Novaya Zemlya testing area is located in the high Arctic and consists of three testing sites; Chernaya Bay where underwater and surface detonations were conducted, Sukhoy Nos where atmospheric detonations were conducted and Matochkin Strait where underground detonations were carried out. FSU monitoring of debris from atmospheric tests at NZ revealed two main trajectories associated with significant radioactive fallout (tropospheric). One trajectory extends due South as far as the Caspian Sea, the other several thousand kilometres South East towards the Sea of Okhotsk (Khalturin *et al.*, 2005).

In the period September 1957 to late December 1962, 91 atmospheric nuclear test detonations were conducted above or on Novaya Zemlya with a total explosive yield equivalent to 240 – 255 Mt TNT (UNSCEAR, 2000a; Khalturin *et al.*, 2005; Björklund and Goliath, 2009). Detonations at NZ were large, and in combination with a large height of detonation ( $H_{det}$ ) and the low polar tropopause this caused considerable contributions to the stratospheric inventory of radioactive debris.

Publications on the radiological condition of Novaya Zemlya are scarce, but local contamination has been documented after some of the detonations (e.g. AMAP (1998), Smith *et al.* (2000), Oughton *et al.* (2004)). Particles from detonations at Novaya Zemlya in October 1958 have been identified in samples from stratospheric air above central Sweden (Sisefsky, 1961). The debris in this filter originated from two tests above Novaya Zemlya the 30. September 1958 with yields of 0.9 and 1.2 Mt. Parts of the same filter (bulked) has later been analysed by Warneke (2002), and a  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio of 0.101 was determined.

During the test series in 1961 and 1962, some very large atmospheric nuclear tests were conducted at this site. Six tests had yields exceeding 10 Mt, and the largest one of these, the Tsar Bomba, detonated the 30 October 1962, had a yield of 50 – 58 Mt (UNSCEAR, 2000a; Björklund and Goliath, 2009). The neutron fluxes in this device would have been very high. In the presence of  $^{238}\text{U}$  as tamper, this device would have generated massive amounts of heavier Pu isotopes. However, it has been indicated that the device was downscaled from its original 100 Mt configuration by substituting

the original U tamper with a lead tamper (e.g. Lapp (1970), Grønhaug (2001) and references therein).

### 2.2.2 US test sites

#### *Nevada test site*

At the Nevada test site 86 atmospheric, air or surface detonations with an accumulated yield of 1.01 Mt took place in the period 1945 – 1962 (UNSCEAR, 2000a; Björklund and Goliath, 2009). Atmospheric tests at this site were generally small (less than 0.1 Mt), and  $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{241}\text{Pu}/^{239}\text{Pu}$  atom ratios (0.00015 – 0.08, and 0.00038 – 0.0072 respectively) in debris at site were low (Hicks and Barr, 1984; Hansen, 1995). The lowest ratios in debris were associated with detonation of pure U-devices. Furthermore, the detonation heights of the tests were relatively low. A majority of the devices detonated at the surface, from steel towers, in balloons or by airdrop. Particles associated with tower shots range from spherical air-detonation resembling particles to conglomerates of particles adhering to each other (Crocker *et al.*, 1966).

#### *US equatorial tests (Bikini, Enewetak, Johnston Island, and Christmas Island).*

At Bikini and Enewetak atolls 63 detonations were carried out at surface, placed on barges, mounted in towers, or by air drop by the US in the period 1946 – 1958 (Björklund and Goliath, 2009). Detonation heights were generally low and environmental materials were readily fused with debris from the detonations. Altogether 35 thermonuclear detonations were performed at the two atolls; most of these were surface detonations, including the first US thermonuclear device (Ivy Mike) and the largest (Castle Bravo). Severe local contamination has occurred on at least one occasion, Castle Bravo detonation, where flake like particulate debris was deposited up to 200 km eastwards from the detonation within hours after the detonation, affecting local residents, fishermen and US service men (e.g. Yamamoto *et al.* (1996), Cronkite *et al.* (1997), UNSCEAR (2000a)). Pu linked to high yield detonations at the Marshall Islands is associated with high atom ratios, e.g. Castle Bravo:  $^{240}\text{Pu}/^{239}\text{Pu}$   $0.32 \pm 0.03$  (Yamamoto *et al.*, 1996), and Ivy Mike:  $^{240}\text{Pu}/^{239}\text{Pu}$   $0.46 \pm 0.01$  (Lindahl *et al.*, 2012).

Tests at Johnston Island and Christmas Island were all detonated at sufficient height to avoid local fallout. At Johnston Island 7 high altitude ( $H_{\text{det}} > 21000$  m) and 5 atmospheric tests were conducted, while at Christmas island 24 atmospheric tests were conducted in the period August 1958 – November 1962 (Björklund and Goliath, 2009).

### 2.2.3 French test sites

A series of 17 low yield (0.0007 – 0.12 Mt) tests was carried out on ground or in tower (Reggane desert) and underground (Ekker) in the period 1960 – 1966 (Björklund and Goliath, 2009). The French aboveground testing in Algeria were carried out during the partial test ban treaty period (1958 – 1961). Air concentrations of radioactivity at this time were low, and debris from at least one of the tests was detected at distant locations (Hvinden, 1960; Patterson and Lockhart, 1960; Lindblom, 1961). Lindblom (1961) found that particles from the largest French atmospheric detonation in Algeria (13 February 1960, 0.07 Mt) had reached Sweden. Local contamination associated with close in fallout from the aboveground detonations and venting from underground tests has also been reported (IAEA, 2005).

### 2.3 Accidents during transport of nuclear weapons

UNSCEAR (1993) lists fourteen accidents involving the loss of nuclear devices, the two most severe were the Palomares (1966) and Thule (1968) aircraft accidents involving the disintegration of two and four thermonuclear devices, respectively. Later investigations (e.g. Lind *et al.* (2005), Jimenez-Ramos *et al.* (2007), Lind *et al.* (2007), Eriksson *et al.* (2008)) indicated the weapon material to be mixed U / Pu metal oxide with low  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios. There was no nuclear yield involved in any of the accidents, and contamination was limited to the local scale (Mitchell *et al.* (1997) and references therein).

### 2.4 Releases from the nuclear fuel cycle

Releases of radionuclides from the nuclear fuel cycle are associated with mining and milling, reactor operation, reprocessing and inadequate storage or dumping of waste. Releases from mining and milling operations are first and foremost associated with remnants of U and daughter products in the mine tailings, and give rise to higher than background releases of natural radionuclides in the vicinity of the sites (UNSCEAR, 1993). Nuclear reactor operation and post irradiation fuel handling have caused both operational and accidental releases of radionuclides to the environment. Reprocessing plants release fission products and transuranium elements, mainly to the aquatic (marine and rivers) environment. Long lived nuclides (e.g.  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ ,  $^{99}\text{Tc}$ ,  $^{129}\text{I}$  and  $^{137}\text{Cs}$ ) are of major concern (UNSCEAR, 1993))

#### 2.4.1 Nuclear reactor accidents

UNSCEAR (1993) states six accidents that have caused exposures of the general public at four installations handling nuclear fuel (two at the FSU Mayak production

centre; long term releases from the Windscale piles and the Windscale fire; the Three Mile Island accident, and the Chernobyl accident). The Fukushima accident in 2011 also caused the release of radionuclides to the atmosphere. The accident took place out of the range of the samples employed in the present work, and furthermore, Pu contamination in Norway from this accident is unlikely. Of the accidents mentioned in UNSCEAR (1993), only the Chernobyl accident and possibly the Windscale fire could be of significance to Norwegian territories.

### *Windscale*

The Windscale piles were two air-cooled, graphite-moderated reactors in operation in the period 1950 – 1957. Major releases of radionuclides have happened at two occasions. Long term release occurred during normal operation in the period 1952 – 1957; fuel elements with broken cartridges and fuel elements jammed in the coolant stream were oxidised at low temperature resulting in the release of U-matrix particles with fission and activation products (Chamberlain, 1987; Salbu *et al.*, 1994; Smith *et al.*, 2007). Particles were described as large (10 – 250 µm), flake like and layered (Figure 6); the structure of the particles could lead to longer residence times in the atmosphere than comparable spherical particles (Salbu *et al.*, 1994).

During a routine operation of annealing Wigner energies stored in the graphite moderator of one of the reactors, the 7 October 1957, the moderator overheated resulting in a fire (UNSCEAR, 1993; Garland and Wakeford, 2007). Volatiles and particulates were released at several occasions during attempts to cool the moderator and extinguish the fire (UNSCEAR, 1993; Garland and Wakeford, 2007). An estimated 1 – 42 g Pu was released (Garland and Wakeford, 2007), and since the reactor was operated for weapon production purposes  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios can be assumed to be low.

Due to the high temperatures and low air flows during the accident the release of U particles would have been low (Chamberlain, 1996). The debris plume from the Windscale accident moved south – southeast towards central Europe before turning northwards and was observed to pass over Norway (Storebø, 1958; Hvinden and Lillegraven, 1963; UNSCEAR, 1993; Garland and Wakeford, 2007).

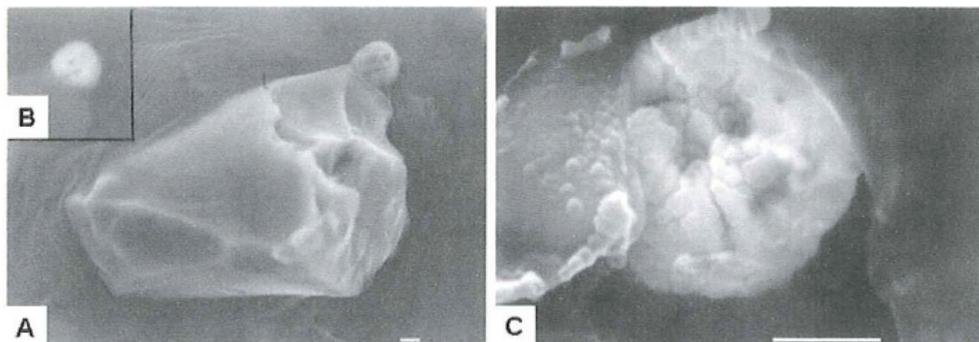


**Figure 6. Fuel particle captured in the vicinity of the Windscale plant in 1956, demonstrating the flake like structure of the particles released during 1952 – 1957. From Salbu *et al.* (1994)**

### *The Chernobyl accident*

During system testing on Saturday 26 April 1986 one of the reactors at the Chernobyl power plant was run at low output. The reactor output became unstable due to a combination of design weaknesses and operator errors. The power level of the reactor increased rapidly and caused a release of the primary coolant and pressure loss within the reactor. The overheated coolant (light water) vaporized instantaneously. A fuel vapour explosion followed, disrupting the core and parts of the building, exposing the graphite moderator to the atmosphere (UNSCEAR, 2000b).

Radioactive debris was released in the initial explosions as well as from the subsequent fire, which lasted until the 5 May. Vast areas of Europe were severely affected, and both volatile species as well as particles originating from the accident have been identified at large distances from the plant, including Scandinavia, 2000 km away from the accident site (e.g. Devell *et al.* (1986), Backe *et al.* (1987) Salbu (1988a), Pöllänen *et al.* (1997) and references therein, UNSCEAR (2000b) and references therein, Salminen-Paatero *et al.* (2012)). About 6 – 8 tons of uranium based fuels with an estimated 20 kg of Pu isotopes ( $^{239}\text{Pu}$  –  $^{242}\text{Pu}$ ) were released during the Chernobyl accident (UNSCEAR, 2000b).



**Figure 7. Ru-particle originating from the Chernobyl accident attached to a carrying silicon particle. The particle was retrieved in Valdres Norway in 1987 (Salbu, 1988b; Lind, 2006). A) General overview of the Ru – silicon conglomerate B) BEI image of the Ru-particle C) High magnification image of the Ru-particle. Bar = 1  $\mu$ m.**

Isotope ratios calculated from core inventory estimates differ significantly from global fallout ratios. A  $^{236}\text{U}/^{239}\text{Pu}$  atom ratio of 9.3 was estimated while for Pu isotope ratios, estimates range considerably (UNSCEAR, 2000b). The  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios was estimated to range between 0.34 and 0.44,  $^{241}\text{Pu}/^{239}\text{Pu}$  between 0.134 and 0.273, and  $^{242}\text{Pu}/^{239}\text{Pu}$  between 0.046 and 0.042 (UNSCEAR, 2000b). Measured  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios within the exclusion zone and in air in Vilnius (Lithuania) during passage of the plume from the accident ranged between 0.38 – 0.41, and fits well with the estimates (Muramatsu *et al.*, 2000; Lujanienė *et al.*, 2009).

Publications addressing debris from the Chernobyl accident deposited in Fenno-Scandinavia indicate highly varying  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios. Lindahl *et al.* (2004) found atom ratios in the range 0.16 – 0.44, mainly due to mixing with global fallout. Salminen-Paatero *et al.* (2012) on the other hand found ratios in the range 0.13 – 0.53 in samples associated with fresh deposition (hot particles, commercially exploited peats, and lichens). The large variation in Pu atom ratios observed in the latter work likely reflects the Chernobyl core included U fuels with varying (low and high) burn up.

Debris from the Chernobyl accident moving towards Fenno-Scandinavia was vertically divided into two main air layers. Materials in the upper layer mainly affected Finland, while materials from the lower layers affected Sweden and Norway (Saltbones, 1986; Persson *et al.*, 1987; Pöllänen *et al.*, 1997). Deposition of debris from the Chernobyl accident is addressed in paper I and II.

## *Nuclear fuel reprocessing*

Several reprocessing plants have been in operation in the northern hemisphere. According to Skipperud (2004), three Former Soviet union weapon production plants: Chelyabinsk-65 (Mayak) since 1948, Tomsk-7 since 1953 and Krasnoyarsk-27 since 1958; two UK weapon production and later reprocessing plants: Windscale, since 1951, Dounreay, since 1955; and the French COGEMA la Hague plant (in operation since 1965) are or have been in operation. The main releases of radionuclides from these plants have been to rivers or the ocean. The Ob and Yenisei Rivers, which were contaminated by FSU weapon production plants eventually discharge into the Kara Sea and the Arctic. Similarly, effluents from the Sellafield, Dounreay and La Hague have been discharged into surface waters in the Irish Sea and the English Channel, respectively, ultimately leading to the North Sea. Several papers have addressed the transport of effluents from European and FSU reprocessing and weapon production into the Arctic (e.g. Raisbeck *et al.* (1995), Beasley *et al.* (1998a) and Herrmann *et al.* (1998)). The isotopic composition of Pu in sediment profiles collected close to the Sellafield reprocessing plant represent a crude history of the reprocessing activities at the site. Pu from the early period (~1960) of weapons grade Pu (~0.06), later  $^{240}\text{Pu}/^{239}\text{Pu}$  in the effluents increased, and peaked around 1980 with a  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio of ~0.25 (Kershaw *et al.*, 1995).

### **2.5 Satellite accidents**

Six satellites carrying radioisotope thermal generators (RTG) have been destroyed during launch and re-entry in the period 1964 – 1983 (Krey *et al.*, 1979; Leifer *et al.*, 1987; UNSCEAR, 1993). For three of these cases the RTG or reactors have been recovered entirely or lost at sea without further releases of radionuclides. Two of the three remaining satellites disrupted completely during re-entry, while the last satellite disintegrated partially or completely upon re-entry (UNSCEAR, 1993).

#### *SNAP-9A*

In 1964, a Transit navigational satellite equipped with a SNAP-9A RTG disintegrated upon re-entry into the atmosphere causing the release of 950 g  $^{238}\text{Pu}$  into the southern hemisphere stratosphere (Hardy *et al.*, 1973; UNSCEAR, 1993). Due to the low  $^{238}\text{Pu}$  background, the input of  $^{238}\text{Pu}$  from this accident was considerable. The inventory of  $^{238}\text{Pu}$  in the environment increased threefold with a 20 : 80 distribution between the northern and southern hemisphere (Hardy *et al.*, 1973).

### *Cosmos-954*

In 1978, the FSU Cosmos 954 satellite partially burned during re-entry. Following disintegration volatiles were largely released in the stratosphere, while large shrapnel and refractories scattered over vast areas in Canada. The power source of this satellite has been indicated to contain enriched U (Krey *et al.*, 1979). Uranium isotopes and an estimated 120 g of  $^{239}\text{Pu}$  was released during the accident (UNSCEAR (1993) and references therein).

### *Cosmos-1402*

In 1983 the FSU Cosmos 1402 satellite re-entered the atmosphere and disintegrated over the South Atlantic Ocean. This accident caused releases of 50 kg of U highly enriched in  $^{235}\text{U}$  (Leifer *et al.*, 1987). In 1984 and 1985, altered  $^{235}\text{U}/^{238}\text{U}$  activity ratios (Bakhtiar *et al.*, 1985) and  $^{239+240}\text{Pu}$  elevated activity concentrations (Salaymeh *et al.*, 1987) in rainwater in Fayetteville (36 °N) indicated that debris from the Cosmos 1402 had reached the northern hemisphere.

### 3 Transport and atmospheric behaviour

Transport and deposition of radioactive debris released to the atmosphere depends on a number of factors. Both source related characteristics like chemical reactivity, hygroscopicity, particle size and release height, as well as environmental conditions like wind direction, humidity, land topography and precipitation affect residence time and deposition.

This chapter will briefly summarise source related and environmental factors affecting the atmospheric transport and deposition of radioactive aerosols and gases.

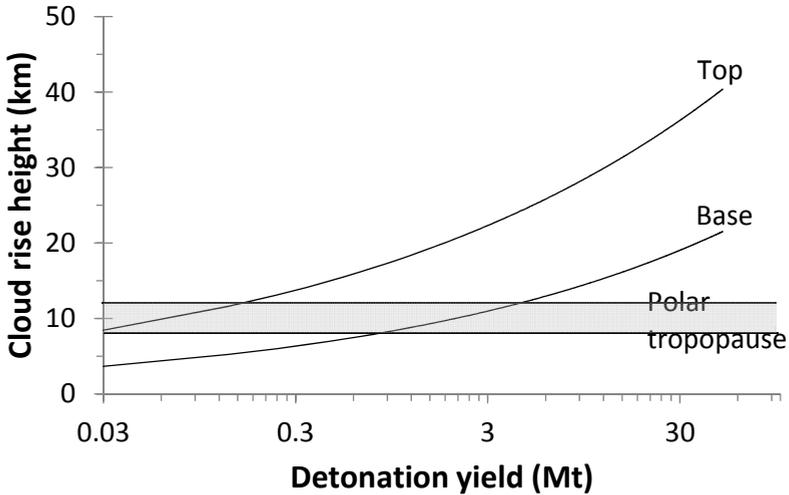
#### 3.1 Plume rise

During nuclear detonations very large amounts of energy were released within a very short time range. Consequently, the device, supporting materials and environmental materials close to the detonation reached temperatures of  $10^6 - 10^7$  K (Glasstone and Dolan, 1980). The extremely hot gases and vapours formed a rapidly rising and expanding fireball containing device remnants, supporting materials and incorporated environmental materials. The height to which the debris cloud rose depended on the detonation height, detonation yield, and meteorological conditions (Glasstone and Dolan, 1980). Atmospheric compartments and cloud rise observations according to Peterson (1970) are given in Figure 8 and Figure 9.

Low yield detonations released the main part of their debris into the troposphere, (Figure 9). Debris injected in the troposphere had a mean residence half-life of 72 days (Holloway and Hayes, 1982), and although it has been postulated that the debris was deposited mainly at the latitude bands in which the detonation took place, debris have been shown to spread significantly (Machta *et al.*, 1956; Hvinden *et al.*, 1964) (paper II).

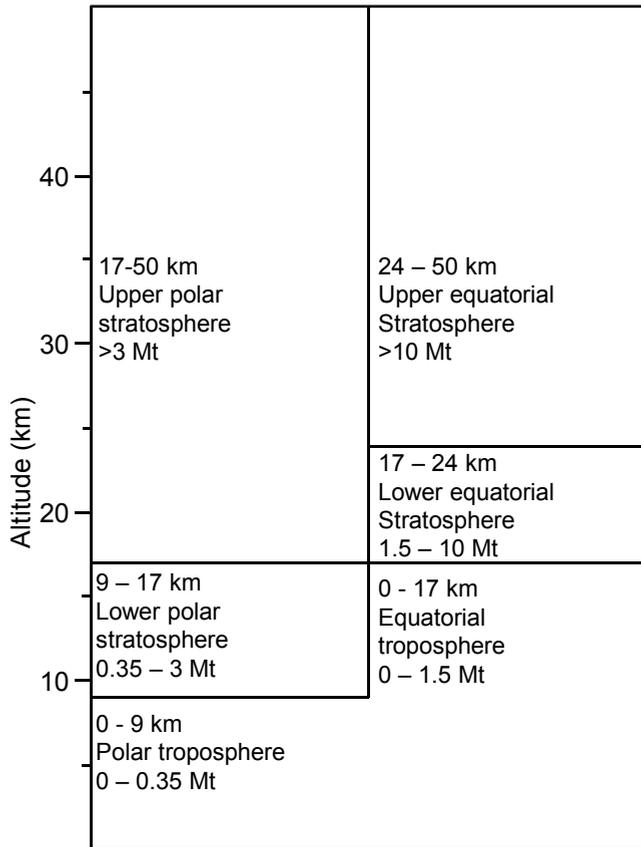
Insertion of debris above the tropopause started to occur for detonations larger than 0.4 Mt in polar regions and 1 Mt in equatorial regions (Peterson, 1970). Debris injected into the stratosphere is indicated to have had residence times in the range 5 months to 2 years, depending on latitude, injection height and season (Junge, 1963; Telegadas and List, 1969; Peterson, 1970). The distribution of debris between the compartments of the stratosphere had significance not only for the residence time, but also the distribution of debris between the hemispheres. Debris injected into the lower polar stratosphere (9 – 17 km) had estimated residence half-lives of 5 months before being deposited in a 20:1 distribution between northern and southern hemisphere respectively. Debris injected in the upper north polar stratosphere on the other hand

had estimated residence half-lives of 2 years and was deposited in a 4:1 distribution between the northern and southern hemispheres (Peterson, 1970).



**Figure 8.** Elevation of base and top of mushroom cloud caps from nuclear detonations as a function of detonation yield in polar regions according to Peterson (1970). The shaded box represents approximate locations of the summer (upper confinement) and winter (lower confinement) polar troposphere.

The stratosphere is assumed to serve as a relatively well mixed inventory of radionuclides from diverse high yield and high elevation detonations, likely due to mixing effect in the lower stratosphere prior to transfer to the troposphere. Nevertheless, it has been indicated that clouds of concentrated activity existed in the stratosphere for months after detonations (Storebø, 1959).



**Figure 9. Atmospheric compartments modified from Peterson (1970). Yield numbers indicate the size of detonations estimated to inject more than 50 % of its debris in the respective compartments. Polar latitudes defined as latitudes above 30 °N or S.**

### **3.2 Atmospheric circulation**

The general atmospheric circulation is based on three idealized large circulation patterns: the Hadley cell, Ferrel cell and the polar cell (Figure 10). Chapters 3.2.1 through 3.2.4 give a brief presentation of the circulation patterns and the effects on a regional scale. The description is based on Ahrens (2003), unless otherwise stated.

#### **3.2.1 Hadley cell**

The highest energy transfer by solar irradiation is usually at tropical latitudes, near equator. This causes the formation of warm moist air which rises until it reaches the tropopause where it is deflected polewards (the tropopause represents a barrier through which tropospheric air generally does not penetrate). The increased elevation and the adiabatic cooling of the air (also possibly the encountering of cold tropopause

air) cause the formation of precipitation, and the rising air keeps the tropopause suspended here at a higher elevation than further north and south. The air deflects and moves polewards while losing heat by radiation on the way south / north. Subsidence patterns starts to emerge around 30 °N/S (the subtropics). The subsidence and convergence of winds from equator causes high pressure areas of surface air to emerge at these latitudes. The higher air pressure forces surface air to move towards equator and the poles, due south or north; however, the Coriolis Effect deflects the wind towards its right and causes surface winds between equator and 30 °N/S to be moving in an easterly direction.

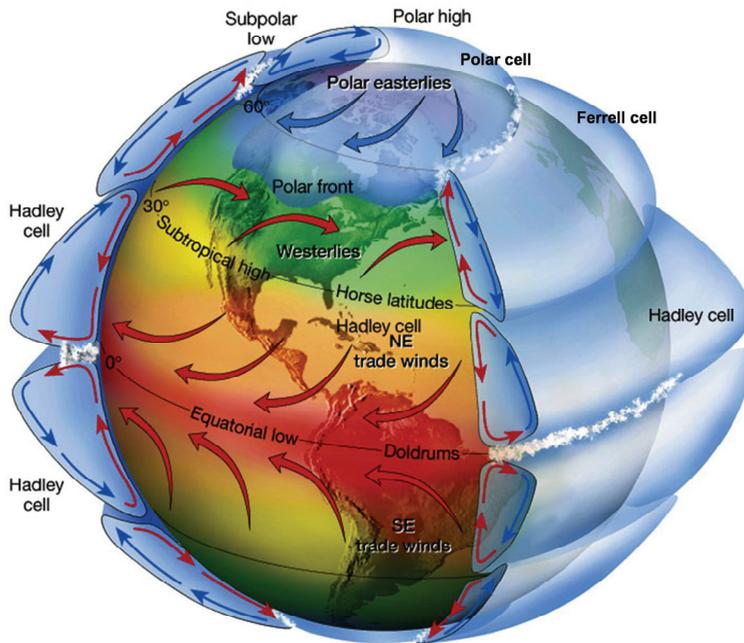


Figure 10. Atmospheric circulation patterns according to Lutgens and Tarbuck (2001).

### 3.2.2 Ferrel cell

The increased surface pressure at subtropical latitudes (30 – 60 ° N / S) caused by the Hadley cell forces mild air to be shifted northwards. Due to the Coriolis effect the prevailing wind direction in this cell is westerly. Further north, at latitudes depending on season and local circumstances the northwards moving air encounters cold polar southwards moving air and forms the polar front of low pressure at approximately 60 °N. At the polar front air masses rise and again move towards the subtropics and the poles.

### 3.2.3 Polar cell

Driven partially by the northwards moving air from the polar front and partially by the subsiding air (and high pressure) at the poles, the polar cell cause easterly winds. As mentioned above the polar front is not stationary and the polar cell may extend as far south as 40 °N covering substantial areas of snow covered continental Eurasia and North America in winter (Barrie, 1986; Warneke, 2002).

### 3.2.4 Arctic contamination

Several authors have indicated that continental Eurasia is an important contributor to arctic pollution (e.g. Barrie and Hoff (1984 ); Barrie (1986), Barrie *et al.* (1992), Stohl (2006)). During winter the arctic air mass extends as far south as 40 °N over continental Eurasia and Northern America; the air mass form an isolated dome limited southwards by the polar front. A semi-permanent high pressure area over Siberia forces transport of tropospheric air on its west side towards the Arctic, while a high pressure ridge above North America extracts air southwards from the arctic (Barrie, 1986; Barrie *et al.*, 1992; MacDonald *et al.*, 2000). Fluctuations in the transport have been observed to depend on the North Atlantic Oscillation (NAO) defined as the pressure difference between a permanent low pressure area above Iceland and a permanent high pressure above the Azores / south-west Europe (Walker and Bliss, 1932). Transport of tropospheric air masses from Eurasia and Northern America towards the Arctic is stronger during periods of a positive North Atlantic Oscillation index, defined as a large difference in the atmospheric pressure at the mentioned locations. The effect is largest for transport from Europe and least for transport from Asia (Eckhardt *et al.*, 2003).

## 3.3 Importance of particle characteristics

During the course of an atmospheric nuclear detonation unspent weapon material, fission products, activation products, and device remnants are evaporated by the energy released in the detonation, and form a fireball. Upon expansion and interaction with the surrounding atmosphere the fireball cools and eventually reach the condensation temperature of the different elements present (Junge, 1963).

For particle categorisation, important distinctions arise between surface detonations and air detonations. Air detonations were those detonations carried out at sufficient height to avoid large amounts of surface materials being incorporated into the fireball. Usually the limit between a surface detonation and an air detonation is set by the expression  $H=55w^{0.4}$ , where H is the height above ground of the detonation and w is the yield in kt (Peterson, 1970). Detonations carried out below this height are denoted

surface detonations, as the fireball from the detonations is in contact with the surface. Local deposition of debris is higher, and in addition larger particles with a higher incorporation of environmental materials are produced. High yield tests, especially FSU tests at Novaya Zemlya were usually performed sufficiently high above ground to avoid interactions between the fireball and the ground (UNSCEAR, 2000a). According to Grønhaug (2001), most FSU air detonations took place above a height of  $H=100w^{0.33}$ , thus ensuring very little interaction with ground materials.

Air detonation particles formed after condensation of device remnants alone or onto atmospheric particulates. The resulting particles were predominately spherical in shape and with a uniform distribution of radioactivity throughout (Crocker *et al.*, 1966; Heft, 1970; Peterson, 1970). Particle diameters in the range 0.01 - 20  $\mu\text{m}$  were frequently reported, e.g. Sisefsky (1961), Sisefsky (1966), Glasstone and Dolan (1980). The particle population after surface detonations tended to form two distinct groups. One group which resembled the air detonation particles mentioned above, and the other group formed from surface materials directly affected by, or drawn into the fireball. The latter group of particles were generally larger and more irregular in shape following condensation of debris onto unmelted or partially melted environmental materials (Heft, 1970). Furthermore, the distribution of radioactivity was more uneven in this group, the activity tended to be higher on the surface of the particles (Crocker *et al.*, 1966; Glasstone and Dolan, 1980).

### **3.4 Factors affecting atmospheric residence time and deposition**

The residence time of particles in the atmosphere is determined by the injection height, particle characteristics and meteorological conditions during release and deposition. Larger particles ( $>20 \mu\text{m}$ ) are deposited relatively rapidly to form local fallout (Junge, 1963). Reports of larger particles transported over a long range are rare e.g. Mamuro *et al.* (1965), Pöllänen *et al.* (1997). Most particles detected at some distance from the detonation or accident site are substantially smaller than 20  $\mu\text{m}$  (e.g. Sisefsky (1966), Kemmochi (1966), Persson and Sisefsky (1971), Pöllänen *et al.* (1997)).

Small particles are not efficiently removed from the atmosphere by gravity alone, e.g. Storebø (1960). Instead, such particles have been found to be subject to considerable wind transport, and have been retrieved at long distances from the detonation sites (Sisefsky, 1961; Mamuro *et al.*, 1962; Mamuro *et al.*, 1963). Sisefsky (1961) located and described more than 900 particles from high yield detonations at Novaya Zemlya in September and October 1958. These highly radioactive particles were captured in the low stratosphere above central Sweden some 17 days after the detonation. They

were found to be spherical, and in the size range 0.2 – 5 µm, the lower limit being set by the resolution of the optical microscope used. Fallout particles with diameters up to 14 µm originating from detonations at Semipalatinsk or Novaya Zemlya have also been retrieved in Japan, some 4000 - 6000 km from the detonation sites (Mamuro *et al.*, 1962). Debris from Chinese nuclear tests was retrieved in Sweden on several occasions, e.g. Sisefsky (1966), (1967), Sisefsky and Persson (1971) and Persson and Sisefsky (1971). Particles were captured above and below the tropopause as well as on ground level after transit times as short as 8 days. A range of different particles were described, from dense relatively small (0.2 – 5 µm) particles originating from air detonations to larger less dense and insoluble particles from surface detonations. Finally, particles from the Chernobyl accident were transported and deposited far from the accident site. Pöllänen *et al.* (1997) A comprehensive review of Chernobyl particles retrieved at various sites in Europe can be found in Pöllänen *et al.* (1997).

Atmospheric particles generated in nuclear detonations rapidly become a part of the atmospheric particle population. As such, they are subjected to the same processes as natural and other anthropogenic particles. The most important mechanism for the removal of tropospheric particles is wet deposition, in which the particles act as cloud condensation nuclei (CCN), onto which atmospheric moisture condenses. Water insoluble particles larger than ~0.1 µm and water soluble particles larger than 0.01 µm may serve as CCN (Wallace and Hobbs, 2006). Further, particles may be incorporated into existing water droplets by diffusion (diffusional collection) or by impact with falling precipitation (inertial collection) (Wallace and Hobbs, 2006). Precipitation thus removes (radioactive) particles from the atmosphere and deposits these on the ground, i.e., there is a correlation between mean precipitation loads and deposition of radioactive debris (Storebø, 1959; Storebø, 1968; Lee *et al.*, 1997; Palsson *et al.*, 2013).

### **3.5 Post depositional processes of relevance to the present thesis**

#### *Humic surface soil*

Refractory radionuclides released during high temperature events like nuclear weapon detonations and power plant accidents are assumed to be deposited as particles or fragments (IAEA, 2011). No publications have been found which directly addresses the mobility of radioactive particles in natural soils. However, several publications have shown that a major proportion of GFO-related Pu and Cs still resided within the upper 10 - 15 cm of undisturbed soils, and that the concentration decrease exponentially with depth (e.g. (Bunzl *et al.*, 1995; Lee and Lee, 2000; Lee and Lee,

2001)). Cs and Pu seem to be strongly bound to both organic and mineral soils (Bunzl *et al.*, 1998; Lee and Lee, 2001; Skipperud, 2004).

### *Ice core*

Contaminants are deposited onto the glacier surface with precipitation (wet deposition) or directly (dry deposition). Meltwater percolation occurs for glaciers in which melting during the warm seasons exceeds the infiltration capacity of the annual layer. Consequently, meltwater containing contaminants percolate past the annual layer potentially disturbing the stratigraphy. The estimated melt index of Svalbard glaciers ranges within 15 – 25 %, and the effective percolation depth of the Austfonna glacier is estimated to range within 2 – 5 years (Tarussov, 1992). Pinglot *et al.* (2003) estimates a downward migration corresponding to between 0 and 1.6 year for the AUS-99 ice core investigated in the present work. The Austfonna ice core has been dated based on the  $^3\text{H}$  concentration profile, with the 1963  $^3\text{H}$  peak at 21.3 m physical depth (Watanabe *et al.*, 2001). However,  $^3\text{H}$  is incorporated in the glacier as water, and may behave more conservatively than particular radionuclides upon infiltration of meltwater.

## 4 Materials and methods

Atmospheric radioactive debris can be characterised by the activity concentrations, radionuclide speciation and atom ratios. Particle diameter, shape and density reveal important information on the release scenario, while the nuclide and isotopic composition serves source and age identification purposes. During the course of the present work, digital autoradiography has been employed in order to locate and isolate individual particles captured in air filters, ESEM (Environmental Scanning Electron Microscope) has been applied for further isolation of potential particles, while synchrotron based x-ray microtechniques (Hasylab) has been attempted for speciation analysis of hotspots identified by autoradiography. Concentrations of U and Pu, and respective atom ratios in the samples have been determined by SF-ICP-MS and AMS. Chapter 4 describes the sample preparation and analytical methods used in the present work.

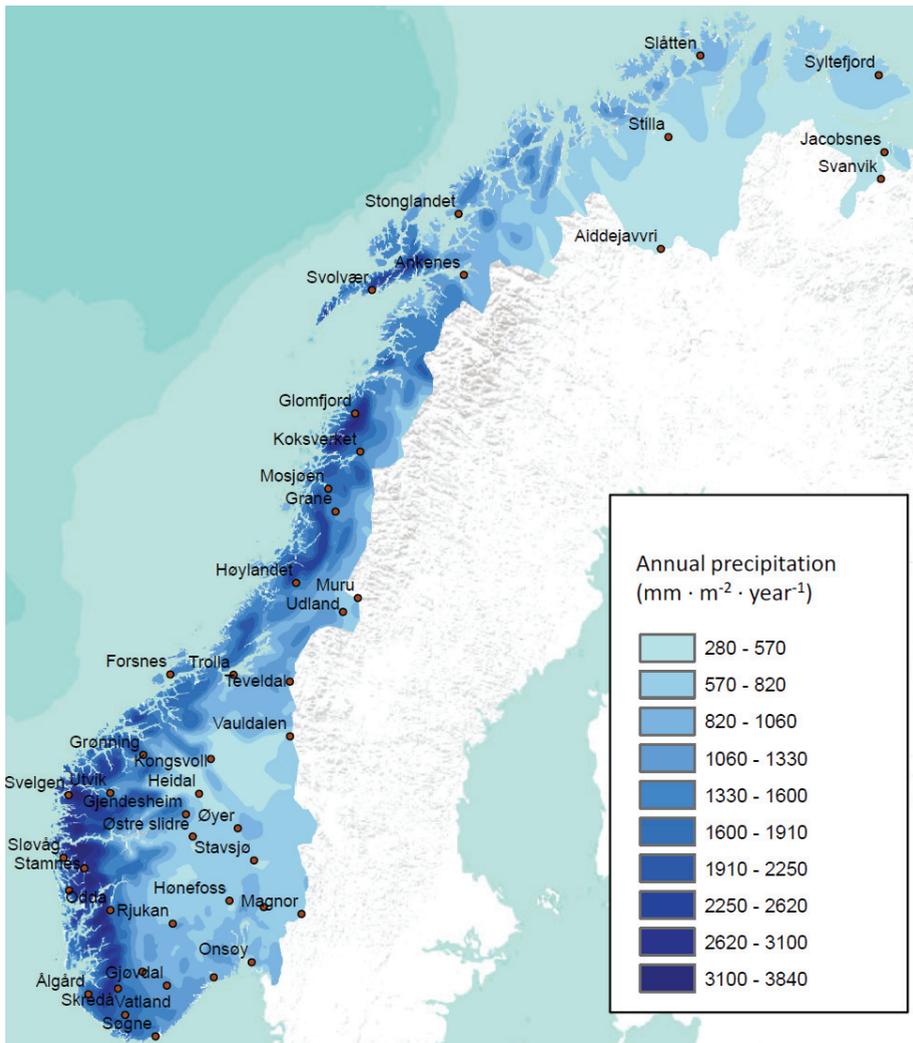
### 4.1 Samples investigated

#### *Humic surface soil samples*

The Humic surface soil samples were collected in 1990 and 2005 (courtesy of Eiliv Steinnes) at 45 locations across Norway, representing all regions (Figure 11). An area of 330 cm<sup>2</sup> to a depth of 7 cm was sampled from the surface soil layer at each location. Assuming that global fallout has been retained within the sampling depth, the samples represent the accumulated deposition of radionuclides released to the atmosphere. Although great care was taken in order to sample exactly the same locations in 1990 and 2005, minor deviations could occur due to problems with relocating the exact position. The samples were air dried at room temperature prior to storage.

#### *Ice core samples*

The ice core was drilled at the summit of the Austfonna glacier (79.8333 °N, 24.0167 °E, and 750 m a.s.l.) in 1999 to a depth of 289 m (Figure 12). The ice core was cut into appropriate sections based on information available from previous studies of the ice core (Pinglot *et al.*, 2001; Watanabe *et al.*, 2001; Pinglot *et al.*, 2003) combined with gross beta activity concentrations in Norwegian ground level air (T. Bergan, pers. comm.).

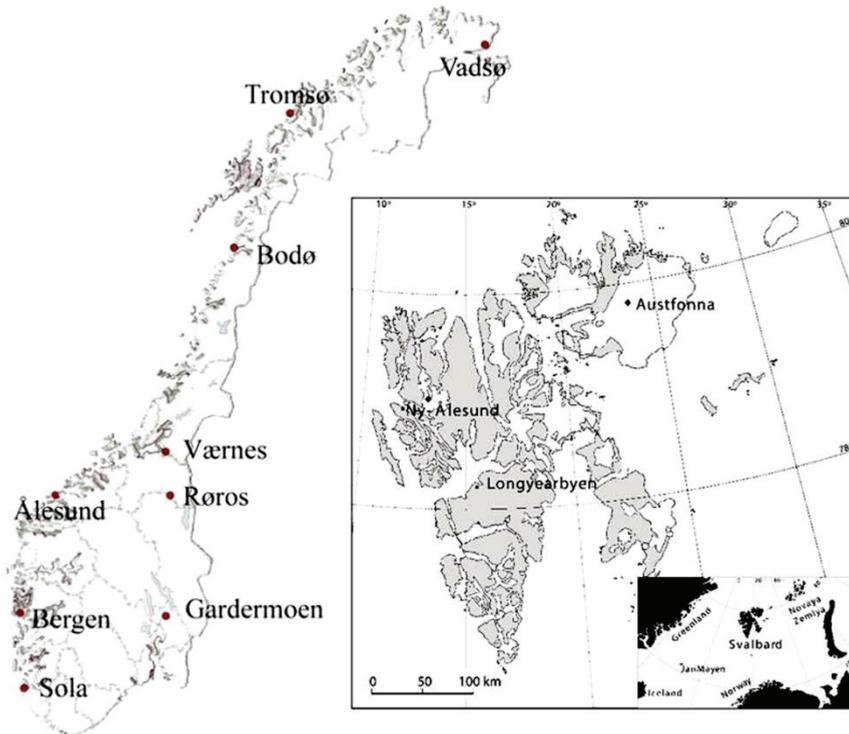


**Figure 11.** The sampling locations for humic surface soil samples (paper III). Mean annual precipitation indicated.

### *Air filter samples*

As a consequence of atmospheric nuclear weapons testing from 1945 onwards, the Norwegian Defence Research Establishment (FFI) operated a surveillance and monitoring program for radioactive fallout (Njølstad, 2006). Gross beta activities were measured in deposition (wet and dry deposition), milk and surface air from 1957 until the mid-1980s. Radioactivity in ground level air was monitored through the operation of 2 – 10 air filter stations nationwide (cf. Figure 12). According to Lockhart *et al.* (1964) the filters were specified to retain 99.9 % of atmospheric

particulates larger than  $0.3 \mu\text{m}$  at the flow rates used ( $18 \text{ m}^3 \text{ h}^{-1}$ ). The filters were changed every 24 h, and gross beta activities were analysed 48 – 72 h after the collection (Hvinden, 1958).



**Figure 12.** Map of and air filter stations in mainland Norway (right), Svalbard with the sampling location of the AUS-99 ice core on Austfonna (left).

Air filters were primarily selected from Bergen, representing a southern coastal site, Røros representing an inland site and Vadsø for its proximity to the NZ testing site. The sample selection was designed to identify periods of atmospheric nuclear testing, hence filters from the pre-moratorium (1957 and 1958), immediately after the cessation of the moratorium (September 1961 and onwards) and filters from the periods of spring maximum deposition in 1962 and 1963 were analysed. Filters were also selected when high gross beta activities indicated the arrival of debris from atmospheric nuclear weapon testing. Finally three samples were prepared representing ten consecutive days of air filter sampling at Bergen (1957) and Røros (1961 and 1962); these samples were prepared without the use of yield monitor, in order to measure  $^{242}\text{Pu}/^{239}\text{Pu}$ , and  $^{244}\text{Pu}/^{239}\text{Pu}$  atom ratios.

An overview of the samples analysed in the present work is given in Table 2.

**Table 2. Overview of the samples analysed in the present work**

Sample type	Time period	Main purpose and limitations	Analytes	Key sources
Humic surface soils, mainland Norway, samples from 45 sites in 1990 and 2005, (n=87)	1945 – 2005	Geographical distributions, fallout inventories, source identification and wash out.	$^{137}\text{Cs}$ , $^{238}\text{Pu}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{241}\text{Pu}$ , $^{242}\text{Pu}$	Integrated fallout: NWT, satellite accidents, Windscale and Chernobyl accidents
Ice core, Nordaustlandet, Svalbard (n=9)	~1949 – 1999	Temporal distribution, source assessment. Low time resolution	$^{234}\text{U}$ , $^{236}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{241}\text{Pu}$	Chronology, all sources. NWT, satellite accidents, reactor accidents
Air filters, mainland Norway, 2 – 10 stations (n=58)	1957 – 1963	Source identification and transport assessment. Good temporal and spatial resolution. No representation of high yield PPG test period (1952, 1954).	$^{234}\text{U}$ , $^{236}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{241}\text{Pu}$ , $^{242}\text{Pu}$ , $^{244}\text{Pu}$	High time resolution chronology. NWT at Northern hemisphere test sites and global fallout, Windscale accident

## 4.2 Sample preparation

Sample preparation in mass spectrometry includes all steps necessary to prepare the analytes which are to be measured in a form suitable for the specific instrument. Some important steps are common for several instruments.

Interfering elements is a common problem in most analytical instruments due to mass or energy overlap. This is usually reduced or eliminated by reducing the presence of the interfering species or elements through proper chemical separation. Interferences cannot always be resolved, and an important example is the energy overlap between  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in alpha-spectrometry; this interference arises from the very similar alpha-energies of the two isotopes, and insufficient energy resolution of the spectrometers. Since the two plutonium isotopes have identical chemical properties it is not possible to separate them prior to analysis, leading to  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  activities being reported as a sum in alpha spectrometry. Computer based spectral deconvolution techniques may be applied to resolve the individual activities of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  analysed by alpha-spectrometry (e.g. Vintró *et al.* (1996); Pollanen *et al.* (2012)), however, this requires that the samples have sufficient activity. For low activity samples, it is still necessary to apply mass spectrometric methods in order to measure the individual activities of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ . Combined methods which samples are first prepared for and analysed by alpha spectrometry, and thereafter re-dissolved or otherwise liberated from alpha-spectrometry planchets and analysed by mass-spectrometric methods provide additional information in the  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratio (e.g. Skipperud (2004); Michel *et al.* (2007); Cizdziel *et al.* (2008)).

Mass spectrometric methods do not suffer from spectral interferences, as these techniques base the separation on mass differences rather than alpha energy differences. The main interference problem for mass spectrometric techniques arises from the presence of species with the same (mass · energy) / charge ratio as the analyte of interest. Molecular interferences as mentioned above could easily be a problem for ICP-MS, and proper measures must be taken to detect and correct for interferences arising from undissociated molecular species.

### *Humic surface soil samples*

Pebbles and conifer cones were removed from the dried samples, and the samples were ground to a fine powder. Aliquots of ~25 g were analysed for  $^{137}\text{Cs}$  by gamma spectrometry. After addition of yield monitor ( $^{242}\text{Pu}$ ) the samples were covered by watch glasses and dry ashed. The ashing temperature was gradually increased to 500 °C to avoid ignition of the samples causing cross contamination and the

formation of refractory species (Vintró and Mitchell, 2006). A temperature of 500 °C was kept for at least 8 h or until the samples were completely ashed. The ashed samples were then leached in aqua regia (3:1 mixture of concentrated HCl and HNO<sub>3</sub>), filtered, and the filtrate was taken to dryness by careful evaporation.

Anion exchange of the samples were performed according to the procedure described by Clacher (1995): the dried sample residue was taken up in 50 – 100 ml 8 M HNO<sub>3</sub>, and Pu was reduced to Pu(III) by addition of FeSO<sub>4</sub> and thereafter oxidised to Pu(IV) by adding NaNO<sub>2</sub>. The samples were then loaded onto pre cleaned DOWEX 1 x 8 anion exchange columns. Pu and Th are selectively bound to the resin, while U, Am and Po pass through. The column was further cleaned by passing 50 – 75 ml 8M HNO<sub>3</sub>. Th was then stripped from the column with 50 ml 9 M HCl and finally Pu was eluted with a 40 ml 1:100 mixture of 50 % HI in 9M HCl. The Pu eluate was evaporated to dryness and re-dissolved in 20 ml 0.5 M HNO<sub>3</sub> for analysis by ICP-MS.

#### *Ice core and air filter samples*

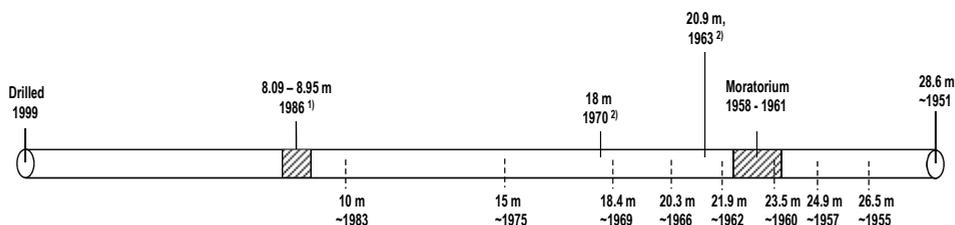
The ice core was cut into sample segments (1.5 – 5 kg) covering different fallout periods, as illustrated in Figure 13. In order to preserve the samples uncontaminated, the cutting was performed in a clean room facility at low temperature (-20 °C). The samples were then melted in room temperature in closed pre-cleaned HDPE bottles, and filtrated with a cut-off of 50 nm (Nucleopore 0.05 µm, Whatman Schleicher&Schuell). The filters were dried and subjected to digital autoradiography as described below, while the filtrates were evaporated to dryness. After completed autoradiography the filters were weighed directly into PTFE ultraclave tubes and yield monitors (~17 pg <sup>233</sup>U and <sup>242</sup>Pu) were added. The ice core filters were digested in 5 ml sub boiled ultrapure HNO<sub>3</sub> (conc.) at high temperature and pressure in an ultraclave unit. The digested filters were recombined with the residues from the dried filtrates, and the sample solutions were diluted to an acid concentration of 8 M.

Most of the air filter were treated as individual samples, and sections corresponding to 110 – 220 m<sup>3</sup> air (1/4 – 1/2 filter) were weighed directly into PTFE ultraclave tubes. Yield monitors (<sup>233</sup>U and <sup>242</sup>Pu, ~17 pg each) were added for quantitative analysis. For the analysis of <sup>241</sup>Pu/<sup>239</sup>Pu, <sup>242</sup>Pu/<sup>239</sup>Pu and <sup>244</sup>Pu/<sup>239</sup>Pu, grouped air filters were selected from ten consecutive days from October 1957 (filters from Bergen) and during spring peak deposition in 1962 and 1963 (filters from Røros). As it was of interest to analyse the <sup>242</sup>Pu/<sup>239</sup>Pu and <sup>244</sup>Pu/<sup>239</sup>Pu atom ratios in the grouped sample, a yield monitor was not added to the latter samples.

Pu and U retained on the filters was leached in sub boil ultrapure HNO<sub>3</sub> (conc.) at high temperature and pressure in an ultraclave unit (UltraCLAVE 3, Milestone Ltd). After leaching, the sample solutions were diluted to an acid concentration of 8 M, and filtered.

Separation of Pu from the sample solution followed the procedure described for humic surface soils above. The U fraction was further purified according to the procedure described by Wilcken (2006): the dried U and Am eluate was dissolved in 15 ml 3 M HNO<sub>3</sub> with 0.5 M Al(NO<sub>3</sub>)<sub>3</sub>, and loaded onto pre-cleaned columns containing 2 ml UTEVA ion-exchange resin. The columns were rinsed with an additional 5 ml 3 M HNO<sub>3</sub>, and converted to the chloride form with 5 ml 9 M HCl. Am and Po was stripped off the columns with 20 ml 5 M HCl with 0.05 M (COOH)<sub>2</sub>. U was eluted with 30 ml 0.01 M HCl.

The Pu and U eluates were evaporated to dryness and taken up in 2 ml concentrated HNO<sub>3</sub> with 2 mg Fe as Fe(NO<sub>3</sub>)<sub>3</sub>, and evaporated to dryness again. The residues were baked at 500 °C overnight. For final target preparation, the Fe / U and Fe / Pu precipitates were pressed into Al sample holders using silver powder as a electric and thermal conductor / binder.



**Figure 13 Illustration of the AUS-99 ice core with sample segments indicated.**

#### **4.2.1 Location of radioactive heterogeneities**

##### *Digital autoradiography*

The filters (ice core filtration membranes and air filters) were mounted on paper and subjected to digital autoradiography. During exposure, the background was reduced by placing the imaging plate (Molecular dynamics, GE Healthcare) and the samples in a protective cassette which was again put in a radiation shielding box of lead. Exposure times of four weeks were routinely used, and the exposed plates were scanned in an image plate scanner (Typhoon 8600, Molecular dynamics) within two hours after termination of the exposure. Indications of radioactive heterogeneities were often encountered, and repeated sample splitting and re-exposures resulted in the isolation of small areas of filter (on the order of a few

mm<sup>2</sup>) which were mounted onto Al stubs with sticky carbon tape and prepared for ESEM-XRMA-analysis.

#### *Environmental scanning electron microscopy with X-ray micro-analysis (ESEM-XRMA)*

Samples isolated by digital autoradiography were analysed by ESEM-XRMA to elaborate the exact position of fallout particles. The samples were run uncoated in the microscope as sample coating may cause interferences in subsequent speciation analysis (Lind, 2006). In order to remediate electrostatic charging of the samples, analyses were performed with the presence of low amounts of atmospheric gas in the sample chamber. To obtain satisfactory specimen penetration depth, counting statistics and detection limits, high acceleration voltages (30 kV) and beam currents (1 – 10 nA) were used.

#### **4.2.2 Determination of concentrations and atom ratios**

##### *Determination of <sup>137</sup>Cs*

Aliquots of the humic surface soil samples, weighed directly into 200 ml NUNC boxes, were measured by low energy (LEGe) gamma spectrometry (Canberra, Meriden CT, USA, relative efficiency 25 % using a purposebuilt geometry setup). The energy resolution (full width at half maximum, FWHM) was 1.76 keV at 1.33 MeV. Counting times ranging between 0.5 and 40 h were used depending on activity to ensure a counting uncertainty below 5 %.

##### *Concentration and atom ratios of Pu isotopes by ICP-MS*

Plutonium atom ratios (<sup>239</sup>Pu, <sup>240</sup>Pu and <sup>242</sup>Pu (yield monitor)) were determined by SF-ICP-MS (Thermo Finnegan Element 2) in low resolution mode ( $R = m / \Delta m = 300$ ). The sample solutions were introduced to the instrument by self-aspiration at a rate of 1.8 ml min<sup>-1</sup>. Concentrations of <sup>239</sup>Pu and <sup>240</sup>Pu were determined through the obtained <sup>239</sup>Pu/<sup>242</sup>Pu and <sup>240</sup>Pu/<sup>242</sup>Pu ratios. Procedural and analytical blanks as well as standard reference materials (IAEA 384, IAEA 300, NIST 4353a) were analysed for method verification.

##### *Determination of Pu and U isotopes by AMS*

The relative abundances of Pu (<sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, <sup>242</sup>Pu and <sup>244</sup>Pu) were determined using the 14 UD tandem accelerator at the Australian National University (ANU), Canberra. Three isotopes (e.g. <sup>239</sup>Pu, <sup>240</sup>Pu and <sup>242</sup>Pu (yield monitor)) were counted sequentially in each set up, and the count rates were normalised against the most abundant isotope (<sup>242</sup>Pu for samples with added yield monitor, otherwise <sup>239</sup>Pu). Counting times varied between 60 and 300 seconds were used, depending on

concentrations. A certified reference material for Pu atom ratios (UKAEA No. UK Pu 5/92138) was measured repeatedly during analyses. The  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratio precision for the reference material was 3.1 % (standard deviation,  $n=13$ ). Detection limits were calculated based on analytical blanks spiked with 17 pg  $^{242}\text{Pu}$ . The detection limits were 4.2 and 1.3 fg for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  respectively. Some samples were prepared without yield monitor in order to analyse  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$  and  $^{244}\text{Pu}$ . For these samples a quantitative detection limit cannot be established. However, blanks without yield monitor run for  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$  and  $^{244}\text{Pu}$  had very low count rates corresponding to 0, 0.48 and 0.1 counts per minutes for the three isotopes, respectively. Due to the low background, blank corrections were not applied to these results.

Similarly to the Pu analysis, three U isotopes ( $^{233}\text{U}$  (yield monitor),  $^{234}\text{U}$  and  $^{236}\text{U}$ ) were measured sequentially, and the concentrations of  $^{234}\text{U}$  and  $^{236}\text{U}$  were established based on the measured  $^{234}\text{U}/^{233}\text{U}$  and  $^{236}\text{U}/^{233}\text{U}$  atom ratios. Analytical blanks spiked with 20 pg  $^{233}\text{U}$  gave 115 and 17.5 counts of  $^{234}\text{U}$  and  $^{236}\text{U}$  for counting times of 0.5 and 3 minutes respectively, corresponding to detection limits of 0.9 pg and 24 fg of  $^{234}\text{U}$  and  $^{236}\text{U}$  respectively. Blank corrections were applied to the results of all U samples.

### 4.3 Methods employed in the present thesis

Radionuclides can be identified and quantified based on radiometric as well as mass spectrometric methods. A range of different methods exhibiting different advantages and drawbacks are available for the purpose. Radiometric methods include  $\alpha$ ,  $\beta$  and  $\gamma$  spectrometry, while mass spectrometric methods include inductively coupled plasma – mass spectrometric methods (ICP-MS), Accelerator Mass Spectrometry (AMS), Thermal Ionisation Mass Spectrometry (TIMS) and Resonance Ionisation Mass Spectrometry (RIMS).

Radiometric methods, e.g. alpha spectrometry offers low detection limits (e.g.  $\sim 1 \times 10^{-14}$  g  $^{239}\text{Pu}$ ). However, long counting times, and insufficient resolution for quantifying  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  individually is a serious drawback (e.g. Hou and Roos (2008) and Skipperud (2004)). Mass spectrometry on the other hand offers more than sufficient resolution, and provides an important tool for source identification, namely the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio.

The rest of the chapter focus on the instrumental techniques used in the present thesis. The isobaric interference from  $^{238}\text{U}$  precludes the determination of  $^{238}\text{Pu}$ . A combined sample preparation for alpha spectrometry and mass spectrometry is frequently used in order to reveal the  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratio and  $^{240}\text{Pu}/^{239}\text{Pu}$

atom ratio in the same sample, e.g. Ketterer *et al.* (2004), Lujanienė *et al.* (2009), Salminen-Paatero *et al.* (2012).

#### 4.3.1 Mass spectrometry

In mass spectrometry, different elements and isotopes of the elements are separated from each other by the mass to charge ratio. A multitude of different mass spectrometers are in use, their general working principle is to accelerate analyte ions to high energies, and separate the different masses by applying magnetic and / or electrostatic fields.

Interferences in mass spectrometry arise from species with the same (mass · energy)/ charge ratio as the analyte of interest. Molecular, poly charged or isobaric interferences may cause problems for precise determination of concentrations and atom ratios. Different strategies are available for the remediation of these interferences. Matrix separation (ion exchange or solvent extraction) resolves a range of interferences, but is labour intensive and time consuming.

##### *Inductively coupled plasma – mass spectrometry (ICP-MS)*

ICP-MS is a versatile and relative inexpensive technique offering good precision and high throughput of samples for measurements of both isotope ratios and concentrations in a range of sample types. Sample introduction systems for the direct volatilisation of solid samples exists, as does ultra-low volume methods, and absolute detection limits in the attogram-range for U and Pu has been reported for the latter (Schaumlöffel *et al.*, 2005).

The analytes are introduced to the instrument as an aerosol or as gas and transported by an argon carrier gas to a hot plasma (6000 - 8000 K) where the sample atoms are ionized to a high degree. The ionised analyte ions are then accelerated and subjected to separation by electrostatic and / or magnetic fields. A variety of mass separation configurations of ICP-MS are in use, including quadrupole, sector field, double focussing sector field and multi-collectors.

ICP-MS is a multi-elemental method, thus complex samples can be introduced and the concentrations of several elements or isotopes of elements can be determined in rapid scans. Short reading times and rapid switching between masses reduce problems associated with signal drift. Consequently, the precision of isotope ratio analysis with ICP-MS is good. Hou and Roos (2008) states precisions in the range 0.1 – 0.5 % for isotope ratio determination using common ICP-MS instruments (double focussing sector field and quadrupole), and even better for multi-collector

ICP-MS instruments. However, isotope ratio precision is ultimately sensitive to the concentrations presented to the instrument.

ICP-MS instruments are inherently more sensitive to interferences and background than AMS-systems (Fifield, 2008), impairing both detection limits and abundance sensitivities. Poor abundance sensitivity is a problem when analysing isotopes with low abundance when there are high abundance isotopes of similar atomic mass present in the sample (e.g.  $^{235}\text{U}$  interfering with  $^{236}\text{U}$ ). Good chemical separation and the reduction of water in the aerosol can resolve some of these problems. However, analysis of  $^{236}\text{U}$  by ICP-MS has an abundance sensitivity of  $^{236}\text{U}/^{238}\text{U}$  around  $1 \times 10^{-7}$  (Boulyga and Heumann, 2006) and is still superseded by AMS in which ratios as low as  $1 \times 10^{-12}$  may be measured (Fifield, 2008).

The primary worry in Pu analysis by ICP-MS is mass interferences due to the presence of  $^{238}\text{U}$ . U may be present in considerable concentrations in the final sample solutions as a result of incomplete separation, or from post separation contamination. Mass overlaps on mass 239 may arise both from the  $^{238}\text{U}^1\text{H}^+$  peak as well as shoulder overlap from the prominent  $^{238}\text{U}$  peak. During the present work, the count rates at mass 239 and 240 was monitored during the analysis of a strong U solution ( $1 \mu\text{g l}^{-1}$ ), and used for establishing a mathematical correction factor based on the  $^{238}\text{U}$  count rates in the samples. This correction was applied to both  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , though the effect was negligible at mass 240.

#### *Accelerator mass spectrometry (AMS)*

AMS is superior to other mass spectrometric techniques in its high sensitivity, high resolution, low detection limits and insensitivity to molecular interferences (Fifield, 2008; Hou and Roos, 2008). Sufficient sample quantities can be obtained through isotope dilution followed by precipitation with silver, e.g. Englund *et al.* (2007) or co-precipitation with iron oxide, e.g. Fifield (2008). The final sample target is prepared by mixing the sample preparates with an appropriate binder / conductor, and pressed into appropriate sample holders.

The principle of the method is similar to ICP-MS; the masses are separated by electric and magnetic fields, and lead to a detector where high concentration analyte atoms are measured as currents, and less abundant analyte atoms are counted individually. The major difference is that in AMS atoms are accelerated to MeV energies as opposed to KeV energies in ICP-MS. The higher energies facilitate the distinction of elements too close in mass to be resolved by ICP-MS.

The ANU AMS system is described essentially as two mass spectrometers separated by a molecule dissociating accelerator stage (Fifield, 2008), and the main

distinctions from other more conventional mass spectrometric methods arises from the high energy obtained in the accelerator and the double mass spectrometer. As described by Fifield (2008), the AMS facility at ANU consists of five main parts (Figure 14):

- The ion source and pre accelerator of -5 kV generating and forcing negative  $\text{PuO}^-$  and  $\text{UO}^-$  molecular ions towards an pre accelerator
- Accelerating the analyte ions through 100 kV in the pre accelerator and directing them towards the positive terminal of the tandem accelerator
- Molecule splitting and ionization by removing electrons forming  $\text{Pu}^+$  and  $\text{U}^+$  ions in a low pressure gas stripper
- Accelerating the positive ions towards the ground potential in the second part of the tandem accelerator. Selection of Pu and U ions with the correct  $mE/q$  ratio
- A detection system sorting out remaining interfering species by mass or energy

$\text{PuO}^-$  and  $\text{UO}^-$  ions are generated in the ion source and pre-accelerated by the negative potential (5 kV) of the ion source and then by a 150 kV pre accelerator before the specific analyte (Pu or U) are inserted into the accelerator by the inflection magnet. The inflection magnet has a resolution of  $M/\Delta M \sim 1000$ , whereas a resolution of 300 would be sufficient to distinguish UO from PuO (Fifield, 2008). Thus, the beam should be sufficiently pure in either U or Pu in the first part of the acceleration. The  $\text{PuO}^-$  or  $\text{UO}^-$  molecules are further accelerated towards the high voltage terminal where a stripping gas ( $\text{O}_2$  or Ar at 0.002 mbar) dissociates the molecules and change the charge by removing electrons. The positive Pu or U ions are then further accelerated towards the ground terminal of the tandem accelerator.  $\text{Pu}^{5+}$  ions are selected in the analysing magnet and are passed through a velocity filter (Wien filter). The detector used depends on the species being analysed. For Pu potential interfering elements which may reach the detector have significantly lower momentum than Pu and are easily distinguished by an ionization chamber detector. For  $^{236}\text{U}$ , on the other hand, the high abundance of natural U isotopes ( $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$ ) necessitates the employment of a time-of-flight (TOF) detector in addition to the ionisation chamber detector (Fifield, 2008).

Generally the AMS detection is vulnerable to any species which, after the stripping and the second part of the acceleration, have obtained the same  $ME/q^2$  as the Pu or U isotope analysed. This may be the case for  $^{238}\text{U}^{17}\text{O}^-$  and  $^{238}\text{U}^{18}\text{O}^-$  formed in the ion source and which is stripped off their O-atoms early in the acceleration process, interfering with  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , these are probably the most prevalent interfering species in Pu analyses, and the situation is impaired by insufficient separation chemistry prior to analysis as well insufficient vacuum in the early accelerator stages (Fifield, 2008). Poly-atomic molecules of lighter elements (e.g.  $^{191}\text{Pt}^{16}\text{O}_4$ ,

$^{191}\text{Pt}^{4+}$ ,  $^{143}\text{Sm}^{3+}$ ,  $^{96}\text{Mo}^{2+}$  and  $^{48}\text{Ti}^{+}$ ) may also cause interference. On the other hand, the probability of any of these to reach the detector is low given their low natural abundance and the complexity of the initial poly-atomic molecules (Fifield, 2008)

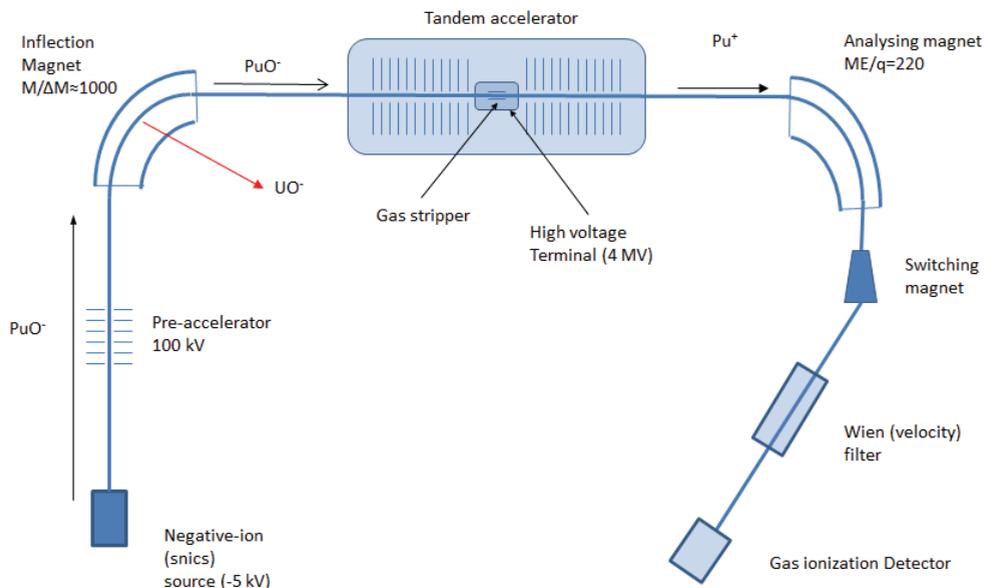


Figure 14. The ANU 14 UD accelerator as inferred from (Fifield, 2008) and (Winkler, 2007).

### 4.3.2 Environmental Scanning Electron microscope (ESEM)

A scanning electron microscope operates under the principle of a focussed electron beam interacting with the sample. The electron beam penetrates the surface of the sample, generating a semi spherical or pear shaped interaction volume, depending on specimen density and accelerating voltage of the incident beam. As a result of the interactions between the electron beam and the sample, several different signal types are generated. Elastic or inelastic scattering of incident electrons causes the emission of backscattered and secondary electrons respectively. Energy deposited by inelastic scattering causes excitation of sample atoms, and later emission of auger electrons or characteristic x-rays (Postek *et al.*, 1980). Backscattered and secondary electrons serve to reveal the topography of the sample, while characteristic x-rays can be used for qualitative or semiquantitative analysis of sample composition.

The backscattered electrons from the sample are particularly useful in the search for actinides as backscattering increases proportionally with atomic number of the sample atoms. A sample with high atomic number will thus appear brighter in the

microscope than a sample of low atomic number, and it is recognized that two elements differing in atomic number by more than three can be distinguished from each other in the electron microscope using BEI mode (Postek *et al.*, 1980). Problems arise when particles are smaller than the resolution of the electron microscope and when high atomic number elements appear with low concentrations embedded in particles of lighter elements. Electron microscope run on well prepared samples should be able to give sub-micrometre resolution (Lind, 2006). However, this requires the instrument to be run under high vacuum. The presence of even minute concentration of air impairs the resolution of the microscope by diffusing the beam. Furthermore, the penetration depth into the sample is low, and the method has relatively high detection limits.

Normally samples prepared for SEM are coated with a material with good electrical conductivity in order to carry away charging effects brought upon the sample by the electron beam. For the case of particle characterisation, sample coating is unfavourable since the coating may interact with later speciation analysis of the particles (Lind, 2006). To overcome the problem of electrostatic charging of the specimen the examination is carried out with a slightly higher pressure in the sample chamber of the electron microscope. Furthermore, beam currents and accelerating voltages were kept high in order to reduce detection limits and increase electron beam penetration depth in the sample (Lind, 2006). Both the presence of air in the sample chamber and the high beam current impairs resolution to such a degree that particles below  $\sim 2 \mu\text{m}$  cannot be distinguished.

#### *X-ray micro analysis (XRMA)*

X-Ray micro-analysis allows for the semi quantitative determination of individual elements in a sample on a microscopic level. An electron beam excites sample atoms by removing electrons from the atom; upon de-excitation characteristic x-rays are emitted (Parrish and Mantler; Postek *et al.*, 1980). The penetration depth is low, on the order of  $1 \mu\text{m}$  for electron beam excitation (Parrish and Mantler), consequently detection of inclusions of Pu or U trapped behind other particles or as inclusions in larger particles could prove difficult. During the present work, ESEM-XRMA served the purpose of examining hotspots identified by digital autoradiography. Substantial effort was put into locating and identifying actinide carrying particles for further examination by  $\mu$ -x-ray fluorescence, however these attempts were futile due to small particle size or a complex matrix.

#### *High intensity micro x-ray techniques (synchrotron)*

The intense, well collimated and polarised x-ray radiation produced in synchrotron facilities used permits non-destructive analysis of elemental composition, structure

and speciation. As opposed to XRMA, the penetration depth of the beam is large, permitting the investigation of internal composition and structure (Lind, 2006). The method has been successfully applied to characterize U and Pu containing particles (Salbu *et al.*, 2001). In the present work the method was tested for radioactive particles contained in the air filters, however it was not found to be useful, likely due to low concentrations.

#### **4.3.3 Digital autoradiography**

Digital autoradiography has got several advantages over conventional film based autoradiography and track-etch techniques in offering high sensitivity and a large dynamic range. The primary detector (image plate) is composed of a highly photostimulable phosphor supported by thin plastic sheet, and in most cases protected by a thin protective layer (Lind, 2006). During exposure the sample is brought in close contact with the imaging plate, and placed within a protective box. Detection limits on the order of 0.002 Bq has been reported for a  $^{238}\text{Pu}$  particle in an autoradiography system similar to the one used in the present work (Zeissler, 1997). The appliance of shielding as described by (Mori *et al.*, 1996) reduces the background and hence the detection limit. During the exposure, the phosphor ( $\text{BaFBr: Eu}^{2+}$ ) captures and stores the irradiated energy until scanned by an image plate scanner. The energy is then released as luminescence, which is detected by the instrument (Lind, 2006).

#### **4.3.4 Atmospheric dispersion modelling**

In this work an atmospheric dispersion model (HYSPLIT) has been used to model releases from selected atmospheric and underground detonations at Novaya Zemlya and Semipalatinsk. Hysplit is a Lagrangian dispersion model in which transport and dispersion of particles or puffs of air are simulated based on forecast or historical meteorological data. The model encompasses simple trajectory calculations, as well as more complex dispersion and deposition simulations with puffs or particles or a combination of the two as an input term for the simulation (Draxler and Hess, 1997). Transport and deposition calculations are performed in a Lagrangian framework, while concentrations are calculated within a fixed grid (Eulerian).

Hysplit has not been designed specifically for modelling nuclear detonations, and does not include calculations of cloud stabilisation height and particle distributions. Thus, this information must be acquired from other sources and included in the model prior to the simulations. Furthermore, the model is sensitive to the quality of the meteorological data forming the basis of the simulations. Available archived

meteorological data improved after the international geophysics year in 1957 – 1958, and this applies in particular to data released from the former Soviet Union.

Geographical location, release heights and particle sizes are factors that have significant influence on transport times and deposition times and locations (Pöllänen *et al.*, 2006; J. Bartnicki, Pers. Comm.). The starting points of the simulations may be one or several geographical points separated by horizontal and / or vertical distance, and several classes of particles with individual densities, size and gravitational settling may be defined into the model alongside gaseous releases.

The removal of radioactive debris on the time scales simulated in this work is assumed to take place through two processes: wet and dry deposition. Large particles are affected by gravity and deposit rapidly after the detonation, whereas smaller particles and gaseous debris have considerable residence times in the troposphere and are subject to transport over large distances. Wet deposition is assumed to be the most efficient deposition mechanism for these particles.

## 5 Summary of findings

### Paper I - Levels and trends of deposition and concentrations of Pu and Cs in Humic surface soils

To differentiate between sources contributing to Pu contamination, Pu concentration and atom ratios were determined in humic surface soil samples from 45 geographically well distributed sites across the Norwegian mainland using SF-ICP-MS and AMS. The  $^{239+240}\text{Pu}$  activity concentrations ranged within 4 – 149 Bq  $\text{m}^{-2}$  (1990) and 0.7 – 63 Bq  $\text{m}^{-2}$  (2005), with the higher concentrations predominantly found at coastal locations. To identify any migration of Pu from the upper 5-7 cm of the surface soils, Pu activity concentrations in samples collected at the same sites in 1990 and 2005 were compared. On average, about 35 % of the  $^{239+240}\text{Pu}$  activity concentrations in the 1990 samples remained in the humic soil samples collected in 2005. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios ranged within 0.164 to 0.211 (1990) and 0.161 to 0.195 (2005), predominantly within the range of global fallout. Results based on the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio, as well as levels of heavier Pu isotopes ( $^{241}\text{Pu}$  and  $^{242}\text{Pu}$ ) indicated a slight (8 to 13 %) contribution of Pu from the Chernobyl accident in some inland areas of Norway.

### Paper II - Chronology of Pu isotopes and $^{236}\text{U}$ in an arctic ice core

In the present work, state of the art isotopic fingerprinting techniques are applied to an Arctic ice core in order to quantify deposition of U and Pu, and to identify possible tropospheric transport of debris from former Soviet Union test sites Semipalatinsk (Central Asia) and Novaya Zemlya (Arctic Ocean). An ice core chronology of  $^{236}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{240}\text{Pu}$  concentrations, and atom ratios, measured by accelerator mass spectrometry in a 28.6 m deep ice core from the Austfonna glacier at Nordaustlandet, Svalbard is presented. The ice core chronology corresponds to the period 1949 to 1999. The main sources of Pu and  $^{236}\text{U}$  contamination in the Arctic were the atmospheric nuclear detonations in the period 1945 to 1980, as global fallout, and tropospheric fallout from the former Soviet Union test sites Novaya Zemlya and Semipalatinsk. Activity concentrations of  $^{239+240}\text{Pu}$  ranged from 0.008 to 0.254 mBq  $\text{cm}^{-2}$  and  $^{236}\text{U}$  from 0.0039 to 0.053  $\mu\text{Bq cm}^{-2}$ . Concentrations varied in concordance with  $^{137}\text{Cs}$  concentrations in the same ice core. In contrast to previous published results, the concentrations of Pu and  $^{236}\text{U}$  were found to be higher at depths corresponding to the pre-moratorium period (1949 to 1959) than to the post-moratorium period (1961 and 1962). The  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio ranged from 0.15 to 0.19, and  $^{236}\text{U}/^{239}\text{Pu}$  ranged from 0.18 to 1.4.

The Pu atom ratios ranged within the limits of global fallout in the most intensive period of nuclear atmospheric testing (1952 to 1962). To the best knowledge of the authors the present work is the first publication on biogeochemical cycles with respect to  $^{236}\text{U}$  concentrations and  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios in the Arctic and in ice cores.

### **Paper III - Long-range tropospheric transport of uranium and plutonium weapons fallout from Semipalatinsk nuclear test site to Norway**

A combination of state-of-the-art isotopic fingerprinting techniques and atmospheric transport modelling using real-time historical meteorological data has been used to demonstrate direct tropospheric transport of radioactive debris from specific nuclear detonations at the Semipalatinsk test site in Kazakhstan to Norway via large areas of Europe. A selection of archived air filters collected at ground level at 9 stations in Norway during the most intensive atmospheric nuclear weapon testing periods (1957–1958 and 1961–1962) has been screened for radioactive particles and analysed with respect to the concentrations and atom ratios of plutonium (Pu) and uranium (U) using accelerator mass spectrometry (AMS). Digital autoradiography screening demonstrated the presence of radioactive particles in the filters. Concentrations of  $^{236}\text{U}$  ( $0.17\text{--}23\text{ nBq m}^{-3}$ ) and  $^{239+240}\text{Pu}$  ( $1.3\text{--}782\text{ }\mu\text{Bq m}^{-3}$ ) as well as the atom ratios  $^{240}\text{Pu}/^{239}\text{Pu}$  ( $0.0517\text{--}0.237$ ) and  $^{236}\text{U}/^{239}\text{Pu}$  ( $0.0188\text{--}0.7$ ) varied widely indicating several different sources. Filter samples from autumn and winter tended to have lower atom ratios than those sampled in spring and summer, and this likely reflects a tropospheric influence in months with little stratospheric fallout. Very high  $^{236}\text{U}$ ,  $^{239+240}\text{Pu}$  and gross beta activity concentrations, as well as low  $^{240}\text{Pu}/^{239}\text{Pu}$  ( $0.0517\text{--}0.077$ ),  $^{241}\text{Pu}/^{239}\text{Pu}$  ( $0.00025\text{--}0.00062$ ) and  $^{236}\text{U}/^{239}\text{Pu}$  ( $0.0188\text{--}0.046$ ) atom ratios, characteristic of close-in and tropospheric fallout, were observed in filters collected at all stations in Nov 1962, 7–12 days after three low-yield detonations at Semipalatinsk (Kazakhstan). Atmospheric transport modelling (NOAA HYSPLIT\_4) using real-time meteorological data confirmed that long range transport of radionuclides, and possibly radioactive particles, from Semipalatinsk to Norway during this period was plausible. The present work shows that direct tropospheric transport of fallout from atmospheric nuclear detonations periodically may have had much larger influence on radionuclide air concentrations and deposition than previously anticipated.

## 6 Results and discussion

In the present work, concentrations, atom- and activity ratios of U, Pu and  $^{137}\text{Cs}$  have been determined using several chemical and instrumental techniques. In order to assure the quality of the obtained data, blanks (analytical and procedural), standard reference materials and in-house standards have been processed and analysed along with the samples and are presented in

Table 4. Standard reference materials and in house standards represent common samples for ICP-MS and AMS and permit the intercomparison of the results from the two instruments (Figure 15).

### *Detection limits*

Instrumental and procedural detection limits in the present work are based on repeated measurements of analytical and procedural blanks. Detection limits for both instruments were based on the analysis of procedural blanks and are presented in Table 3. ICP-MS analysis suffers higher and more variable background than AMS analysis. The detection limits for ICP-MS was calculated as the average intensities for the blank samples + 3  $\sigma$ . For AMS analysis, background counts were low, and the detection limit was calculated as the blank count rates (cps) on the respective mass.

**Table 3. Detection limits (g) based on repeated analysis of blank samples.**

	$^{234}\text{U}$	$^{236}\text{U}$	$^{239}\text{Pu}$	$^{240}\text{Pu}$
ICP-MS	-	-	$2.65 \times 10^{-13}$	$7.45 \times 10^{-14}$
AMS	$9.00 \times 10^{-12}$	$2.40 \times 10^{-14}$	$4.20 \times 10^{-15}$	$1.30 \times 10^{-15}$

### *Accuracy*

Repeated analyses of Pu concentrations and atom ratios in certified reference materials and in-house standards were performed throughout the course of the present work for quality assurance and quality control; the results are presented in Table 4.

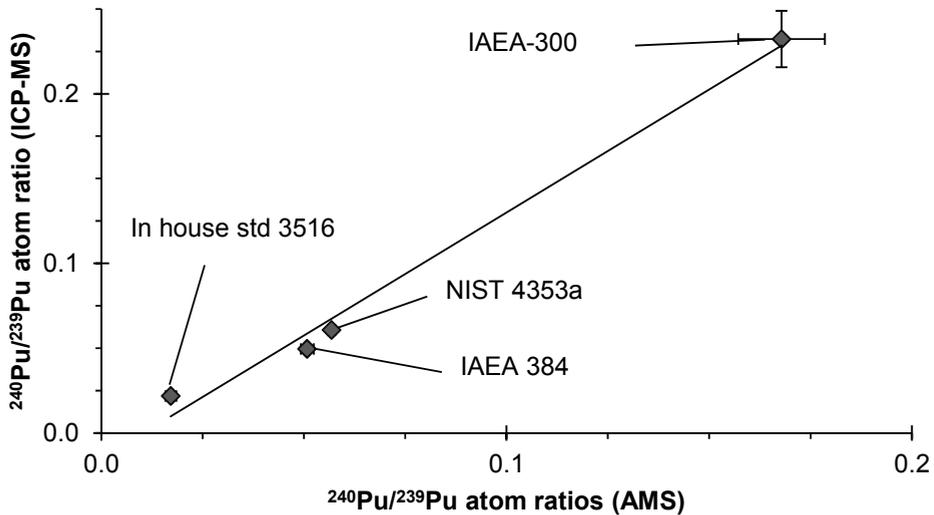
**Table 4. Concentrations and atom ratios of plutonium in certified reference materials and in house standards analysed in the present work. The UKAEA Pu 5/92138 has certified  $^{239}\text{Pu}/^{240}\text{Pu}$ : $^{242}\text{Pu}$  atom ratios close to unity and is used for instrumental calibration, for this reason it is analysed without yield monitor addition. Consequently, concentrations are unavailable for this material. Certified or recommended values from reference sheet when available, literature values included for comparison.**

Reference material	$^{239}\text{Pu}$ conc. pg g <sup>-1</sup>	$^{240}\text{Pu}$ conc. pg g <sup>-1</sup>	$^{239+240}\text{Pu}$ Bq kg <sup>-1</sup>	$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio	Reference
IAEA 384 (Fangataufa sediment)	46 ± 6 42 ± 4* 41.2 ± 0.3	2.2 ± 0.5 2.1 ± 0.2* 2.06 ± 0.01	125 ± 18 107 ± 2 112.02 ± 0.74	0.0501 ± 0.0008 0.048 ± 0.006 0.0501 ± 0.0005*	This work (n=5) Povinec <i>et al.</i> (2007) Godoy <i>et al.</i> (2009)
NIST 4353 (Rocky flats soil)	4.9 ± 0.3 6.1 ± 0.7** 3.97 ± 0.02 2.1 ± 0.1	0.29 ± 0.01 0.34 ± 0.04** 0.218 ± 0.001 0.14 ± 0.02	13.8 ± 0.7 16.8 ± 1.8 10.65 ± 0.04 5.88 ± 0.28	0.058 ± 0.002 0.053 – 0.060** 0.055 ± 0.001 0.065 ± 0.009	This work (n=3) Reference sheet Cizdziel <i>et al.</i> (2008) Child <i>et al.</i> (2008)
IAEA 300 (Baltic sea sediment)	1.17 ± 0.04 - 1.05 ± 0.04 0.97 ± 0.06	0.20 ± 0.01 - 0.18 ± 0.01 0.19 ± 0.01	4 ± 1 3.44 – 3.65 3.9 ± 0.1 3.8 ± 0.2*†	0.22 ± 0.03 - 0.17 ± 0.01 0.19 ± 0.003	This work (n=4) Reference sheet Child <i>et al.</i> (2008) Nygren <i>et al.</i> (2003)
in-house standard 2626 (sediment)	-	-	-	0.127 ± 0.004 0.115 ± 0.007	This work (ICP-MS) (n=3) L. Skipperud (Pers. Comm).
in-house standard 3516 (soil)	-	-	-	0.019 ± 0.003 0.0194±0.0008	This work ICP-MS and AMS (n=5) L. Skipperud (Pers. Comm).
UKAEA Pu5/92138	-	-	-	0.97 ± 0.014 0.966 ± 0.001	This work (n=16) Reference sheet

\* - information / uncertified value, \*\* - Calculated from reference sheet, † - Nygren *et al.* (2003) reports considerable inhomogeneities in the IAEA-300 reference material affecting both Pu concentrations and atom ratios.

### Precision

A certified reference material for Pu atom ratios (UKAEA Pu5/92138) was analysed repeatedly during AMS-analysis in the present work. Atom ratio precisions of 2.5, 2.7 and 1.4 % was calculated for  $^{240}\text{Pu}/^{239}\text{Pu}$ ,  $^{239}\text{Pu}/^{242}\text{Pu}$  and  $^{240}\text{Pu}/^{242}\text{Pu}$ , respectively (relative standard deviation,  $n=16$ ). The precisions for  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios, determined by SF-ICP-MS in repeated measurements of reference materials IAEA 300, IAEA284 and in house standards 2626 (sediment) and 2516 (soil) was below 7 % RSD (0.4 – 7 %).



**Figure 15.** Comparison of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in standard reference materials and in house standards (IAEA300, IAEA384, NIST4353a and in-house standard 3516) analysed by AMS and SF-ICP-MS.

Activity concentrations of  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  ratios were determined in selected humic surface soil samples. The activity concentrations were well correlated ( $R^2=0.97$   $n=25$ ) between the two instruments, as shown in Figure 16.

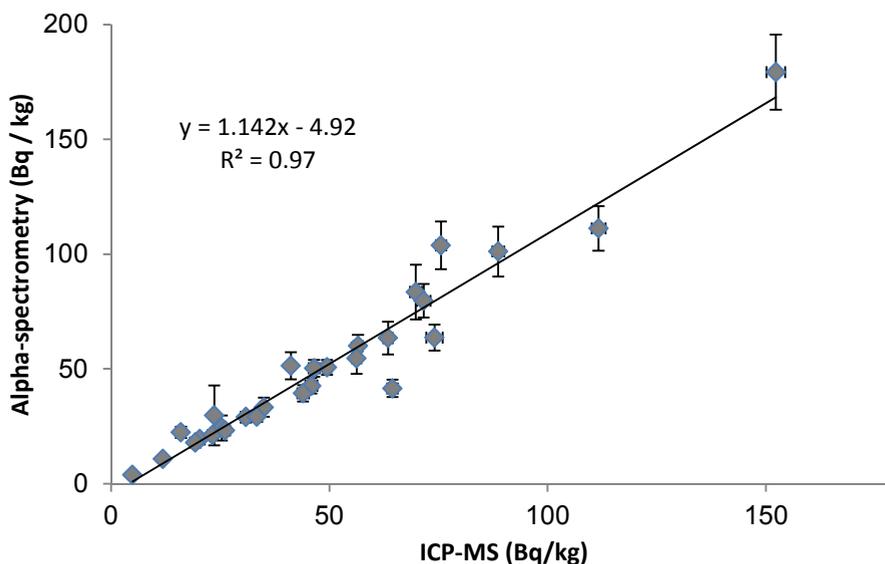


Figure 16. The correlation between concentrations of  $^{239+240}\text{Pu}$  determined in humic surface soil samples with ICP-MS and alpha-spectrometry respectively.

## 6.1 Plutonium deposition in the terrestrial environment of Norway

### 6.1.1 Concentrations of Pu in the terrestrial environment

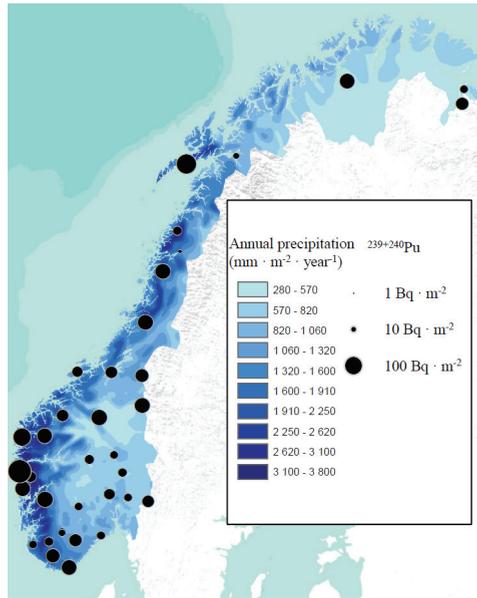
Deposition of  $^{239+240}\text{Pu}$  in the humic surface soils (Paper I) were found within the ranges 4 – 149  $\text{Bq m}^{-2}$  (1990) and 0.63 – 63  $\text{Bq m}^{-2}$  (2005). The  $^{239+240}\text{Pu}$  activity concentrations were largely within the range of 14 – 135  $\text{Bq m}^{-2}$  for soil samples collected at similar latitudes in 1970 (Hardy *et al.*, 1973; Kelley *et al.*, 1999). Thus a substantial proportion of the Pu deposition during the main period of deposition (1945 - 1963) still seems to be present within the upper 7 cm of humic surface soil layers in Norway.

The calculated integrated deposition in the Austfonna ice core was  $8.2 \pm 0.1 \text{ Bq m}^{-2}$  for  $^{239+240}\text{Pu}$  and  $0.151 \pm 0.005 \text{ mBq m}^{-2}$  for  $^{236}\text{U}$ . The  $^{239+240}\text{Pu}$  values were in accordance with the value stated by Hardy *et al.* (1973) (average value for latitudes 78 – 80 and 80 – 90 °N).

### 6.1.2 Geographical distribution of Pu

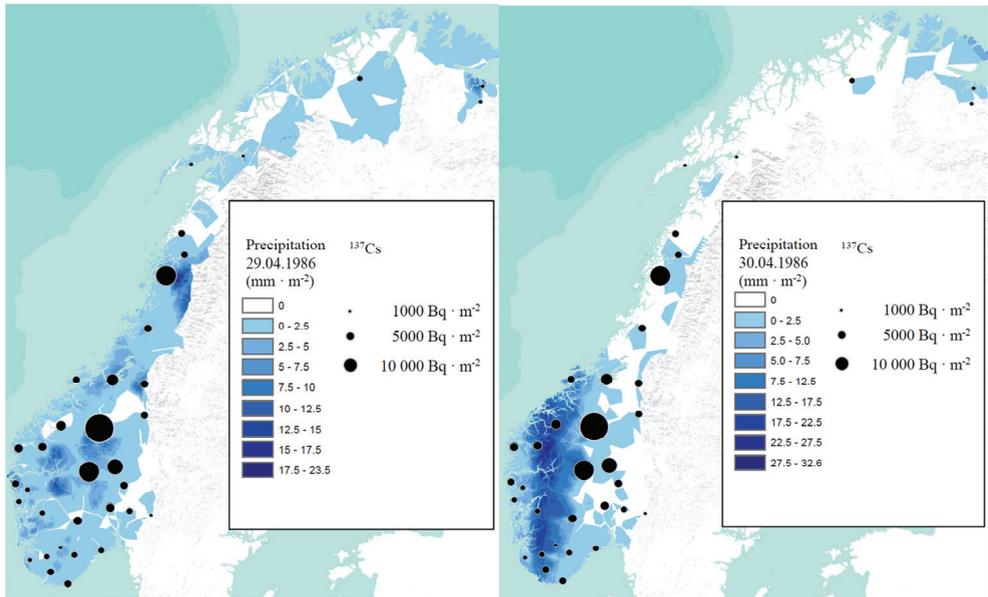
The highest concentrations of  $^{239+240}\text{Pu}$  in the humic surface soil samples were found at coastal sites with high annual precipitation (Figure 17 and Paper I). Significant variations were observed between sites with similar precipitation levels. Orographic precipitation (Storebø, 1959; Storebø, 1968) and washout due to precipitation rates and chemistry (Steinnes and Njastad, 1993) may explain some of

this variation. Similarly, the lowest activity concentrations were found at inland locations with low annual precipitation. This is generally in agreement with the global fallout theory. However, a good correlation between precipitation and activity concentrations could not be established. This may be attributed to soil chemistry, precipitation chemistry and precipitation rates in the years following the initial deposition.



**Figure 17. Activity concentrations of  $^{239+240}\text{Pu}$  in humic surface soils in Norway (Paper I) and annual precipitation levels.**

In contrast to Pu, the highest concentrations of  $^{137}\text{Cs}$  were found at inland locations associated with high precipitation in the first few days after the Chernobyl accident (Figure 18). The deposition of  $^{137}\text{Cs}$  from the Chernobyl accident depended heavily on the location of the plume and the precipitation pattern in the days after the accident. Thus heterogeneous distributions should be expected. The main deposition of debris from the Chernobyl accident in Norway originated from the releases during the initial explosion the 26 March 1986. This debris reached central Scandinavia and Norway the 29 and 30 April 1986 (Saltbones, 1986), elevated concentrations of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  in air were observed until 9 May 1986 (Sæbø *et al.*, 1998). Furthermore, radioactive particles containing Chernobyl origin Zr, Ru, I, Cs, Ba and Ce isotopes were observed in rain water collected in Oslo on 6 May 1986 and in environmental samples collected in 1987 (Salbu, 1988a; Salbu, 1988b).



**Figure 18.** Measured activity concentrations of  $^{137}\text{Cs}$ , samples collected in 1990 in Norway (Paper I) plotted along levels of precipitation in the days following the Chernobyl accident (McInnes).

### 6.1.3 Trends in deposition of Pu

Concentrations of  $^{239+240}\text{Pu}$  and  $^{236}\text{U}$  in the Austfonna ice core ranged within  $0.008 - 0.254 \text{ mBq cm}^{-2}$  and  $0.0039 - 0.053 \text{ } \mu\text{Bq cm}^{-2}$ , respectively (paper II). The concentrations were found to be higher at depths corresponding to the pre-moratorium period 1956 – 1959 than the post-moratorium period (cf. Figure 19). This is in contrast to previously published values (e.g. Warneke *et al.* (2002), Koide *et al.* (1985), UNSCEAR (2000a) and Olivier *et al.* (2003)), in which the highest deposition of radionuclides were reported to have taken place in the period 1962–1964. The Pu and  $^{236}\text{U}$  deposition profile is supported by  $^{137}\text{Cs}$  deposition profiles in the same core and several other ice cores from the same glacier (Pinglot *et al.*, 2003). A few possible reasons for this anomaly will be presented below.

- Variations in the ice core accumulation rates, reflected in annual precipitation or melting. This could cause  $^{239+240}\text{Pu}$  and  $^{236}\text{U}$  concentrations to be higher in those segments of the core corresponding to 1955 – 1958 than in the segments corresponding to 1959 - 1962 and 1962 – 1962 without reflecting variations in actinide deposition.
- Fluctuations in the south – north tropospheric transport in combination with a higher number of detonations at Semipalatinsk and tropospheric debris from US equatorial detonations. Several papers have addressed long range transport of

pollutants from Eurasia and north America reaching the Arctic, particularly during winter, e.g. Barrie and Hoff (1984), Barrie (1986), Stohl (2006) and (Warneke *et al.*, 2010). According to Barrie (1986), the arctic winter air mass extends as far south as 40 °N over continental Eurasia and a persistent high pressure area forces air pollutants from Central Europe and Eurasia towards the Arctic. Tropospheric transport towards the Arctic is enhanced during positive North Atlantic Oscillation phases (Eckhardt *et al.*, 2003).

- A post deposition redistribution of radionuclides embedded glacier ice may arise from infiltration and percolation of meltwater. Tritium ( $^3\text{H}$ ), which is used to establish the 1963 maximum deposition in the ice core is bound in water molecules, and is likely to behave more conservatively in the ice profile than debris particles. This could cause Pu and  $^{236}\text{U}$  to be depleted in layers corresponding to 1963, and enhanced in deeper layers, leaving  $^3\text{H}$  relatively unaffected.

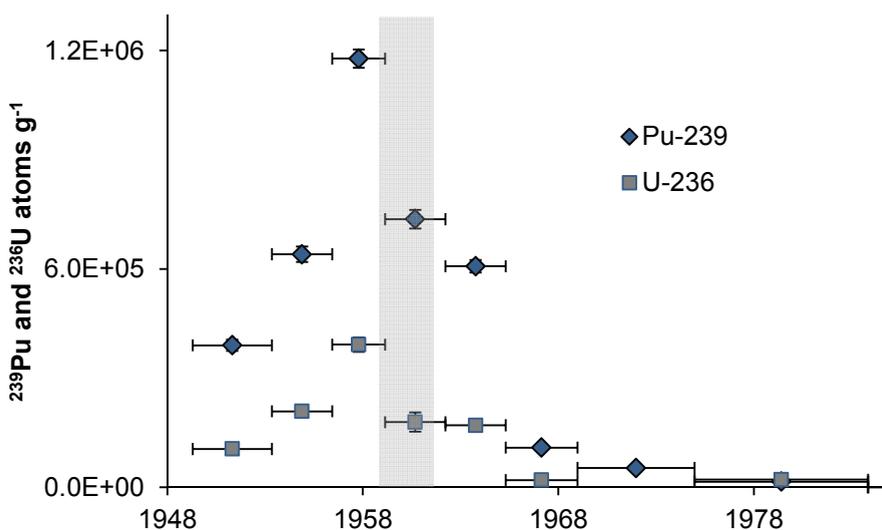


Figure 19. Concentrations of  $^{239}\text{Pu}$  and  $^{236}\text{U}$  (atoms  $\text{kg}^{-1}$ ) in the Austfonna ice core. The shaded grey box indicates the moratorium period (November 1958 – September 1961).

#### 6.1.4 Wash-out (humic surface soil samples)

By comparing the  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$  concentrations in the upper 7 cm of humic surface soil samples collected in 1990 with those from 2005, the average retention of  $^{239+240}\text{Pu}$  was found to be 35 %, ranging within 2 and 78 % when obvious outliers had been removed from the dataset (appendix 2). The outliers included samples with identical or higher activity concentrations in 2005 than in 1990, and 4

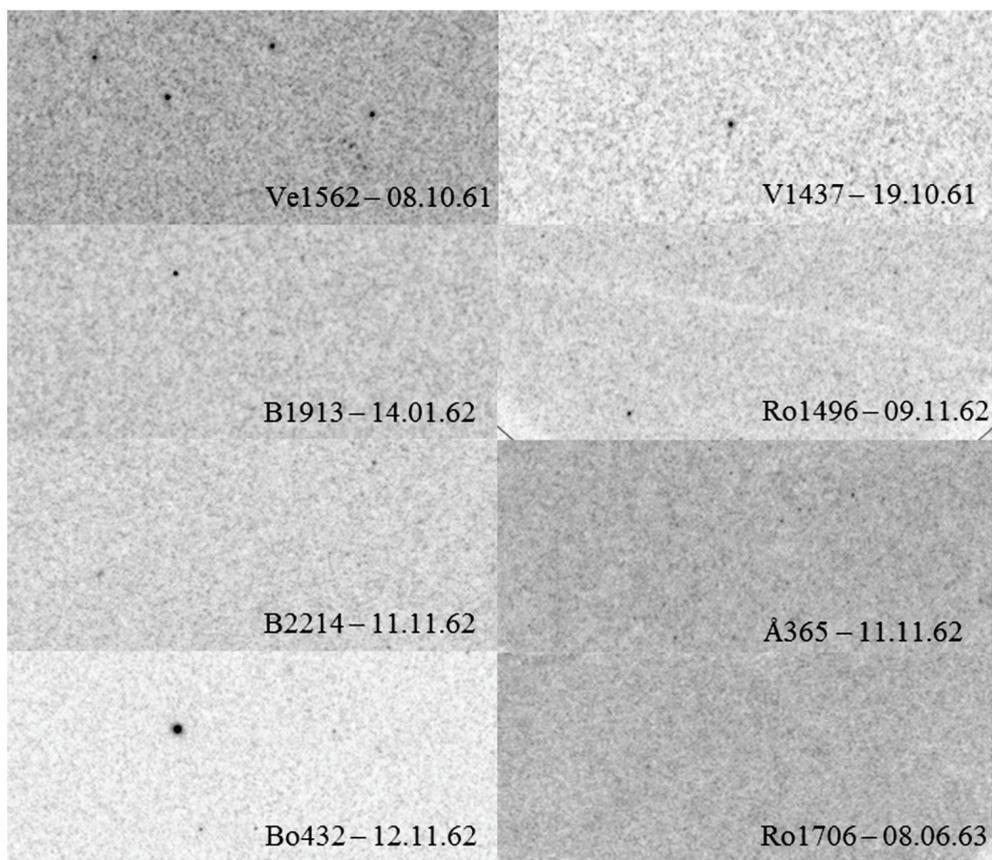
samples where the humic layer in 2005 had an insufficient depth. Similarly, the average retention of  $^{137}\text{Cs}$  in the samples was 35 %, and ranging within 9.2 and 92%.

The long-term bioavailability and mobility of radionuclides released to the environment depends primarily on the release scenario, speciation and interactions between the radionuclides and environmental materials after deposition. Depending on the chemical conditions in the recipient medium, the mobility of the deposited radionuclides may change over time. Weathering and bio-erosion increase the mobility of the deposited radionuclides, while sorption, chemisorption and incorporation into mineral lattices decrease the mobility (Salbu, 2000b). Both Pu and Cs are assumed to be relatively immobile in organic rich and mineral soils and sediments (Skipperud, 2004; Qiao *et al.*, 2012).

## **6.2 Radioactive particles in surface air and ice core samples**

Results from digital autoradiography of the air filters (Paper III) and the AUS-99 ice core (Paper II) indicated the presence of radioactive particles in samples from periods with atmospheric nuclear testing at the FSU test sites Novaya Zemlya and Semipalatinsk. In periods without atmospheric nuclear testing radioactive few particles could be identified, despite high concentrations of actinides in the filters (Paper III and Appendix 1). Since refractory radionuclides released from atmospheric nuclear detonations are assumed to form particles (Heft, 1970; Salbu, 2008), it is likely that these filters contain particles too deeply embedded in the filter material to be detected by digital autoradiography. Filters with high activities (gross beta, Pu, U,) were usually darker than the background in the autoradiographs, even in the absence of hotspots.

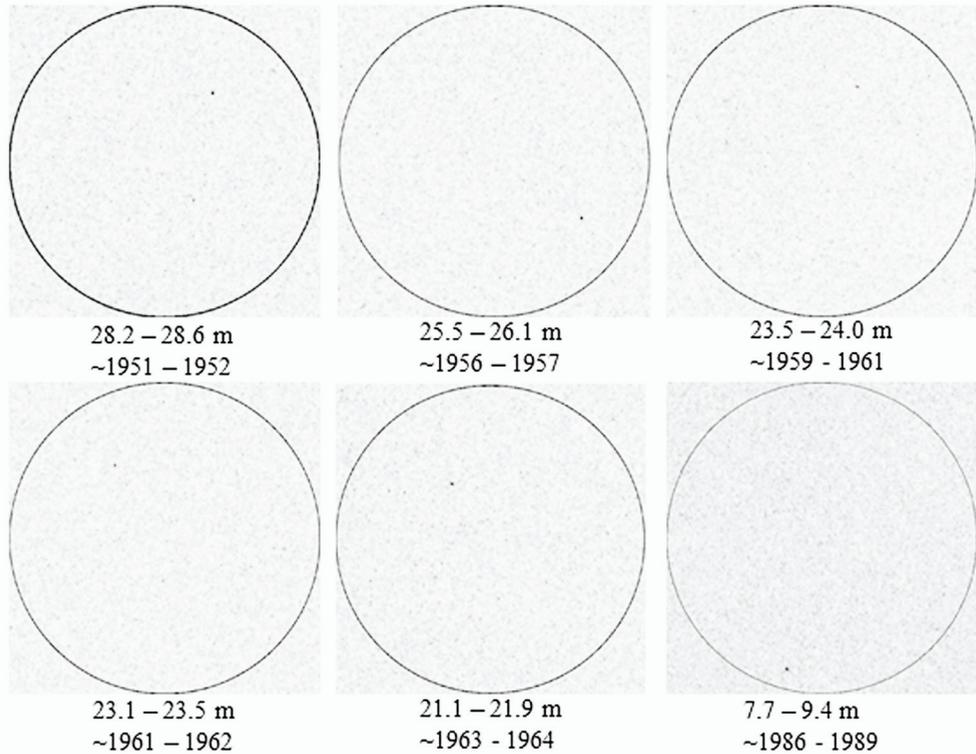
Fallout particles as large as 20  $\mu\text{m}$  have been reported in fresh fallout. However, large particles are assumed to be rapidly removed from the atmosphere by gravitational settling and particle diameters larger than 6  $\mu\text{m}$  are seldom found in debris transported over longer distances, e.g. Mamuro *et al.* (1962), Sisefsky (1966), and Sisefsky (1966). Stratospheric radioactive debris particles tended to be larger in periods with atmospheric testing than in periods without, e.g. Chagnon and Junge (1965), Feely *et al.* (1966), Telegadas and List (1969). Figure 20 shows autoradiographs of a selection of air filters collected in the period 1961 – 1963.



**Figure 20. Autoradiographs of selected air filters with radioactive heterogeneities. The filters with highest associated Pu and U concentrations were from November 1962. Hotspots indicating the presence of particles can be seen in the filters from the autumn and winter of 1961 and 1962, but were absent or ambiguous in filters associated with spring peak deposition as illustrated by the autoradiograph of the filter from Røros, the 8 June 1963 (lower right). Location codes: B – Bergen, Ro – Røros, V – Vadsø, Ve – Værnes and Å – Ålesund.**

A selection of digital autoradiographs of ice core filters is shown in Figure 21. These indicate the presence of low activity particles at depth ranges corresponding to the atmospheric nuclear testing periods at Novaya Zemlya and Semipalatinsk (1951 – 1962). Radioactive heterogeneities were also seen at depths corresponding to the time period 1986 – 1989. These particles may originate from the Chernobyl accident in 1986 or a vented underground detonation at Novaya Zemlya in 1987. The vented underground detonation at Novaya Zemlya is further described in Mikhailov (2004) as a situation in which highly radioactive gases escaped through fissures and into the open atmosphere and lingered over the area for several days. Both volatiles and refractory nuclides escaped; the escaped gases were detected as far south as Risø in Denmark (Bjurman *et al.*, 1990).

Radioactive particles were frequently sampled and characterised during the atmospheric nuclear testing periods e.g. Sisefsky (1961), Mamuro *et al.* (1962), Edvarson and Sisefsky (1966), Sisefsky (1966). However, to the author's best knowledge this is the first time fallout particles from atmospheric nuclear weapon tests have been localised in ice core samples.



**Figure 21. Digital autoradiographs of ice core filters with radioactive heterogeneities. The size of the filters are indicated by the outer circle ( $\phi=47$  mm) whereas autoradiography hotspots indicating the presence of radioactive particles are inscribed in smaller circles.**

Attempts were made to isolate and characterise individual particles using ESEM-XRMA and the x-ray microprobe, beamline L, HASYLAB. However, the Pu and U signals could not be observed, probably due to the complex sample matrix (cellulose asbestos filters), and that concentrations of Pu and U in the particles were below the detection limits for these methods. Fallout particles have been reported to consist mainly of Fe, Al, Ca, Si and Pb, i.e., materials associated with bomb casing, tamper and environmental materials, (Sisefsky, 1964; Crocker *et al.*, 1966; Kemmochi, 1966). Thus, the relative concentration of actinides in particles may prove too low to be directly detected by ESEM-XRMA.

### 6.3 Atom and activity ratios

Source identification of radioactive contamination is important in order to reveal transport routes and transit times of pollution as well as predicting the extent of future accidental or operational releases. In the present work Pu atom and activity ratios were utilised in combination with atmospheric dispersion modelling for source identification and source assessment purposes.

#### 6.3.1 $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratios

Activity ratios of  $^{238}\text{Pu}/^{239+240}\text{Pu}$  were determined in a subset of the humic surface soil samples; ratios were found to range within 0.022 and 0.07 ( $0.04 \pm 0.01$ ,  $n=27$ ). The activity ratios were in general higher than the  $^{238}\text{Pu}/^{239+240}\text{Pu}$  atom ratio established by Hardy *et al.* (1973) ( $0.026 \pm 0.04$ , 25 northern hemisphere locations with latitude higher than  $30^\circ\text{N}$  decay corrected to 01.01.2012). This might indicate an influence from Chernobyl debris. Activity ratios of  $^{238}\text{Pu}/^{239+240}\text{Pu}$  in debris from the Chernobyl accident were substantially higher than global fallout, Lujaniene *et al.* (2009) observed ratios in the range 0.44 – 0.5 in air in Vilnius in the days following the accident, while ratios in the range 0.03 – 0.70 and 0.04 – 0.57 were found in debris deposited in Finland and Sweden respectively (Lindahl *et al.*, 2004; Salminen-Paatero *et al.*, 2012). However, the distribution of elevated  $^{238}\text{Pu}/^{239+240}\text{Pu}$  atom ratios did not reflect known deposition of Chernobyl debris. It is conceivable that the  $^{238}\text{Pu}$  derived from the SNAP-9A breakdown have had different chemical properties than the  $^{239+240}\text{Pu}$  deposited after nuclear weapon tests. A lower wash-out of SNAP-derived  $^{238}\text{Pu}$  in the soils could then possibly offer an explanation of the higher activity ratios found.

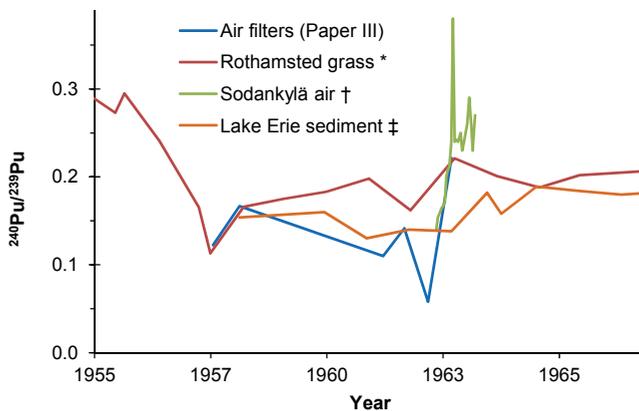
#### 6.3.2 $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios

The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios obtained in the present thesis varied within 0.161 – 0.211 in humic surface soil samples, (paper I), 0.15 – 0.19 the Austfonna ice core, (paper II), and 0.0517 – 0.237 in air filters from the period 1957 - 1963, (paper III). Figure 22 and Figure 23 shows summary plots of the  $^{240}\text{Pu}/^{239}\text{Pu}$  isotopic composition of the samples analysed in (paper III) along with literature values for comparison.

Radionuclides deposited at a site represent a mixture of fallout from regional and global fallout from atmospheric nuclear tests, reactor accidents and any locally deposited radionuclides. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios observed in the humic surface soils samples were used to assess the relative impact of fallout from atmospheric nuclear detonations and the Chernobyl accident in Norway. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios found (0.164 – 0.211 in samples from 1990 and 0.161 – 0.195 in samples

from 2005) indicated integrated global fallout to be the most important source. However, high atom ratios at some inland locations were attributed to a slight Chernobyl influence.

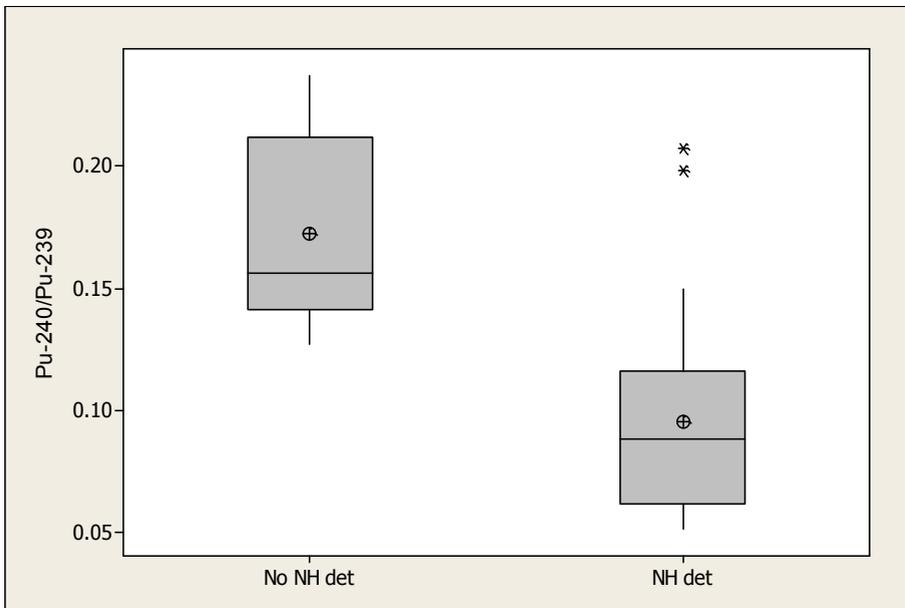
The ice core samples (Paper II) offer poorer time resolution and less secure dating than the air filters (Paper III). The time resolution is 1 - 3 years at best, depending on in core concentrations and post – depositional processes. Downward migration due to melting and infiltration disturb the isotopic signal (e.g. Tarussov (1992), Pinglot *et al.* (2003)). Despite the close geographical and climatological proximity of the Austfonna glacier, from which the core was extracted, to Former Soviet Union test sites Novaya Zemlya and Semipalatinsk, no significant deviations from global fallout levels of the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios were observed. In Figure 22  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios observed in the air filter samples (paper III) are compared with literature values. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios observed in the air filter samples (paper III) were found to be in agreement with ratios observed by Warneke *et al.* (2002) and Salminen-Paatero *et al.* (2012) in periods with coherent sampling times (1957, 1958 and 1963). In the remaining periods, autumn 1961 and 1962, the atom ratio of Pu in air reflected weapon debris from low and intermediate yield detonations.



**Figure 22.** Summary plot of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios discussed. Activity weighted yearly mean values for all stations have been used for the air filter samples (Paper III); 1962 split into spring (March and April) and winter (November). Literature values included for comparison: \* - Rothamsted herbage archive, Warneke *et al.* (2002), † - Surface air Sodankylä from Salminen-Paatero *et al.* (2012), ‡ - Lake Erie sediment from Winkler (2007).

During time periods associated with atmospheric nuclear testing at Novaya Zemlya and Semipalatinsk the air filters from all stations had  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios of  $0.06 \pm 0.01$  (activity weighted mean  $\pm 1$  s.d.), i.e., significantly lower than global

fallout ratios. This is opposed to periods without atmospheric nuclear testing wherein the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios of  $0.18 \pm 0.04$  approached stratospheric fallout ratios (cf. Figure 23 and appendix 1). Similar tendencies have been reported elsewhere, e.g. Salminen-Paatero *et al.* (2012) and Warneke *et al.* (2002) (Figure 22). The results of Pu isotope measurements in air filter samples (Paper III) suggest that debris from low yield detonations has had a strong influence on actinide concentrations in ground level air during periods of atmospheric testing at FSU test sites Semipalatinsk and Novaya Zemlya. The higher  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios seen in periods without atmospheric nuclear testing and in connection with spring peak deposition indicate influence of debris from high yield detonations, originally injected into the stratosphere.



**Figure 23.** Summary plot of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios during 1957 – 1963. The data are grouped into filters from periods with northern hemisphere atmospheric testing (NH det) versus periods without northern hemisphere atmospheric testing (no NH det). Ringed cross – mean of observations within the category. \* - outliers.

Debris from low yield detonations generally characterised by low  $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{241}\text{Pu}/^{239}\text{Pu}$  atom ratios (e.g. Hicks and Barr (1984), Beasley *et al.* (1998b), Lind (2006)), while high yield detonations generally yielded high  $^{240}\text{Pu}/^{239}\text{Pu}$ ,  $^{241}\text{Pu}/^{239}\text{Pu}$  atom ratios (e.g. Diamond *et al.* (1960), Yamamoto *et al.* (1996), Lindahl *et al.* (2012)). Debris from reactors features a range of different  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios, depending on burn up and the history of the fuel. Characteristic for fuels from power production reactors and recycled fuels is, however, elevated  $^{240}\text{Pu}/^{239}\text{Pu}$

atom ratios due to accumulation of heavier Pu isotopes and fission of  $^{239}\text{Pu}$  (Lovins, 1980). Publications stating the  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios in debris from nuclear detonations are rare, and mostly concerning local fallout from low yield detonations (Beasley *et al.*, 1998b) and global fallout (Ketterer *et al.*, 2007; Sakaguchi *et al.*, 2009). Information on this ratio in debris from high yield detonations has not been found. However, the ratio is expected to be higher than in global fallout.

Detonations at STS were generally of low yield (average 59 kt, range 0.001 – 1600 kt). (UNSCEAR, 2000a; Björklund and Goliath, 2009). In contrast detonations at NZ were of high and very high yield (average 2900 kt, range 2 – 5800 kt) (UNSCEAR, 2000a; Björklund and Goliath, 2009). The arctic location of NZ would cause most of the debris from atmospheric nuclear detonations conducted here to be apportioned to the stratosphere, and thus to be dispersed and deposited as global fallout (Peterson, 1970; UNSCEAR, 2000a). Furthermore, it has been indicated that most detonations at NZ were carried out under wind directions that would cause tropospheric debris to be deposited onto FSU territories (Khalturin *et al.*, 2005).

Previous work on high time resolution samples has been done by Warneke *et al.* (2002) and Salminen-Paatero *et al.* (2012). Like the air filter samples analysed in Paper III, the samples analysed by Salminen-Paatero *et al.* (2012) permit a time resolution down to 24 hours, while the herbage archive used by Warneke *et al.* (2002) has been collected biannually (spring and autumn) to annually, with a time resolution down to three months at best. The air filter samples were collected daily, and the fine time resolution and secure dating allows for observation of short term trends and episodes which is not possible in other sample types like ice cores, sediment cores or soil samples.

### **6.3.3 $^{236}\text{U}/^{239}\text{Pu}$ atom ratios in ice core samples and air filter samples**

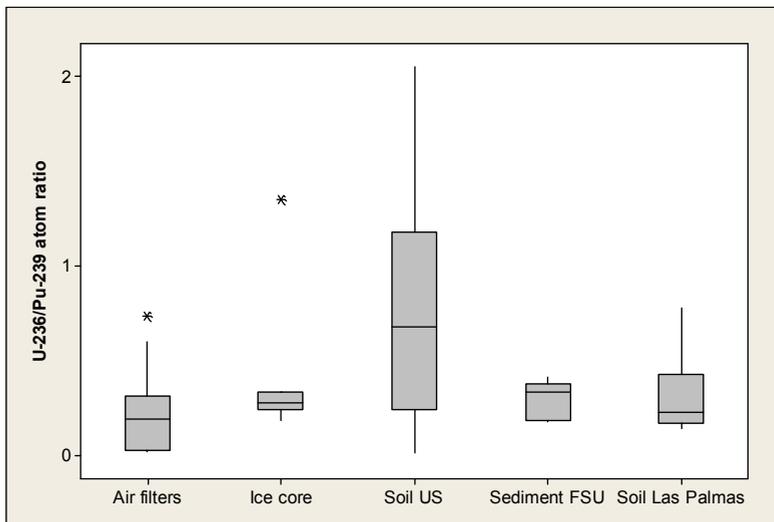
In the present work  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios have been determined in an Arctic ice core from Nordaustlandet and in selected air filter samples;  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios in the range 0.0188 – 0.7 (air filters) and 0.18 – 1.4 (ice core) were found.

The  $^{236}\text{U}/^{239}\text{Pu}$  atom ratio in the Austfonna ice core was predominately found within the global fallout range reported by Ketterer *et al.* (2007). There was a tendency of higher  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios at depths corresponding to pre-moratorium deposition than at depths corresponding to post-moratorium deposition; however, uncertainties were high. The  $^{236}\text{U}/^{239}\text{Pu}$  atom ratio observed at depths corresponding to the period 1975 – 1983 is very high ( $1.4 \pm 0.4$ ), however,

this is in agreement with reports of  $^{236}\text{U}/^{239}\text{Pu}$  in riverine- and sea-water (Eigl *et al.*, 2013) and soil samples (Srncik *et al.*, 2011; Ketterer *et al.*, 2013).

The  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios observed in the air filters (Paper III) were predominately within the range of global fallout reported by Ketterer *et al.* (2007). During the passage over Norway of a radioactive cloud attributed to low yield tests at STS in October / November 1962 (further discussed in chapter 6.5), very low  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios were observed. These ratios were significantly different from the values reported by Ketterer *et al.* (2003) to be representative of global fallout, but largely in agreement with ratios observed in local fallout at STS by (Beasley *et al.*, 1998b).

The  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios obtained from the analysis of the ice core and the air filters samples (paper II and paper III) are illustrated in Figure 24, along with literature data for comparison. Soil US and Sediment FSU refers to  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios in soil samples from Washington state (US), and the Pechora region in the FSU calculated from the data presented by Ketterer *et al.* (2013); Soil Las Palmas refers to bulk analysis of soil samples from La Palma, Canary Islands, as reported in Srncik *et al.* (2011).



**Figure 24. Boxplot of  $^{236}\text{U}/^{239}\text{Pu}$  atom ratios observed in the present thesis. Air filter samples from Norway during 1957 – 1963 (Paper III), the ice core from Nordaustlandet (Paper II). \* denotes outliers. Literature values included for comparison: Soil US – soil samples from Washington state USA and Sediment FSU – lake sediments from the Pechora region (Ketterer *et al.*, 2013). Bulk soil samples (0 – 5 cm) from La Palma, Canary Islands Srncik *et al.* (2011).**

The observed variations in the  $^{236}\text{U}/^{239}\text{Pu}$  atom ratio likely reflect variations in weapon design. The main source of  $^{236}\text{U}$  in nuclear weapon debris is thermonuclear fission – fusion – fission reactions in which fast neutrons released from a fusion

secondary stage induces fission in a tertiary tamper stage of natural or depleted U (Winkler *et al.*, 2012). In this process  $^{236}\text{U}$  is produced in neutron capture in  $^{235}\text{U}$  or the reaction  $^{238}\text{U} (n, 3n) ^{236}\text{U}$ . Nuclear weapon material in pure fission weapons or thermonuclear primaries has been indicated to consist of pure U, Pu or a combination of the two (e.g. Bukharin (1998), Eriksson *et al.* (2005), Ranebo *et al.* (2007), Lind *et al.* (2007)). The production of  $^{236}\text{U}$  in pure fission and fission - fusion detonations is likely lower than in fission – fusion – fission detonations, and the  $^{236}\text{U}/^{239}\text{Pu}$  atom ratio in debris from low yield detonations has been indicated to be low (Paper III and Beasley *et al.* (1998b)).

#### 6.4 Plutonium isotopes heavier than $^{240}\text{Pu}$

Analysis of Plutonium isotopes heavier than  $^{240}\text{Pu}$  were performed in selected humic surface soil samples (paper I), pooled air filters from 1957 (Sola 9 – 21 October 1957), 1962 (Røros 16 – 26 march 1962) and 1963(Røros 1 – 11 June 1963), and selected filters from November 1962 (paper III). The results are presented in Table 5 and Figure 25.

The  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratio observed in the pooled air filter samples and the humic surface soil samples ranged within  $1.3 \times 10^{-3}$  and  $10 \times 10^{-3}$ . The lowest ratios were found in pooled air filter samples from 1961, and the highest atom ratios were found in the humic surface soil samples.

A higher  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratio was found in the pooled filter sample from 1957 than from 1962. The same tendency is observed in the  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratios in a time resolved sediment core from Lake Erie, wherein substantially higher ratios ( $5.36 \times 10^{-3}$ ) were found the moratorium than after ( $3.20 \times 10^{-3}$  for the years 1961 to 1964) (Winkler *et al.*, 2004). The pooled filters from 1957 would have collected debris from atmospheric detonations in the few months preceding the sampling. Transfer of debris from the stratosphere to the troposphere is at its lowest during the months September through October (UNSCEAR, 2000a), and contemporary atmospheric testing activities would likely easily overwhelm the Pu atom ratio signatures of stratospheric debris. The pooled filters from 1962 would have captured almost exclusively stratospheric debris for the following reasons:

- The sampling period was during the peak transfer of debris from the stratosphere to the troposphere as defined in (UNSCEAR, 2000a)
- Only the US detonation the 05.03.1962 was close enough geographically to affect the filters. However, this detonation was as small cratering detonation (Björklund and Goliath, 2009), depositing most of its debris close to the detonation site. Debris from the detonation conducted above Christmas Island

the 25.04.1962 is unlikely to have reached Norway soon enough to influence Pu concentrations and atom ratios in the air filters from the sampling period.

The most likely origin of the debris in the pooled air filters from 1962 is then FSU test series in September through November 1961. This test series encompassed small, large and very large detonations injecting debris into different parts of the atmosphere. The stratospheric residence time of debris from the largest detonations were long, and peak deposition of this debris would not be expected to take place until the spring of 1963 (Peterson, 1970). Thus the radionuclide composition in the troposphere in the first half of 1962 should be dominated by debris from detonations in the range 0.7 – 4 Mt. Eight detonations performed in 1961 had a yield in the size range suitable for inserting debris into the lower polar stratosphere. The  $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratios measured in the spring of 1962, 0.145 and  $1.3 \times 10^{-3}$  is likely representative of the mixed debris from these detonations.

**Table 5. Atom ratios obtained in paper I (upper part, humic surface soil samples) and paper III (lower part, air filter samples).**

Site	Date / year	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{241}\text{Pu}/^{239}\text{Pu}$ $\times 10^{-3}$	$^{242}\text{Pu}/^{239}\text{Pu}$ $\times 10^{-3}$	$^{244}\text{Pu}/^{239}\text{Pu}$ $\times 10^{-4}$
Glomfjord	2005	$0.20 \pm 0.02$	$2.3 \pm 0.8$	$10 \pm 2$	n.a.
Trolla	2005	$0.17 \pm 0.01$	$1.5 \pm 0.6$	$4.5 \pm 0.7$	n.a.
Fyllingsdalen	2005	$0.17 \pm 0.01$	$1.4 \pm 0.4$	$5.1 \pm 0.9$	n.a.
Kløfta	2005	$0.19 \pm 0.02$	$0.9 \pm 0.5$	$6 \pm 1$	n.a.
Svolvær	2005	$0.17 \pm 0.01$	$1.3 \pm 0.3$	$3.8 \pm 0.5$	n.a.
Røros	09.11.1962	$0.0605 \pm 0.0008$	$0.46 \pm 0.06$	n.a.	n.a.
Bergen	11.11.1962	$0.0517 \pm 0.0009$	$0.62 \pm 0.09$	n.a.	n.a.
Ålesund	11.11.1962	$0.0574 \pm 0.0006$	$0.37 \pm 0.04$	n.a.	n.a.
Ålesund	12.11.1962	$0.061 \pm 0.001$	$0.25 \pm 0.05$	n.a.	n.a.
Bergen	9.10-21.10.57	$0.116 \pm 0.007$	n.a.	$2.8 \pm 0.5$	n.a.
Røros	16.4 - 26.4.62	$0.145 \pm 0.003$	$0.4 \pm 0.1$	$1.3 \pm 0.1$	b.d.
Røros	1.6 - 11.6.63	$0.220 \pm 0.003$	$1.6 \pm 0.2$	$4.5 \pm 0.2$	$1.7 \pm 0.5$

n.a. – not analysed, b.d. – below the detection limit  
reference date 01.01.2012.

The pooled air filter sample from 1963 had a  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratio higher than established integrated global fallout (Kelley *et al.*, 1999) and contemporary levels (Winkler, 2007). However, consistently high gross beta activities indicate that

these filters were deployed during the spring peak deposition of stratospheric fallout. The fallout thus captured would include debris from the high yield detonations from 1961 and possibly 1962, and high Pu atom ratios should be expected. It should also be mentioned that the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in this sample agrees well with ratios reported by Warneke *et al.* (2002) and Salminen-Paatero *et al.* (2012) for the same time period.

The  $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratios in the humic surface soil samples were well correlated ( $R^2_{\text{adj}}=0.85$  and  $p=0.01$ ). The higher  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratios found at Glomfjord and Kløfta may reflect a slight deposition of Chernobyl-related plutonium. The calculated  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratio in the fuel of the Chernobyl reactor at the time of the accident ranged within 0.039 – 0.048 (UNSCEAR (2000b), and references therein), i.e., approximately 10 times the global fallout ratio of  $0.0039 \pm 0.0007$  (Kelley *et al.*, 1999).

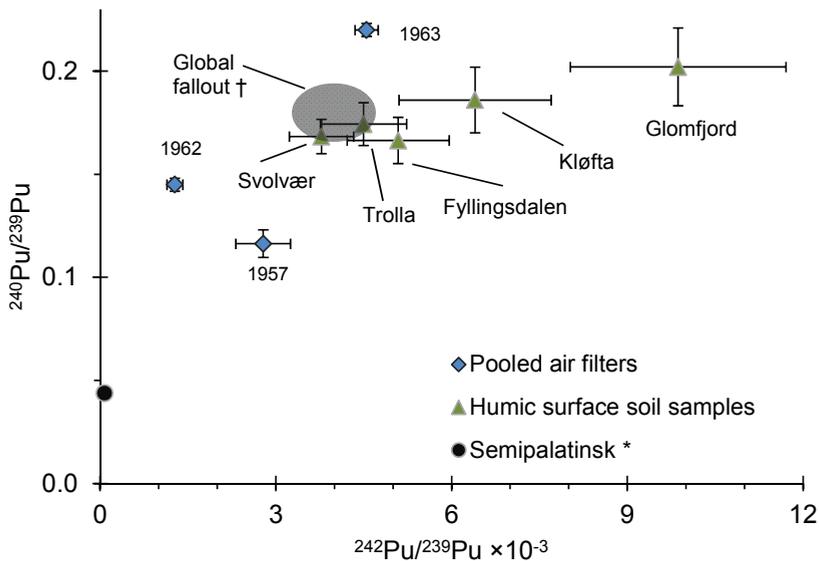


Figure 25. Atom ratios of  $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{242}\text{Pu}/^{239}\text{Pu}$  in the present work, reference date 01.01.2012. The pooled air filter samples are based on ten consecutive days of sampling in Bergen in autumn 1957 and Røros in spring 1962 and 1963. Literature values included for comparison, \* Beasley *et al.* (1998b), † Kelley *et al.* (1999).

## 6.5 Source identification

### 6.5.1 Debris from the Windscale accident

Pu activity concentrations and  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios were determined in air filters from Sola, 9th, 16th, 17th, 22nd October 1957. In addition  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios were determined in grouped air filters from Bergen covering the period 9 to 21 October. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios were found to range within 0.09 – 0.12 in this period. Atmospheric dispersion modelling (e.g. Bergan *et al.* (2008)) and elevated gross beta activities in the air filters indicate that the debris cloud from the Windscale fire arrived to the Norwegian West coast the 16 October. The highest gross beta concentrations were measured at Sola and Bergen (321 and 249 mBq m<sup>-3</sup> respectively, the 16 October), however, the Pu concentrations did not peak on the same day as the gross beta concentrations. The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio observed in the filter from the 16th October was lower than the other filters from the same period, however, it is not likely that the low  $^{249}\text{Pu}/^{239}\text{Pu}$  atom ratio reflects Windscale Pu. The releases of Pu from the Windscale fire have been estimated to be low, assuming an  $^{240}\text{Pu}/^{239}\text{Pu}$  below 0.07 for weapons grade Pu, the total estimated releases of Pu would be less than 45 g (Garland and Wakeford, 2007).

Between 22 August and 10 October 1957, 7 nuclear detonations with yields in the range 0.006 – 2.9 Mt were conducted at FSU test sites NZ and STS. Pu in debris from these detonations should be expected to overwhelm the non-local fallout from the Windscale accident. Thus, the Pu captured in these filters likely originates from nuclear weapons rather than the Windscale plant.

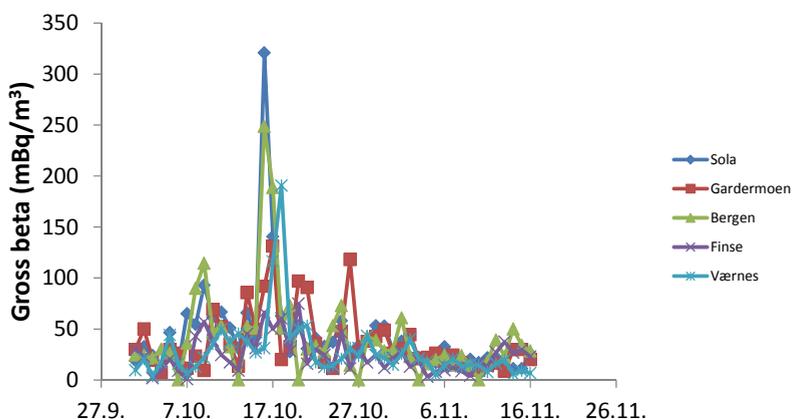


Figure 26. Gross beta activities in Norwegian ground level air measured during the first few days after the Windscale accident in 1957 (Bergen). Peak activities were seen at the west coastal stations Sola and Bergen before proceeding east / northeast to Gardermoen and Værnes.

### 6.5.2 Debris from the Chernobyl accident

In Paper I, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios (0.161 – 0.211) for both series of the humic surface soil samples were mainly reflecting global fallout. Significant deviations from global fallout atom ratios were found for some inland locations (Kongsvoll, Skredå, Vauldalen, Kløfta, Stavsjø and Øystre Slidre) indicating influence of Chernobyl related debris. A selection of  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios obtained in this work and in southern Sweden and Finland is shown in Figure 27.

Lower variation was observed in the results from this work compared to the results from Sweden (Lindahl *et al.*, 2004) and Finland (Salminen-Paatero *et al.*, 2012). Lichens, as used by both authors, absorbs most of its nutrients and hence contaminants, from the air (Lindahl *et al.*, 2004; Steinnes, 2008). Chernobyl derived Pu deposited onto lichens is thus less likely to be diluted by global weapons fallout Pu. Similarly, peats from commercially exploited peatlands, as used by Salminen-Paatero *et al.* (2012), are also likely to represent less diluted Chernobyl fallout, provided that harvest of the peat layer was conducted between the main deposition of global fallout and the Chernobyl accident. Additionally, as reviewed by Pöllänen *et al.* (1997), deposition of Chernobyl related debris was highly heterogeneous with numerous hot spots ranging in size from 30 cm up to several kilometres.

The influence of debris from the Chernobyl accident on  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in the humic surface soil samples was assessed in (paper I) using the formula:

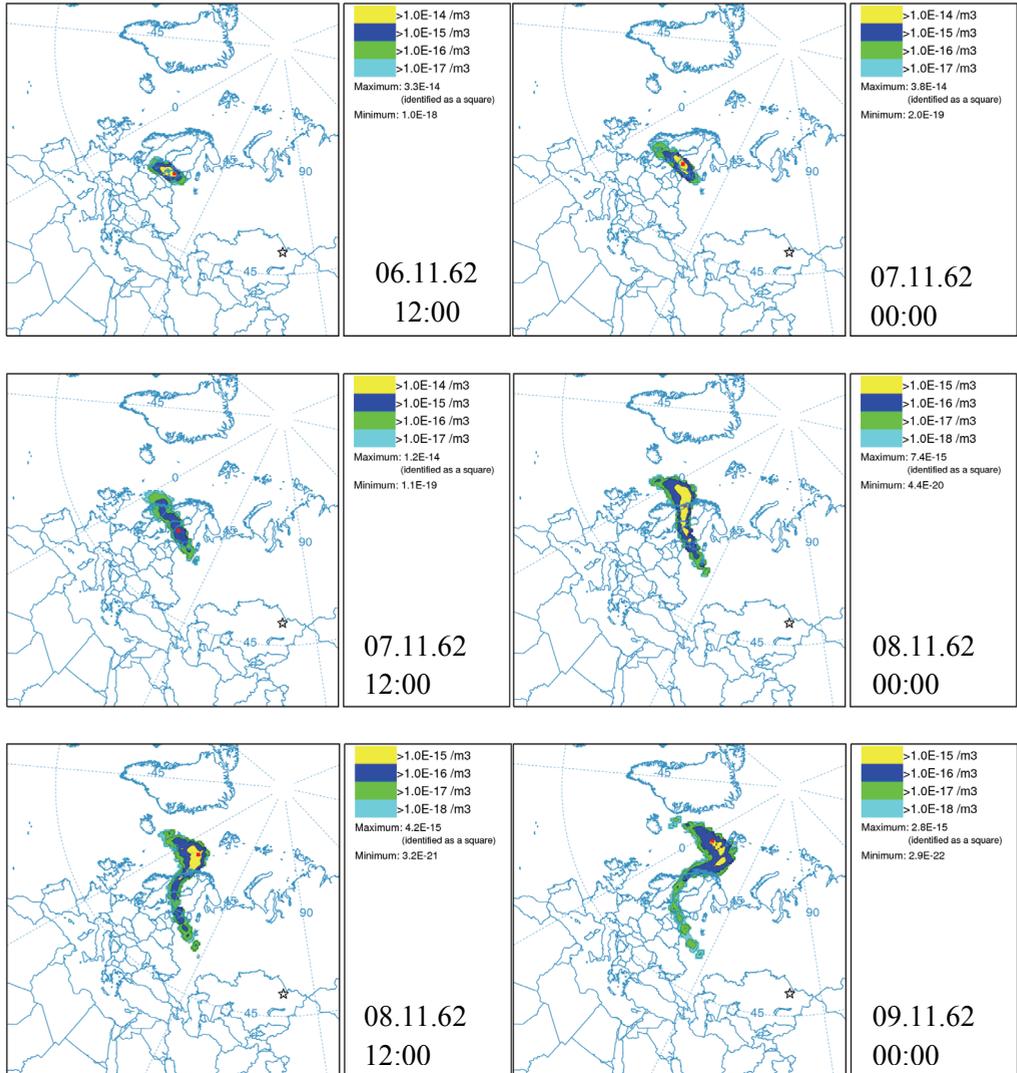
$$X = \frac{(R_{\text{Sample}} - R_{\text{GFO}})}{(R_{\text{Chb}} - R_{\text{GFO}})} * 100\%$$

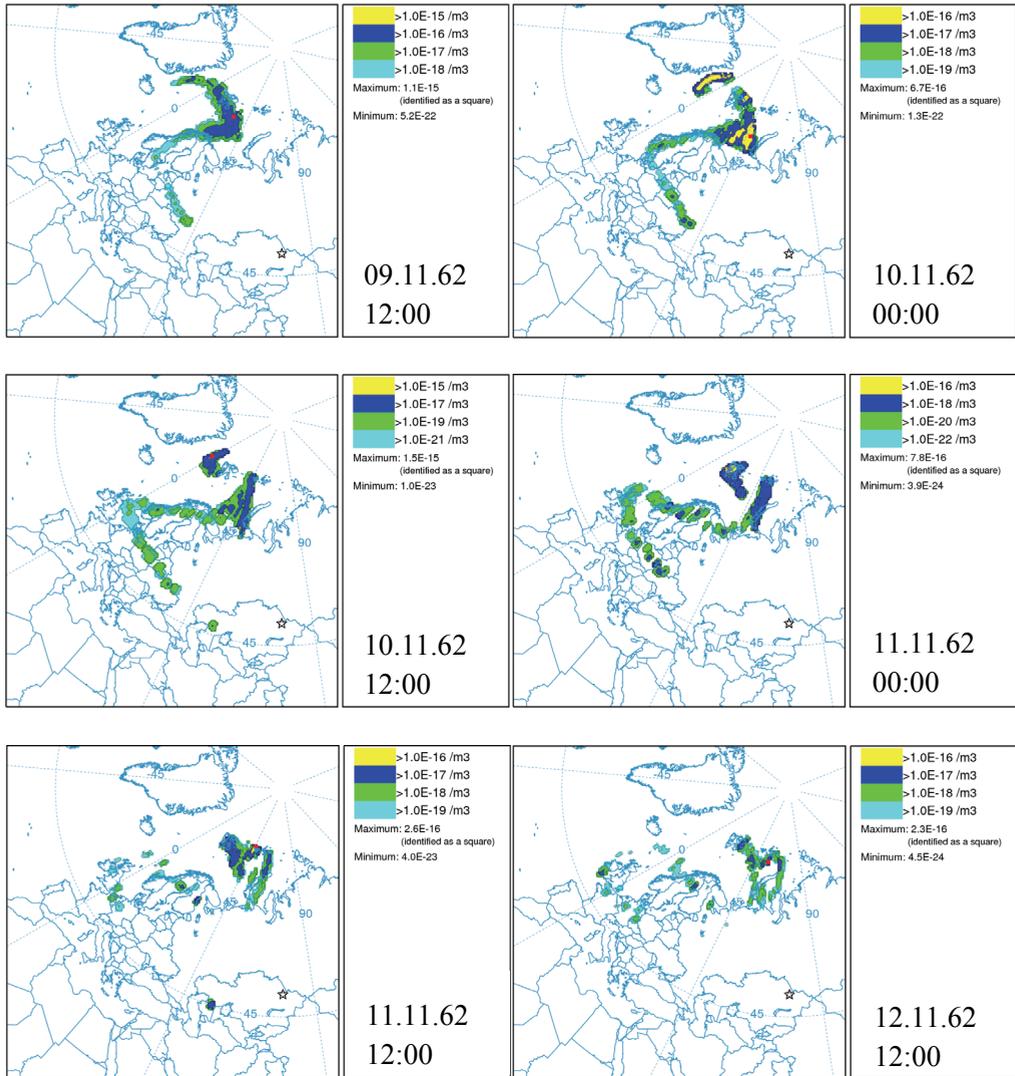
where  $R_{\text{GFO}}$  is the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in global fallout, (0.18 according to Kelley *et al.* (1999)) and  $R_{\text{Chb}}$  ( $0.40 \pm 0.07$  is the estimated  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio given in UNSCEAR (2008)). The calculated Chernobyl debris influence on the  $^{239+240}\text{Pu}$  concentrations at sites Kongsvoll, Skredå, Vauldalen, Kløfta, Stavsjø and Øystre Slidre was calculated to range within 8 and 13 %.

The  $^{241}\text{Pu}/^{239}\text{Pu}$  atom ratios in the humic surface soil samples suffered from large uncertainties as a consequence of low concentrations. Nevertheless, the Glomfjord sample displayed a  $^{241}\text{Pu}/^{239}\text{Pu}$  atom ratio higher than global fallout indicating a Chernobyl influence of the deposition at this site. The indication of a Chernobyl influence at Glomfjord was further corroborated by the  $^{242}\text{Pu}/^{239}\text{Pu}$  atom ratio, which also indicated that the Kløfta sample had been influenced by Chernobyl derived Pu. The  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio measured in soils from 1986 from areas close to Kløfta and Glomfjord (Ullensaker (0.43) and Rana (0.35)), indicated that



Hysplit simulations were performed simulating the transport of debris from detonations at Semipalatinsk and Novaya Zemlya. The results indicate that the elevated activity concentrations seen in surface air in Norway during 7 to 13 November 1962 originated from one or more of the three atmospheric nuclear detonations above Novaya Zemlya in the period 30 October to 1 November 1962. Figure 28 shows the HYSPLIT simulation of a release above Semipalatinsk test sited the 31 November 1962.





**Figure 28. Summary of a selected HYSPLIT simulation of the radioactive cloud from a low yield detonation above Semipalatinsk the 31.10.1962. Release heights between 1750 – 5000 m were simulated, and post detonation debris concentrations in air between 0 and 75 m above ground were simulated. The figure shows the position of the plume from 6.11.1962 at 12:00 and with 12 hour increments thereafter.**

Air concentrations of Pu and  $^{236}\text{U}$  concentrations during this episode were exceptionally high (11.7 - 782  $\mu\text{Bq m}^{-3}$  and 4.6 – 20.1  $\text{nBq m}^{-3}$  respectively). However, it is unlikely that this would affect integrated fallout. Deposition is strongly correlated to precipitation (e.g. Storebø (1968), Pálsson *et al.* (2012)),

which was very low during the passage of the plume (www.eklima .no; D.R. Kristoffersen, pers. comm.).

The time resolution of Pu in the ice core (Paper II), was insufficient for the direct identification of detonations. High pre moratorium  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios indicative of fallout from the US thermonuclear detonations in 1952 and 1954, documented in other publications (e.g. Koide *et al.* (1985), Olivier *et al.* (2004), Warneke *et al.* (2002)) were not observed. Positive inflections in the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio caused by fallout from the large Former Soviet Union test series in 1961 and 1962 could not be identified either. The difference between the current results and the results of Warneke *et al.* (2002) are easily explained by the poorer resolution of the Austfonna ice core, and the fact that the highest atom ratios observed by Warneke *et al.* (2002) were associated with low deposition rates. The high atom ratio observations made by Koide *et al.* (1985) and Olivier *et al.* (2004) on the other hand are associated with high deposition rates, and the reason for the discrepancy must be sought elsewhere. As reviewed by Barrie (1986), the arctic winter air mass covers large areas of continental Europe and Central Asia as far south as 40 °N. Pollutants tropospherically transported from Europe and Central Asia enters the Arctic from South-East (Barrie, 1986; Stohl *et al.*, 2000). Furthermore, the deposition profile observed at Nordaustlandet contrasts with deposition profiles established for other sites (Koide *et al.*, 1985; Warneke *et al.*, 2002; Olivier *et al.*, 2004). This indicates a different deposition regime at Svalbard. The mixture of debris from low yield, low  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio debris from STS with debris from high yield, high  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio debris from NZ could offer an explanation.

## 7 Concluding remarks

In the present thesis source identification and source assessment have been conducted based on Pu and U atom and activity ratios in radioactive debris in humic surface soil samples from mainland Norway (Paper I); an arctic ice core covering the period 1949 - 1999 (Paper II); and air filter samples from Norway during 1957 - 1963 (Paper III). Radioactive particles have been searched for and identified in the air filter samples the ice core samples (Paper II) and (Paper III). Atmospheric dispersion modelling (NOAA HYSPLIT\_4) has been used for source identification purposes in (Paper III).

The main objectives of the present work have been to assess the inventory and provenance of Pu from atmospheric nuclear weapon detonations and reactor accidents deposited on Norwegian territories. A combination of advanced mass spectrometry techniques and atmospheric dispersion modelling has been applied.

In the following section the working hypotheses of the present work will be answered based on the obtained results.

H1. *Pu originating from nuclear weapons tests should follow the precipitation pattern, and be enriched at the western coast of Norway*

- Based on concentrations and atom ratios of Pu in humic surface soils, the dominant source was found to be global fallout. Deposition was found to be highest along the western coast in accordance with high annual precipitation rates.

H2. *Traces of Pu associated with reactor fallout should be more unevenly distributed than debris from weapons tests, and it should be possible to identify the signal at certain inland sites in Norway.*

- Pu in integrated fallout samples like surface soils is largely attributed to global fallout Pu. Thus the impact of deposition from a source must be substantial in order to be detected by means of isotope ratio measurements. Nevertheless, higher than global fallout  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios were observed at some eastern inland sites in Norway. Further analysis on the abundances of heavier Pu isotopes ( $^{241}\text{Pu}$  and  $^{242}\text{Pu}$ ) for a sub-selection of the soils confirmed influence of Chernobyl related Pu at several inland sites.

H3. *Inert radioactive particles are expected to be present in samples affected by fallout from the nuclear weapons tests and nuclear reactor accidents*

- Autoradiography hotspots indicated the presence of radioactive particles on filters collected during periods associated with atmospheric nuclear tests at

FSU test sites Novaya Zemlya and Semipalatinsk. Autoradiography of air filters revealed the presence of radioactive particles almost exclusively in periods during and after atmospheric nuclear weapons detonations, while there were very few particle indications during periods of high stratospheric deposition (spring peak deposition). Hotspots were also observed in ice core filtrates, during time periods associated with FSU atmospheric testing activities (1949 – 1965) and the Chernobyl accident or a vented underground test at Novaya Zemlya (1985 - 1989).

*H4. Over time the Pu concentrations in soil will decrease*

- Pu concentrations in parallel samplings of humic surface soils in 1990 and 2005 were analysed. A clear decrease in  $^{239+240}\text{Pu}$  concentrations was found.

*H5. Pu and U isotopic composition or atom ratios can be utilized for source identification. If the time resolution is sufficiently good, single events contributing to the deposition of Pu and  $^{236}\text{U}$  can be identified.*

- Source identification and source assessment was performed both for high time resolution samples and integrated fallout samples. Pu and U isotopes captured in archive air filters (time resolution 24 h) allowed for the identification of tropospherically transported fallout from FSU test site Semipalatinsk. Global fallout was found to be the dominant source of Pu deposition in mainland Norway. However, an impact from the Chernobyl accident was shown as well.

In summary, the following conclusions can be drawn:

- Pu deposition in the terrestrial environment of Norway was found to be primarily caused by global fallout. However, a slight Chernobyl influence was found at several inland sites.
- Pu and  $^{236}\text{U}$  concentration profiles in an ice core from Austfonna (Svalbard) showed higher concentrations at depths corresponding to pre-moratorium deposition (1956 – 1959) than post-moratorium (1959 – 1962). This is in contrast to timer resolved sampling from other locations, and indicates either the influence of post depositional processes altering concentration profiles of particulate radionuclides in the glacier, or a different deposition pattern at this site compared to other sites.
- Pu in air showed a clear influence of debris from FSU test sites NZ and STS during periods associated with testing at these sites, attaining  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios significantly lower than global fallout. Similarly, the presence of radioactive particles was much higher during these periods. During periods without atmospheric nuclear testing at NZ and STS,  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios reflected global (stratospheric fallout).

**Appendix 1. Summary of gross  $\beta$  activities, Pu concentrations,  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios and particle indications in the air filters analysed in paper III**

Site	Date	Gross $\beta$ mBq m <sup>-3</sup>	Particles	$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio	$^{239+240}\text{Pu}$ $\mu\text{Bq m}^{-3}$
Sola	09.10.1957	93	0	0.13 $\pm$ 0.01	7.4 $\pm$ 0.2
Sola	16.10.1957	321	0	0.09 $\pm$ 0.02	3.0 $\pm$ 0.2
Sola	17.10.1957	141	0	0.115 $\pm$ 0.006	3.5 $\pm$ 0.2
Bergen	08 - 20.10.1957	941	-	0.116 $\pm$ 0.007	n.d.
Sola	22.10.1957	41	0	0.15 $\pm$ 0.02	2.2 $\pm$ 0.1
G.moen	01.06.1958	127	0	0.169 $\pm$ 0.008	22.1 $\pm$ 0.5
G.moen	02.06.1958	152	0	0.170 $\pm$ 0.007	24.9 $\pm$ 0.5
G.moen	03.06.1958	155	0	0.162 $\pm$ 0.007	28.4 $\pm$ 0.6
Sola	27.09.1961	235	1	0.090 $\pm$ 0.008	6.9 $\pm$ 0.2
G.moen	28.09.1961	327	0	0.09 $\pm$ 0.01	1.96 $\pm$ 0.08
Røros	06.10.1961	207	2	0.088 $\pm$ 0.005	2.86 $\pm$ 0.05
Bergen	19.10.1961	428	2	0.086 $\pm$ 0.009	3.2 $\pm$ 0.1
Røros	19.10.1961	98	0	0.20 $\pm$ 0.07	15 $\pm$ 3
Vadsø	19.10.1961	130	1	0.14 $\pm$ 0.03	1.3 $\pm$ 0.1
Bergen	27.10.1961	65	0	0.09 $\pm$ 0.02	1.8 $\pm$ 0.1
Røros	27.10.1961	35	0	b.d.	b.d.
Vadsø	27.10.1961	83	0	0.15 $\pm$ 0.02	1.73 $\pm$ 0.09
Bodø	10.11.1961	1093	0	0.093 $\pm$ 0.014	14.9 $\pm$ 0.7
Tromsø	11.11.1961	888	0	0.07 $\pm$ 0.002	1.7 $\pm$ 0.1
Tromsø	12.11.1961	1747	7	0.067 $\pm$ 0.006	16.3 $\pm$ 0.5
Ålesund	17.11.1961	305	1	0.100 $\pm$ 0.007	4.3 $\pm$ 0.1
Røros	11.01.1962	199	0	0.13 $\pm$ 0.01	2.25 $\pm$ 0.09
Bergen	17.01.1962	297	0	0.110 $\pm$ 0.007	18.1 $\pm$ 0.5
Røros	04.03.1962	58	0	0.127 $\pm$ 0.005	12.3 $\pm$ 0.2
Bergen	15.04.1962	209	0	0.138 $\pm$ 0.006	26.7 $\pm$ 0.5
Bergen	16.04.1962	205	0	0.137 $\pm$ 0.005	27.8 $\pm$ 0.4
Bergen	17.04.1962	263	0	0.142 $\pm$ 0.003	30.3 $\pm$ 0.4
Bergen	18.04.1962	104	0	0.148 $\pm$ 0.006	15.4 $\pm$ 0.3
Bergen	19.04.1962	157	0	0.138 $\pm$ 0.007	21.4 $\pm$ 0.5
Bergen	20.04.1962	252	0	0.141 $\pm$ 0.004	36.0 $\pm$ 0.5
Bergen	21.04.1962	219	0	0.152 $\pm$ 0.005	29.1 $\pm$ 0.4
Bergen	22.04.1962	207	0	0.140 $\pm$ 0.005	27.8 $\pm$ 0.4

Bergen	23.04.1962	222	0	0.156 ± 0.005	34.0 ± 0.5
Bergen	24.04.1962	154	0	0.152 ± 0.006	19.7 ± 0.4
Vadsø	25.04.1962	78	0	0.14 ± 0.01	3.2 ± 0.1
Røros	15 - 25.04.1962	1992	2	0.145 ± 0.003	n.d.
Sola	09.11.1962	3142	15	0.060 ± 0.001	169 ± 2
G.moen	09.11.1962	9092	15	0.0614 ± 0.0007	575 ± 5
Røros	09.11.1962	11527	22	0.0574 ± 0.0006	487 ± 4
Bodø	09.11.1962	8132	2	0.054 ± 0.001	301 ± 3
Tromsø	10.11.1962	2050	4	0.063 ± 0.003	116 ± 2
Bergen	11.11.1962	6327	8	0.059 ± 0.001	267 ± 2
Ålesund	11.11.1962	15467	9	0.0517 ± 0.0009	782 ± 7
Værnes	11.11.1962	2362	3	0.062 ± 0.002	121 ± 1
Vadsø	11.11.1962	464	8	0.077 ± 0.004	11.7 ± 0.2
Ålesund	12.11.1962	8781	9	0.061 ± 0.0008	631 ± 5
Ålesund	13.11.1962	1762	0	0.064 ± 0.003	87 ± 1
Ålesund	17.11.1962	48	1	0.21 ± 0.01	2.48 ± 0.09
Bergen	28.05.1963	162	0	0.190 ± 0.006	20.1 ± 0.3
Bergen	29.05.1963	130	0	0.195 ± 0.007	27.1 ± 0.5
Bergen	30.05.1963	307	0	0.23 ± 0.01	18.2 ± 0.4
Bergen	31.05.1963	499	0	0.204 ± 0.007	26.3 ± 0.5
Bergen	01.06.1963	394	1	0.237 ± 0.005	53.9 ± 0.7
Bergen	03.06.1963	332	0	0.227 ± 0.007	35.3 ± 0.6
Bergen	06.06.1963	376	0	0.221 ± 0.005	20.3 ± 0.3
Røros	08.06.1963	817	0	0.225 ± 0.004	157 ± 2
Røros	31.05-10.06.1963	4681	0	0.227 ± 0.004	n.d.

**Appendix 2. Summary of some results from the humic surface soil samples in paper I, the  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratio and retention of  $^{239+240}\text{Pu}$  and  $^{137}\text{Cs}$ . Numbers in brackets are outliers, cf. chapter 6.1.4.**

Site	$^{238}\text{Pu}/^{239+240}\text{Pu}$ 1990		Retention			
	Activity ratio	±	$^{137}\text{Cs}$		$^{239+240}\text{Pu}$	
			%	±	%	±
Onsøy	0.049	0.004	n.a.	n.a.	n.a.	n.a.
Kløfta	0.040	0.005	(136)	(4)	60	6
Magnor	0.07	0.03	46	4	2.84	0.2
Stavsjø	0.043	0.007	39	2	52	4
Østre slidre	0.059	0.006	76	2	16	2
Øyer	n.a.	n.a.	50	2	75	17
Gjendesheim	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Heidal	0.030	0.003	n.a.	n.a.	n.a.	n.a.
Hønefoss	n.a.	n.a.	13.2	0.9	5.61	0.7
Langesund	0.063	0.005	65	2	(90)	(7)
Rjukan	0.045	0.003	16.8	0.7	(218)	(11)
Gjøvdal	0.027	0.002	23	1	66	9
Hylestad	0.040	0.004	n.a.	n.a.	n.a.	n.a.
Søgne	0.045	0.005	18	1	5.52	0.5
Vatland	0.046	0.004	47	2	65	13
Skredå	n.a.	n.a.	27	2	28	2
Ålgård	0.06	0.01	33	2	(158)	(9)
Odda	0.039	0.005	33	1	57	11
Fyllingsdalen	0.043	0.003	54	4	(98)	(3)
Stamnes	0.025	0.002	20	1	(1.9)	(0.3)
Sløvåg	0.036	0.004	43	2	4	1
Svelgen	n.a.	n.a.	35	2	71	2
Utvik	0.036	0.002	37	1	2.10	0.1
Grønning	0.022	0.001	21	1	78	8
Kongsvoll	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Vauldalen	n.a.	n.a.	38	2	10.98	0.6
Trolla	n.a.	n.a.	20.9	0.9	44	3
Forsnes	0.040	0.006	n.a.	n.a.	n.a.	n.a.
Teveldal	n.a.	n.a.	15.0	0.4	5.39	0.9

Udland	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Muru	0.043	0.009	n.a.	n.a.	n.a.	n.a.
Høylandet	n.a.	n.a.	45	1	51	3
Grane	0.034	0.004	n.a.	n.a.	n.a.	n.a.
Mosjøen	0.052	0.003	9.2	0.5	3	1
Koksverket	0.036	0.003	28	2	(497)	(91)
Glomfjord	0.055	0.005	38	2	73	14
Ankenes	0.038	0.006	59	4	(157)	(8)
Svolvær	n.a.	n.a.	92	5	34	2
Stonglandet	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Stilla	n.a.	n.a.	9.9	0.6	21	1
Slåtten	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Aiddejávri	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Syltefjord	0.033	0.002	n.a.	n.a.	n.a.	n.a.
Jakobsnes	n.a.	n.a.	23	1	(98)	(10)
Svanvik	n.a.	n.a.	20	1	6.93	0.5

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