

# Thule-2003 - Investigation of radioactive contamination

**Nielsen, Sven Poul; Roos, Per**

*Publication date:*
2006

*Document Version*
Publisher's PDF, also known as Version of record

RISØ

Risø-R-1549(EN)

# Thule-2003 - Investigation of Radioactive Contamination

Sven P. Nielsen and Per Roos

Risø National Laboratory
Roskilde
Denmark
May 2006

**Author:** Sven P. Nielsen and Per Roos
**Title:** Thule-2003 – Investigation of Radioactive Contamination
**Department:** Radiation Research

**Risø-R-1549(EN)**
**May 2006**

**Abstract**

Analyses of marine and terrestrial samples collected in August 2003 from Bylot Sound at Thule, Northwest Greenland, show that plutonium from nuclear weapons in the American B52 plane, which crashed on the sea ice in January 1968, persists in the environment.

The highest concentrations of plutonium are found in the marine sediments under the location where the plane crashed. The distribution of plutonium in the marine sediment is very inhomogeneous and associated with hot particles with activities found up to 1500 Bq $^{239,240}$Pu. Sediment samples collected in Wolstenholme Fjord north of the accident site show plutonium concentrations, which illustrates the redistribution of plutonium after the accident.

The total plutonium inventory in the sediments has been assessed based on systematic analyses considering hot particles. The inventory of $^{239,240}$Pu in the sediments within a distance of 17 km from the point of impact of the B52 plane is estimated at 2.9 TBq (1 kg). Earlier estimates of the inventory were approximately 1.4 TBq $^{239,240}$Pu.

Seawater and seaweed samples show increased concentrations of plutonium in Bylot Sound. The increased concentrations are due to resuspension of plutonium-containing particles from the seabed and transport further away from the area. Plutonium concentrations in seawater, seaweed and benthic animals in Bylot Sound are low but clearly above background levels.

All soil samples collected from Narssarssuk show accident plutonium with levels above background. Plutonium is very inhomogeneously distributed and associated with particles in the surface layers. Hot particles were found in soil with activities up to 150 Bq $^{239,240}$Pu.

Plutonium in the marine environment at Thule presents an insignificant risk to man. Most plutonium remains in the seabed under Bylot Sound far from man under relatively stable conditions and concentrations of plutonium in seawater and animals are low. However, the plutonium contamination of surface soil at Narssarssuk could constitute a small risk to humans visiting the location if radioactive particles are resuspended in the air so that they might be inhaled.

**ISSN 0106-2840**
**ISBN 87-550-3508-6**

**Group's own reg. no.:**
1400102-02

**Sponsorship:**
Ministry of the Environment

**Pages: 40**

Risø National Laboratory
Information Service Department
P.O.Box 49
DK-4000 Roskilde
Denmark
Telephone +45 46774004
bibl@risoe.dk
Fax +45 46774013
www.risoe.dk

# Contents

# 1 Introduction

In January 1968 a B52 plane from the US strategic Air Command caught fire and crashed on the sea ice in Bylot Sound about 12 km west of the Thule Air Base, Northwest Greenland. The plane carried four nuclear weapons. Part of the weapons plutonium was distributed over some square kilometres of the ice in the explosive fire that followed and radioactive particles were carried by wind in the direction of Narssarssuk approximately 7 km south of the crash site. Plutonium-contaminated ice was recovered and shipped back to the US, as was the plutonium-contaminated wreck. The underlying sea sediments received a fraction of the weapons plutonium when the sea ice melted the following summer and probably also during the accident as the impact caused the ice to break up (U.S. Air Force, 1970).

Previous sample collections in 1968, 1970, 1974, 1979, 1984, 1991 (Aarkrog 1971, 1977; Aarkrog et al., 1981, 1984, 1987, 1988, 1997) have estimated that the pollution remaining in the seabed in Bylot Sound by 1968 amounted to approximately 1.4 TBq $^{239,240}$Pu (about 0.5 kg), 0.025 TBq $^{238}$Pu, 4.6 TBq $^{241}$Pu and 0.07 TBq $^{241}$Am. Results from a sample collection in 1997 indicate, however, that previous estimates of the plutonium inventory may not have included radioactive particles in the seabed sediments in sufficient detail (Eriksson, 2002).

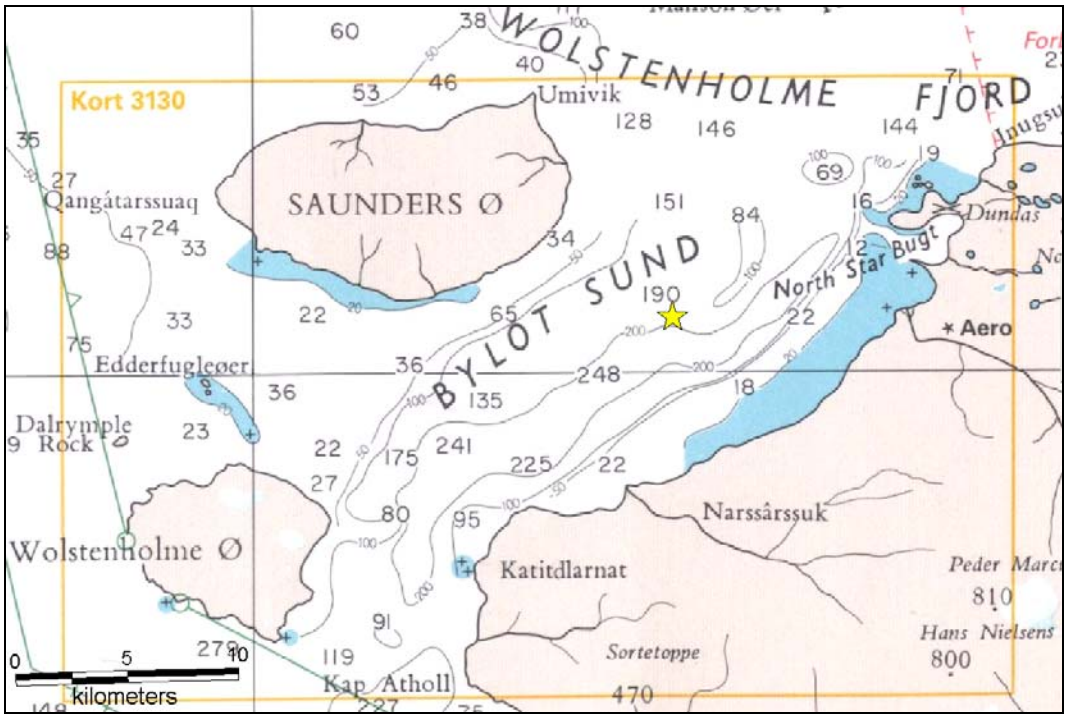


*Fig. 1.1. Map of Bylot Sound showing the site of the aircraft accident (star).*

The Thule Air Base is located at 76°30'N in NW Greenland near the former main living area for the Thule culture, an indigenous Inuit society surviving mainly through hunting marine mammals and birds. In 1953 the main part of the Inuit population was forced to move about 100 km further north to Qaanaaq because of expansion of the activities at the Thule Air Base. However, they still use the area for hunting marine mammals and birds. The rich marine benthic fauna, the basis for the important walrus population was intensively studied in 1939-1941 (Vibe, 1950).

Selected data from the sampling in 1997 has been published (Dahlgaard et al., 1999a,b; Eriksson et al., 1999; McMahon et al., 2000).

The present investigation was carried out during 2003-2005 and supported financially by the Danish Environmental Protection Agency, Ministry of the Environment.

# 2 Scope of Investigation

The present investigation was carried out to obtain updated information on the plutonium contamination after the airplane accident at Pittuffik (Thule Air Base) in January 1968. The investigation has focused on the following:

1. Investigation of plutonium in the marine sediments focusing on a quantitative assessment of radioactive particles (so-called hot particles)1
2. Sediment sampling in Wolstenholme Fjord north of the accident site in order to evaluate re-distribution of plutonium after the accident
3. Analyses of plutonium in seawater and seaweed in order to evaluate resuspension from the seabed
4. Investigate the transfer of plutonium and americium to benthic animals
5. Carry out acoustic mapping of the seabed sediments in Bylot Sound in order to improve the success rate of sediment sampling and the data on plutonium in the sediments
6. Investigate the fate of plutonium on land where data from previous investigations have shown presence of plutonium from the accident (collection of soil/peat profiles).

# 3 Sampling

The sampling was carried out in August 2003 from the investigation vessel 'Adolf Jensen' belonging to Pinngortitaleriffik (Grønlands Naturinstitut) in Nuuk. The participants from Risø National Laboratory in the sampling campaign were Research Specialist Henning Dahlgaard, Postdoc Mats Eriksson and Research Technician Svend K. Olsen. Participants from GEUS, the Geological Survey of Denmark and Greenland, were Senior Scientist Jørn Bo Jensen and Senior Advisor Peter Trøst Jørgensen, who carried out the sediment mapping. Skipper Flemming Heinrich was head of the 6-man crew of Adolf Jensen.

Some participants went on board the Adolf Jensen in Upernavik on 12 August with the equipment and two days later the remaining participants met the vessel in Thule. Sediment mapping using acoustic techniques was carried out in Bylot Sound during 14-19 August in parallel with collection of seaweed and soil samples using Adolf

1 Radioactive particles or hot particles are referred to as localized aggregations larger than about 0.5 μm containing radioactive atoms that give rise to an inhomogeneous distribution of radionuclides significantly different from that of the matrix background (IAEA Coordinated Research Project, G41003, 2000-2007, Radiochemical, chemical and physical characterisation of radioactive particles in the environment).

Jensen's rubber dinghy. Seaweed samples were collected from the coast at Narssarssuk south of the accident site, at Dundas, from the north coast of Wolstenholme Island and from the Eider Islands.

Seawater samples were collected during 18-21 August. Locations were chosen within the contaminated area in Bylot Sound and outside the contaminated area. A location south of Wolstenholme Island served as reference to background levels and locations north of Saunders Island served as expected low-level sites. Accident-related plutonium was expected to be found in near-bottom samples from Bylot Sound, but not in the other seawater samples. Three samples (170-200 litres) were taken at each location, a surface sample collected at 5 m below the surface and two deep samples at 15 and 35 m above the seabed. Seawater samples were passed through 0.45 µm Millipore filters to separate particles from seawater. Additional surface seawater samples were collected in Bylot Sound and at Dundas to determine the levels of the anthropogenic radionuclides $^{90}$Sr, $^{137}$Cs and $^{99}$Tc in the Thule area and compare with levels in a sample, which was collected at Illulissat some 1000 km SSE of Thule.

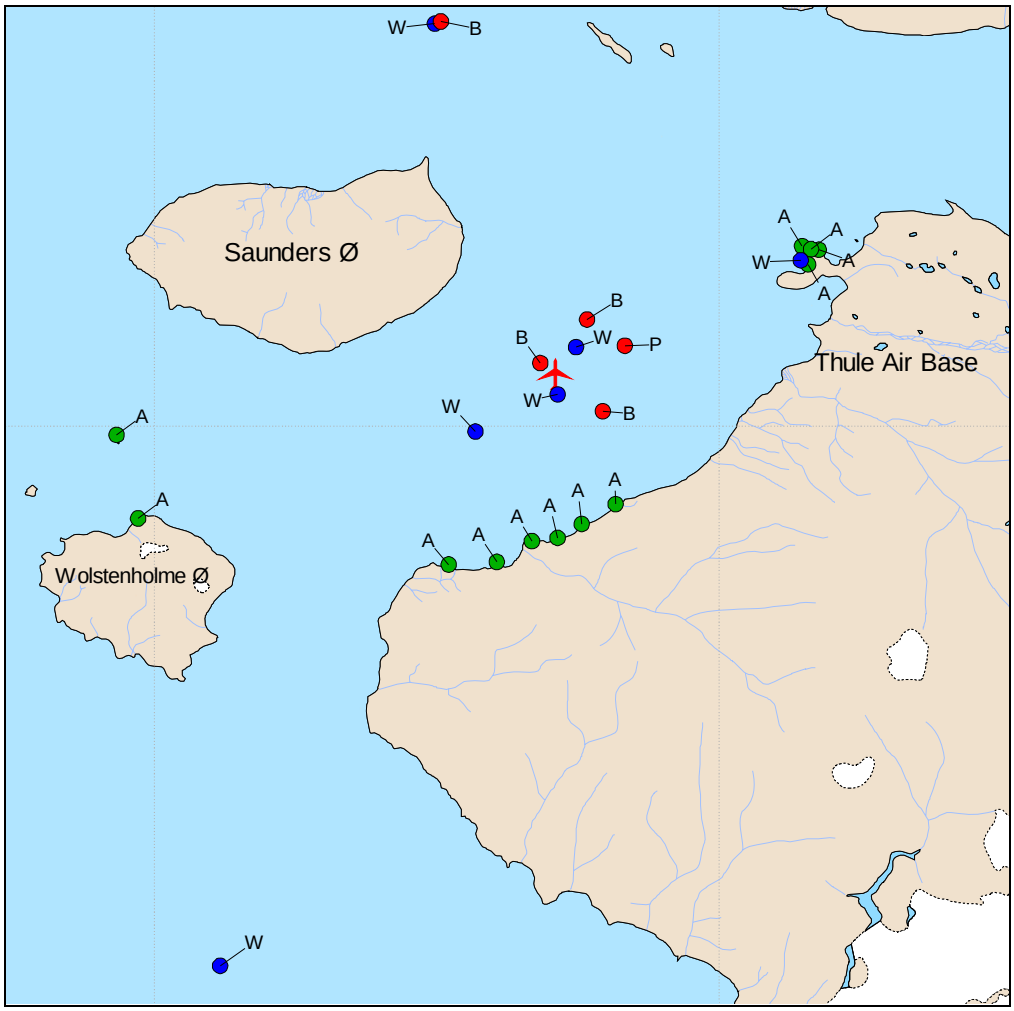


*Fig. 3.1. Sampling locations for seawater (W), seaweed (A), benthic fauna (B) and seal (P).*

Sediment cores were collected with a GEMAX gravity corer from 31 locations of which 10 were stations that had been sampled previously with success. All new

stations were selected based on information from the sediment mapping done by GEUS. Sampling at these new stations was a great success demonstrating that the time and money spent on the sediment mapping was very useful. Previous sampling campaigns have experienced significant problems with hard-bottom conditions prohibiting collection of sediment cores.

Benthic biota was collected along four 500-m transects using a Sigsbee trawl. Three of the trawl hauls were taken within a few kilometres from the accident site at 200-250 m while the fourth was taken 17 km north of the accident site at 135 m. The biota was taken from the trawl, cleaned initially and transferred to tubs where the animals were cleaned more thoroughly in seawater. Special focus was on collecting bivalves, whelks, shrimp and brittle stars; some small fish and squid were also collected. Shrimp, whelks, fish and squid were cleaned for a few hours in surface water. Bivalves and brittle stars were kept overnight in surface water to allow them to depurate. In Bylot Sound a seal was shot from which samples of flesh and liver were collected. Unfortunately, time did not allow for obtaining samples of birds, seal and walrus from local hunters.

Low levels of plutonium contamination on land were found at Narssarssuk in 1968 (US Air Force, 1970) and this area was therefore selected for collection of soil samples. Samples were collected from 8 locations and at each location 3 soil profiles were taken within an area on one square metre. Thus 24 soil profiles were obtained from this area.

Sampling locations are shown in Figs. 3.1 and 3.2.

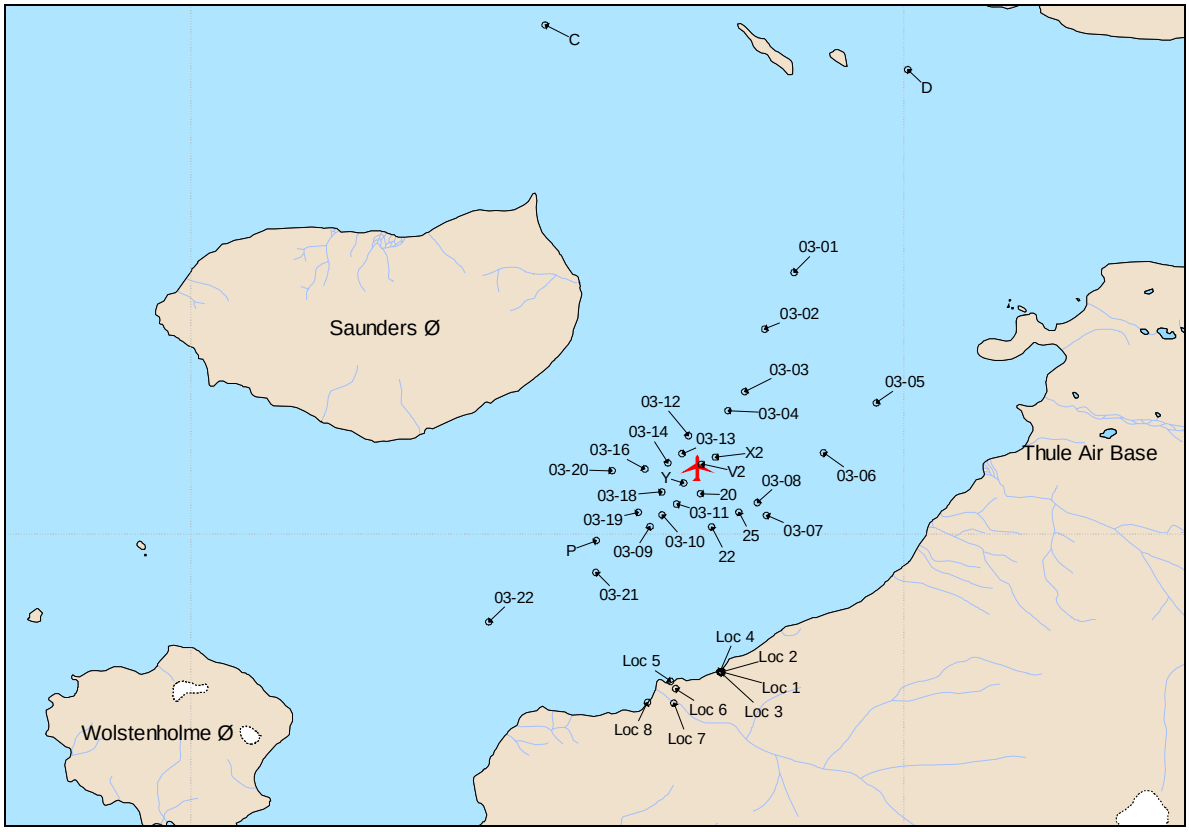


*Fig. 3.2. Sampling locations for marine sediment in Bylot Sound and Wolstenholme Fjord, and for soil at Narssarssuk (Loc. 1-8).*

# 4 Acoustic mapping

Sediment mapping was carried out by GEUS from Adolf Jensen and included recording of bathymetry and characterisation of the seabed. Bathymetry, sediment profiles and surface seabed mosaic were mapped by chirp and side-scan sonar. The seismic grid used for the sediment mapping is shown in Fig. 4.1 and the bathymetry in Fig. 4.2.

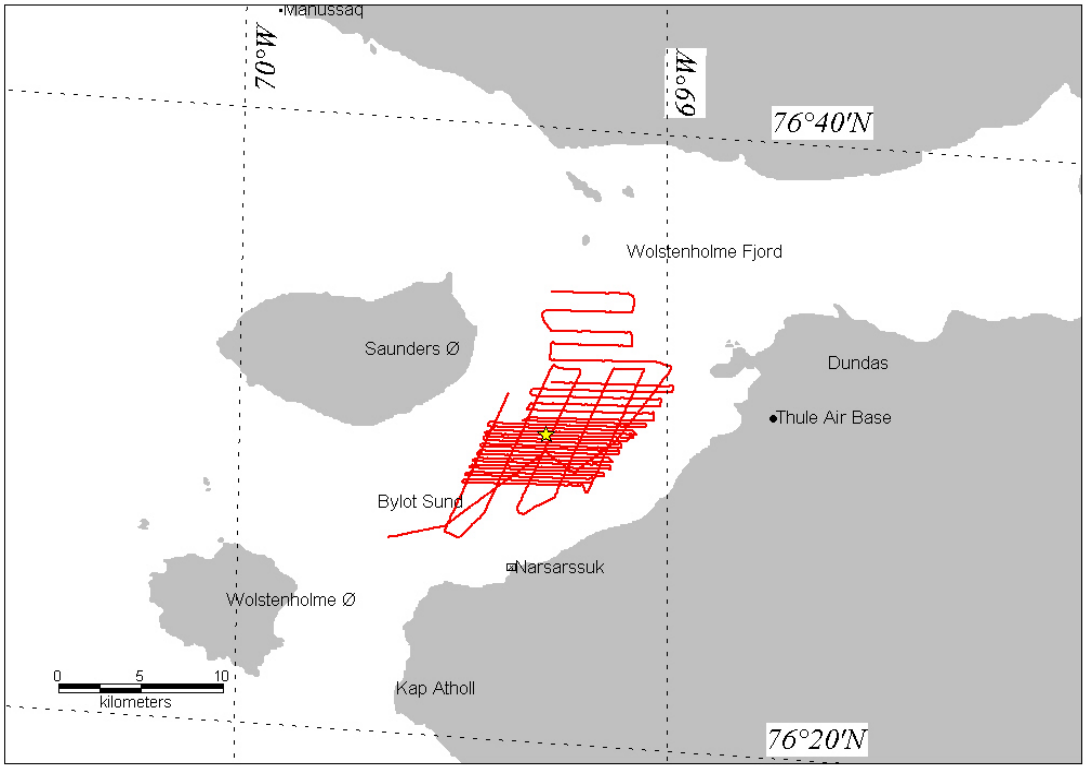


*Fig. 4.1. Seismic grid for the sediment mapping in the central part of Bylot Sound. The point of impact of the B52 plane is marked with a star.*

The following conclusions were drawn from the acoustic mapping, which is reported in detail by Jensen (2003):

- The bathymetry data show more details of the water depths in Bylot Sound than was known previously
- The most pronounced morphological features are a north-south going ridge in Bylot Sound with water depths less than 100 m and a southern channel partitioned in a northern and southern basin with water depths down to 250 m.
- Five characteristic seabed types reflect the sedimentation pattern in the area investigated:
  - Relatively soft bottom with accumulation of sediment
  - Hard bottom with some accumulation of sediment
  - Transition zone between hard residual bottom and sedimentation bottom
  - Hard residual bottom dominated by erosion

- Bottom with scour marks from icebergs which frequently strike the bottom and disrupt the sedimentation layers

– The sediment accumulation areas are interesting for sampling in connection with plutonium since these areas represent sinks for fine-grained material in Bylot Sound

– The predominant bottom type in the area near the point of impact of the B52 aircraft is a transition zone between hard residual and accumulation bottom. It is important that samples for plutonium are collected from the accumulation bottom.

– The residual bottom and the bottom with iceberg scouring marks are not interesting for sediment sampling since these bottoms do not represent recent sediments and are often disturbed.

– The screening of the side-scan data showed a nearly rectangular object of about 5x5 m, which apparently has a depression in the middle. The object may be a foreign body on the seabed.

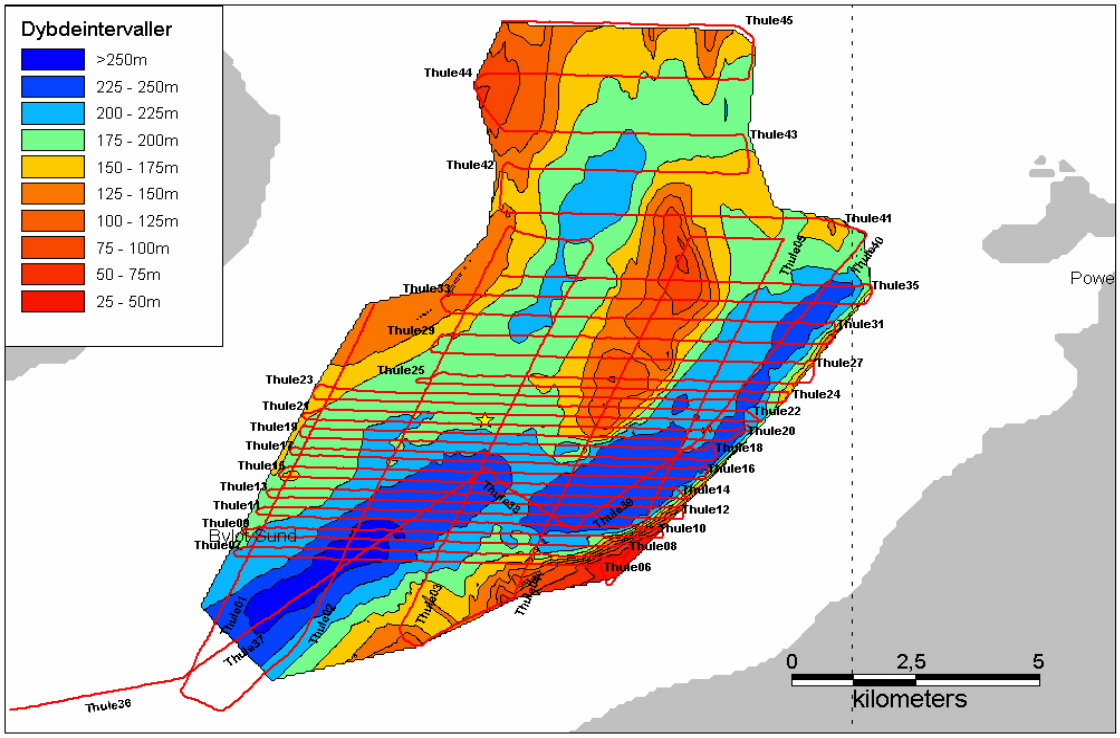


*Fig. 4.2. The bathymetry recorded in the area of investigation. The point of impact of the B52 plane is marked with a star.*

# 5 Analytical

## 5.1 Sample preparation

Sample preparation is needed to transform the sample into a reference condition or to reduce the mass and volume prior to chemical separation and/or radiometric

analysis. Standard preparation methods used include drying at 105 ºC, freeze drying and ashing at 450 ºC. Soil and sediment samples were sieved by removing stones larger than 1.5 mm and in addition soil samples were ashed in order to eliminate the organic content, which varied from 5 to 50%. The methods applied are shown in Table 5.1.1.

*Table 5.1.1. Methods used for preparation of samples prior to radionuclide analysis.*

| Sample type | Drying | Freeze drying | Sieving | Ashing |
|---|---|---|---|---|
| Seawater, filter | | | | X |
| Seaweed | X | | | X |
| Sediment | | X | X | |
| Soil | | X | X | X |
| Benthic biota | | X | | X |
| Seal | | X | | X |

## 5.2 Radiochemical analyses

Gamma-emitting radionuclides ($^{137}$Cs and naturally occurring radionuclides) were analysed by direct measurements of samples after mass reduction. For radionuclides emitting alpha and beta particles such as plutonium isotopes, $^{241}$Am, $^{90}$Sr and $^{99}$Tc, chemical procedures are required to separate the radionuclides in question from the sample matrix and interfering radioisotopes prior to measurement. Measurements of alpha-emitting radionuclides were made by alpha spectrometry using Si detectors and beta-emitting radionuclides were measured by low-level gas-flow Geiger-Müller counters.

The radiochemical analyses of plutonium, americium, strontium and technetium were based on procedures developed by Chen (2001, 2002). Chemical yields were determined using radioactive tracers.

Due to the low concentrations of plutonium and technetium in seawater at Thule, large samples of 175 litres were collected for determination of these radionuclides. Other sample types were collected in kilogram quantities. Typical amounts of sample material used for the different analyses are listed in Table 5.2.1.

*Table 5.2.1. Typical amounts of sample used for analyses (kg).*

| Sample type | Pu-isotopes/$^{241}$Am | Gamma ($^{137}$Cs+other) | $^{90}$Sr | $^{99}$Tc |
|---|---|---|---|---|
| Seawater | 175 | 50 | 50 | 175 |
| Seaweed | 0.01 (ash) | 0.15 (ash) | Na | 0.01 (dry) |
| Sediment | 0.001 (dry) | 0.015 (dry) | Na | Na |
| Soil | 0.001 (ash) | 0.015-0.25 (ash) | Na | Na |
| Benthic biota | 0.0003-0.002 (ash) | Na | Na | Na |
| Seal | 0.01 (ash) | 0.07 (ash) | Na | Na |

Na – not analysed

The chemical procedure for analysis of plutonium was further improved compared to that described by Chen (2001). The improved procedure gives more complete dissolution of plutonium in the samples, higher chemical yields, higher decontamination for interfering naturally occurring radionuclides (isotopes of uranium, thorium and polonium) and better energy resolution for the alpha-spectrometric measurements. The performances of the previous and the improved chemical procedure were compared by analysing two sediment samples using the two procedures. The results of the two methods agreed well with each other, within

5%. Furthermore, the improved procedure was used for results on plutonium in fish and sediment submitted to international intercomparisons (IAEA 2004, 2005), which confirmed the good analytical quality of the results.

Special analytical procedures were applied to investigate the leachability of plutonium and americium from radioactive particles found in samples of soil and sediment. The chemical form of the accident plutonium at Thule is $PuO_2$, which is known to be difficult to dissolve completely (Eriksson, 2002). A number of hot particles were subject to a series of tests including successive leachings with aqua regia at high temperatures. The tests demonstrated that treatment of soil and sediment samples with aqua regia at 150 °C for 4 h dissolved about 99% of both plutonium and americium from the samples. This procedure was therefore used for the chemical analyses mentioned above.

## 5.3 Gamma screening

Gamma screening was carried out on selected sediment cores and all surface soil samples. Usually, analyses are performed on small aliquots selected from homogenised samples assuming that the aliquots are representative for the entire sample. The purpose of the screening was to account for all hot radioactive particles in the sample material and obtain an estimate of the amount of radioactivity associated with these particles. Plutonium isotopes do not emit gamma radiation but $^{241}Am$ does, and this radionuclide is a decay product of $^{241}Pu$ which at the time of the accident in 1968 accounted for about 2‰ by mass of the weapons plutonium. The gamma radiation from $^{241}Am$ is therefore an indicator of accident plutonium in sediment and soil. The screening was carried out on relatively small aliquots typically in the range of 5-15 g avoiding severe attenuation in the samples of the low-energy gamma radiation from $^{241}Am$. The screening estimates of activity in the samples assume uniform distributions of radioactivity throughout the samples, which is clearly not the case if a sample contains one or more hot particles. Therefore, a number of sediment cores were selected for a second screening after re-packing all sub-samples and thus re-distributing possible hot particles in the aliquots. Comparison of the activity estimates of the sediment cores screened twice gives an indication of the accuracy of the activity estimates associated with the screening.

About 2500 gamma analyses were carried out covering about 43 kg dw sieved soil and 23 kg dw sieved sediment. The gamma-spectrometry measurements were made using high-purity Ge detectors.

## 5.4 Sediment size distribution

In order to characterise the sediment and determine the distribution of plutonium on different grain size fractions, a number of sediment samples were sorted by wet sieving.

The wet sieving was done using sieves of size 0.4, 0.125 and 0.06 mm. Four different fractions were collected (>0.4 mm, 0.125-0.4 mm, 0.06-0.125 mm and <0.06 mm) and analysed for Pu-isotopes. Each size fraction was analysed in five 1-g replicates.

### 5.5 Analytical quality

The reliability of Risø's analytical results on plutonium isotopes, $^{241}$Am and other radionuclides is tested regularly by participation in international intercomparison exercises on analyses of radionuclides in environmental samples. For the last 10 years Risø has participated in about 2 international intercomparisons per year many of which have included plutonium and americium in environmental samples. The intercomparisons demonstrate good quality of Risø's analytical results (Fogh 2000; Fogh et al. 2002; IAEA 1999, 2000a, 200b, 2001, 2004, 2005; Nielsen, 1996, 2004; Outola 2005).

# 6 Results

## 6.1 Sediments

Sediment samples were collected from 27 locations in Bylot Sound, 2 locations in Wolstenholme Fjord north of Bylot Sound and 2 locations in Baffin Bay about 30 km southwest of Wolstenholme Island. Water depths at the locations were in the range 185-259 m in Bylot Sound, 90-120 m in Wolstenholme Fjord and 645-690 m at the two locations in Baffin Bay. The locations in Bylot Sound and Wolstenholme Fjord are shown in Fig. 3.2.

The sediment cores were generally collected to 30 cm depth and sliced in 3-cm sections. From one to three sediment cores were collected at each location resulting in more than 500 sediment samples. The samples ranged in size from tens to hundreds of grams of dry weight sieved material. Nearly all sediment cores were analysed by gamma spectrometry for gamma emitting radionuclides and about 300 samples were analysed by radiochemical procedures followed by alpha spectrometry for plutonium and americium. The gamma analyses involved standard long-term analyses of aliquots from the samples as well as short-term screening analyses, which systematically included all material in the sediment cores analysed in 5-15 g aliquots in order to account for all hot particles.

The radiochemical procedures and alpha-spectrometry measurements were carried out on 1-g aliquots of the samples permitting identification of $^{238}$Pu, $^{239,240}$Pu and $^{241}$Am. Profiles of $^{239,240}$Pu concentration (Bq kg$^{-1}$ dw) with depth are given for the locations investigated in Figs. 6.1.1-6.1.3. Most of the profiles are based on results of the alpha-spectrometry measurements but in some cases $^{241}$Am concentrations determined by gamma spectrometry have been used to estimate the plutonium profiles. The concentrations of $^{239,240}$Pu vary considerably across the locations. The maximum concentration at each location ranges from 2 Bq kg$^{-1}$ at location Ny-5 (Fig. 6.1.1) to 5400 Bq kg$^{-1}$ at location 03-20 (Fig. 6.1.2). Location Ny-5 is a background site some 50 km from the contamination in Bylot Sound and the concentrations of $^{239,240}$Pu in the sediments here are low and due to global fallout from nuclear weapons testing. Location 03-20 in Bylot Sound is 3 km from the point of impact of the B52 plane and the maximum concentration of $^{239,240}$Pu in the sediment from this profile is clearly due to the presence of a hot particle in the 0-3 cm layer. The maximum concentrations across the locations occur at varying depths in the sediments ranging from the surface layer to about 15 cm under the surface. The profiles illustrate the non-uniform distribution of plutonium with depth in the sediments and the effects of the biological mixing due to the benthic fauna.

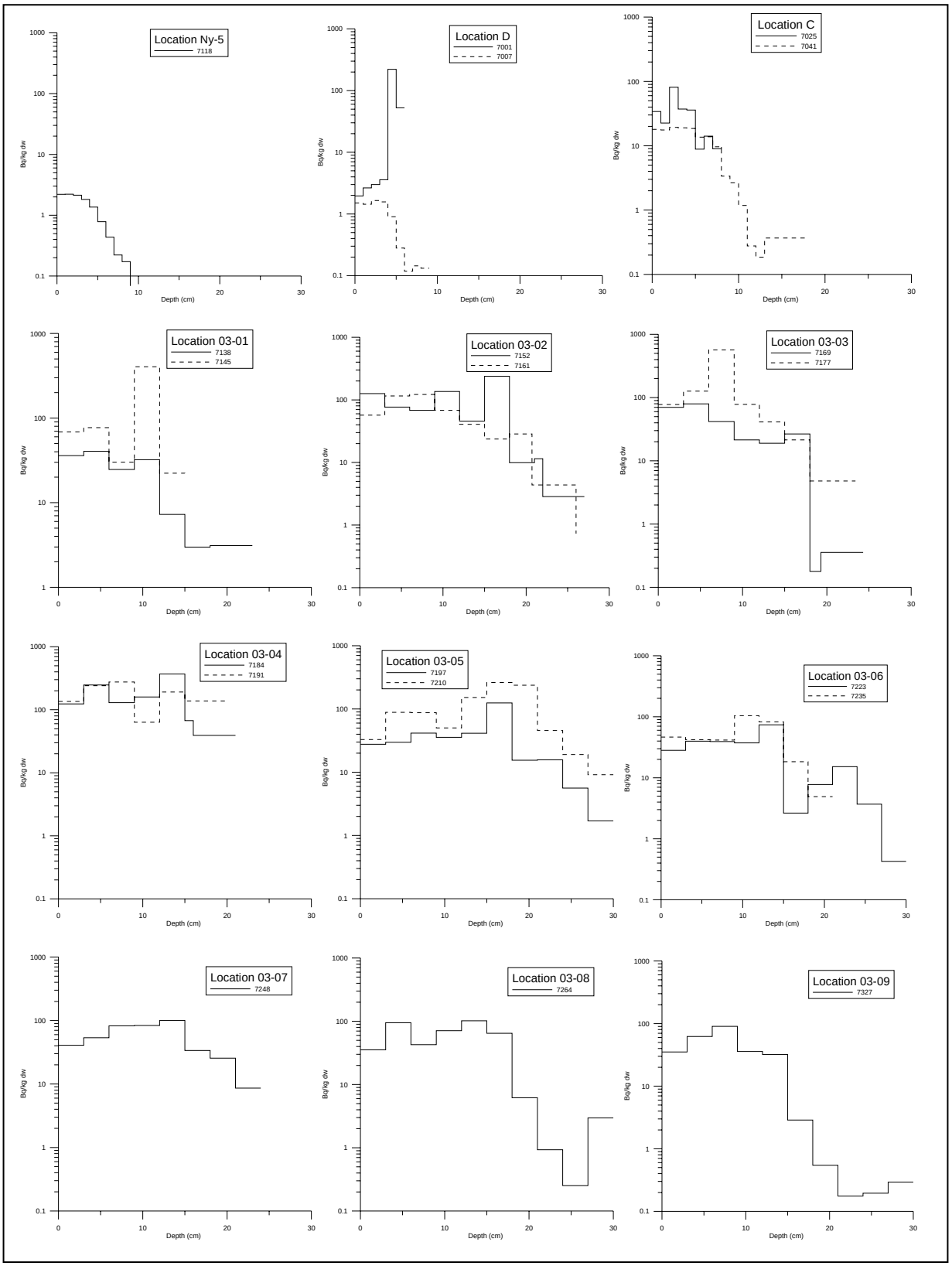


*Fig. 6.1.1. Concentrations of $^{239,240}$Pu in sediment cores from Bylot Sound (Bq kg$^{-1}$ dw).*

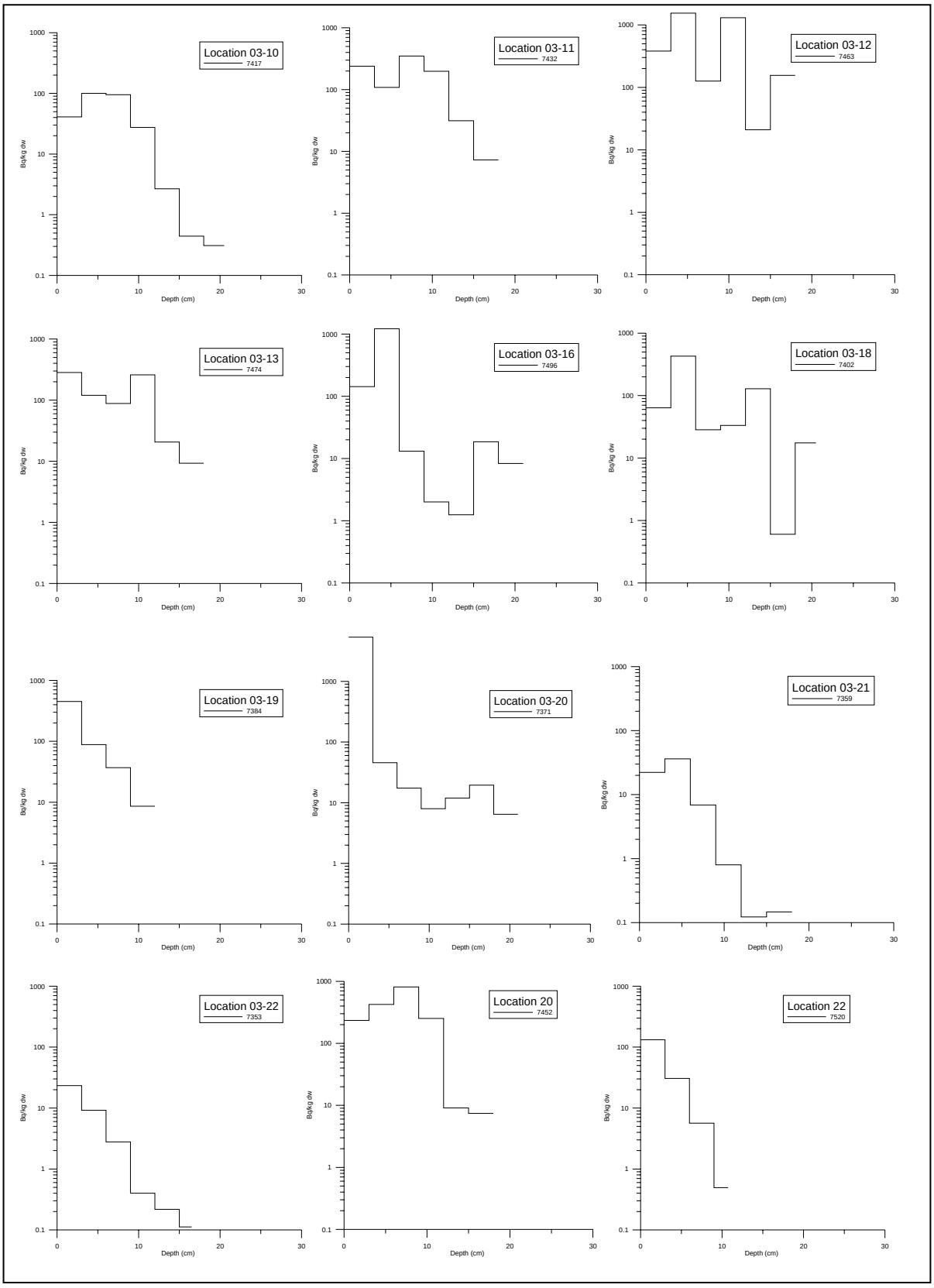


*Fig. 6.1.2. Concentrations of $^{239,240}$Pu in sediment cores from Bylot Sound (Bq kg$^{-1}$ dw).*

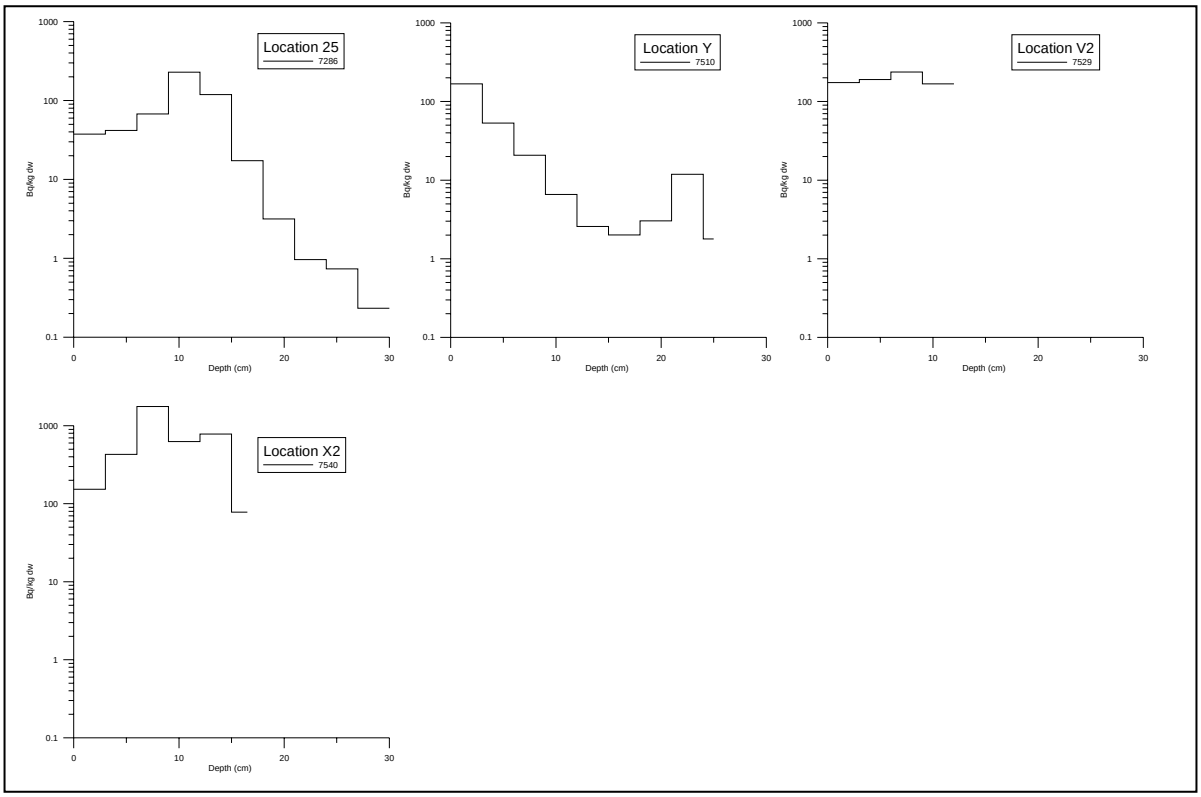


*Fig. 6.1.3. Concentrations of $^{239,240}$Pu in sediment cores from Bylot Sound (Bq kg$^{-1}$ dw).*

Concentrations of $^{238}$Pu were detected in 150 of the sediment samples analysed by alpha spectrometry. The activity ratios of $^{238}$Pu to $^{239,240}$Pu show clear differences between locations. The activity ratios from the sediment samples in Bylot Sound are characteristic of accident plutonium and show little variability with an average value of 0.013±0.005 (1 SD, n=127). The average activity ratio at the background site Ny-5 is 0.035±0.020 (1 SD, n=9), which is close to the typical value of the activity ratio in global fallout of 0.04 (Perkins and Thomas, 1980). The activity ratios at the locations C and D in Wolstenholme Fjord some 17 km from the point of impact are intermediate with a mean of 0.029±0.024 (1 SD, n=14). Concentrations of $^{241}$Am were detected in 37 sediment samples, and also here the activity ratios show clear differences by location. The activity ratios of $^{241}$Am to $^{239,240}$Pu at location Ny-5 show a mean value of 0.46±0.13 (1 SD, n=10) while the ratios in samples from Bylot Sound and Wolstenholme Fjord show a mean value of 0.15±0.05 (1 SD, n=27).

The depositions of $^{239,240}$Pu on the seabed in terms of total activity by area are shown in Table 6.1.1 in which the locations are sorted by distance from the point of impact of the B52 plane. The comparison of the depositions obtained from the different analytical methods shows considerable variation within locations thus illustrating the inhomogeneous distribution of plutonium in the sediments. In some cases hot particles were included in the 1-g aliquots analysed by chemical separation and alpha spectrometry, e.g. in the sediment core from location 03-16 where the aliquot from the 3-6 cm layer contained a $^{239,240}$Pu activity of about 1 Bq. In other cases the standard gamma analyses of typical 15-g aliquots contained hot particles, e.g. in the sediment core from location 03-20 where an aliquot from the 0-3 cm layer contained a $^{239,240}$Pu activity of about 100 Bq. The systematic screening of all material from the sediment cores also in several cases revealed hot particles, which were not identified from the alpha-spectrometric analyses or the standard gamma analyses. The most obvious example of this is from the sediment core from location 03-18

where an aliquot from the 3-6 cm layer contained a $^{239,240}$Pu activity of about 1500 Bq.

*Table 6.1.1. Total deposition of $^{239,240}$Pu (kBq m$^{-2}$) at locations in Bylot Sound determined by three analytical methods: 1) radiochemical separation and alpha spectrometry, 2) standard gamma spectrometry and 3) screening by gamma spectrometry. Distances are shown from the locations to the point of impact of the B52 aircraft.*

| Location | Distance (km) | Alpha (kBq m$^{-2}$) | Gamma standard (kBq m$^{-2}$) | Gamma screening (kBq m$^{-2}$) |
|---|---|---|---|---|
| V2 | 0.2 | 17.5 | 19.7 | 25.4 |
| Y | 0.7 | 6.6 | 4.4 | 14.4 |
| 03-13 | 0.7 | 6.7 | 19.2 | 35.5 |
| X2 | 0.8 | 21.7 | 91.3 | 61.8 |
| 20 | 1.0 | | 36.0 | |
| 03-12 | 1.2 | 14.7 | 86.1 | 35.6 |
| 03-11 | 1.5 | | 19.5 | |
| 03-18 | 1.6 | 16.9 | 13.1 | 223.0 |
| 03-16 | 1.9 | 37.6 | 23.6 | |
| 03-10 | 2.1 | 6.0 | 10.4 | 31.7 |
| 22 | 2.2 | 3.4 | 1.7 | 6.0 |
| 25 | 2.2 | 9.9 | 13.4 | |
| 03-04 | 2.4 | 13.3 | 25.6 | 40.2 |
| 03-04 | 2.4 | | 25.5 | |
| 03-08 | 2.5 | 8.1 | 11.5 | 12.5 |
| 03-19 | 2.7 | | 12.3 | |
| 03-09 | 2.7 | 5.9 | 5.8 | 5.6 |
| 03-07 | 3.1 | | 8.9 | |
| 03-20 | 3.1 | 2.8 | 120.1 | 23.5 |
| 03-03 | 3.3 | 5.3 | 9.6 | 9.8 |
| 03-03 | 3.3 | | 20.2 | |
| 03-06 | 4.6 | 5.2 | 7.7 | 13.9 |
| 03-06 | 4.6 | | 6.7 | |
| 03-21 | 5.3 | 1.7 | 3.8 | |
| 03-02 | 5.6 | 14.7 | 11.1 | 18.9 |
| 03-02 | 5.6 | | 9.5 | |
| 03-05 | 6.9 | 7.0 | 10.9 | 12.2 |
| 03-05 | 6.9 | | 21.0 | |
| 03-01 | 7.9 | 3.1 | 3.8 | |
| 03-01 | 7.9 | 4.1 | 14.5 | 11.9 |
| 03-22 | 9.4 | 1.0 | 2.2 | |
| D | 16.4 | 2.6 | 0.4 | |
| D | 16.4 | 0.1 | | |
| C | 17.0 | | 1.7 | |
| C | 17.0 | 1.0 | 1.1 | |
| Ny-5 | 47.8 | 0.05 | 0.06 | |
| 1412 | 56.1 | | 0.01 | |

A rough view of the activity distribution in Bylot sound sediments may be obtained by plotting the number frequency of measured $^{241}$Am activities in sediment samples of size 5-15 g. Such a plot is shown in Fig. 6.1.4. It may be anticipated that the activity distribution of particles in the sediments is log normal. The distribution has

a geometric mean activity of 0.27 Bq $^{241}$Am, which corresponds to about 2 Bq $^{239,241}$Pu.

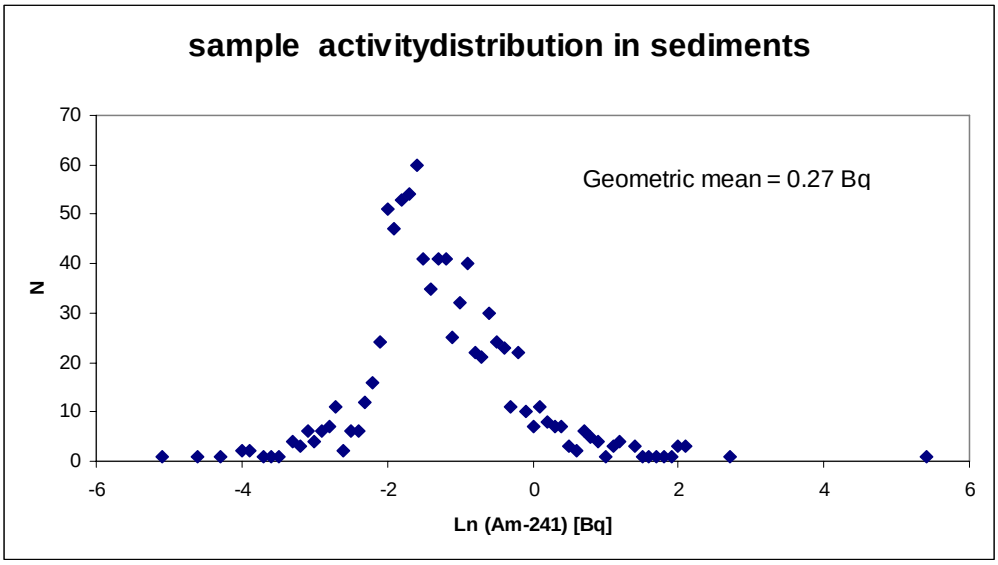


*Fig. 6.1.4. Activity distribution of $^{241}$Am in sediment samples.*

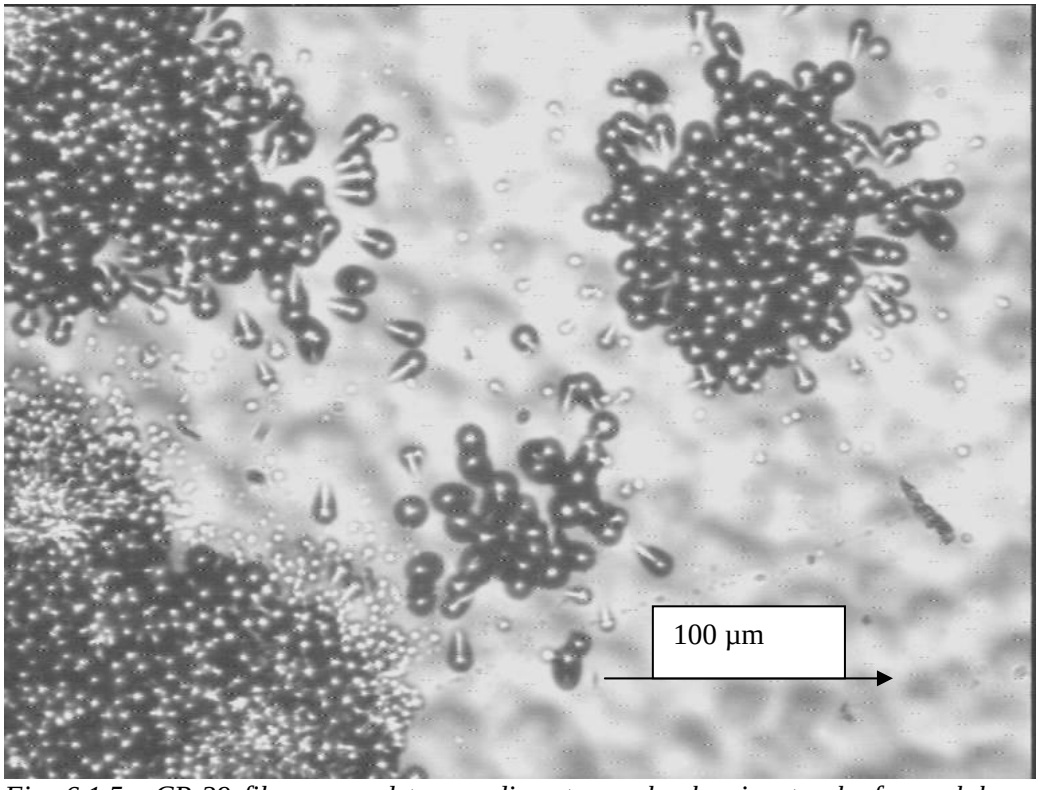


*Fig. 6.1.5. CR-39 film exposed to a sediment sample showing tracks from alpha particles due to plutonium and americium.*

The isolation of individual (larger, several tens of micrometer) Pu-particles from the sediments indicate that they are usually coated with some material but seldom associated to other particles. Evidence from this comes from many of the track etch films used to localize the individual particles. If mainly associated to other particles, shadow effects due to shielding by the sedimentary matter of emitted alpha particles should have been seen frequently but this has not been the case. Fig 6.1.5 shows a typical example of a track etch film (CR-39) exposed to a sediment sample.

### 6.1.1 Size fractionation

The upper 12 cm of sediments from four sediment cores were subject to wet sieving in order to evaluate the activity distribution of plutonium among the different size fractions. Of the four cores selected two were from station P while the two other cores were collected at stations 03-14 and 20, respectively. Fig. 6.1.1.1 shows the distribution of mass by grain size for the four samples, and Fig. 6.1.1.2 shows the corresponding distribution of $^{239,240}$Pu concentration by grain size.

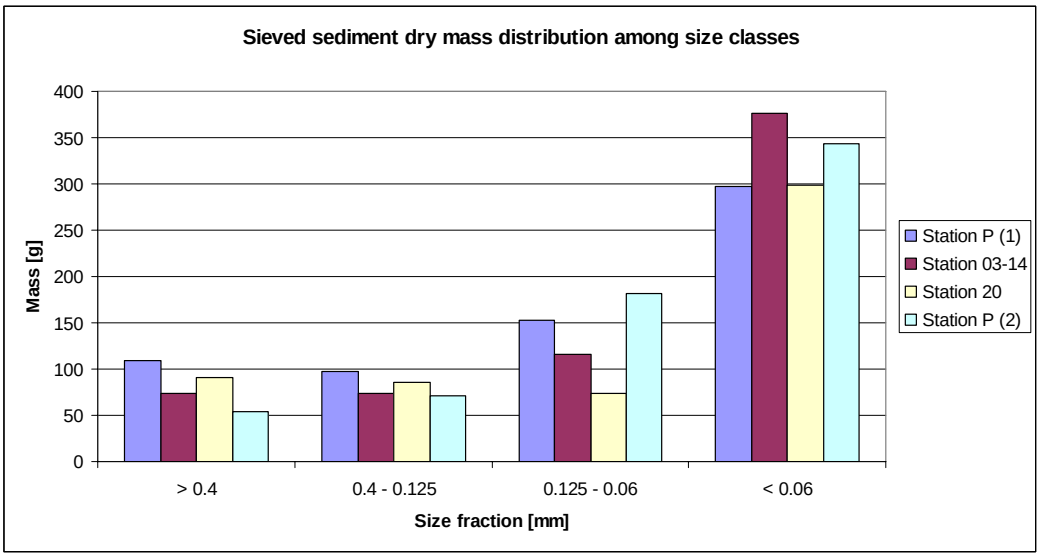


*Fig. 6.1.1.1. Distribution of mass by grain size for four sediment samples.*

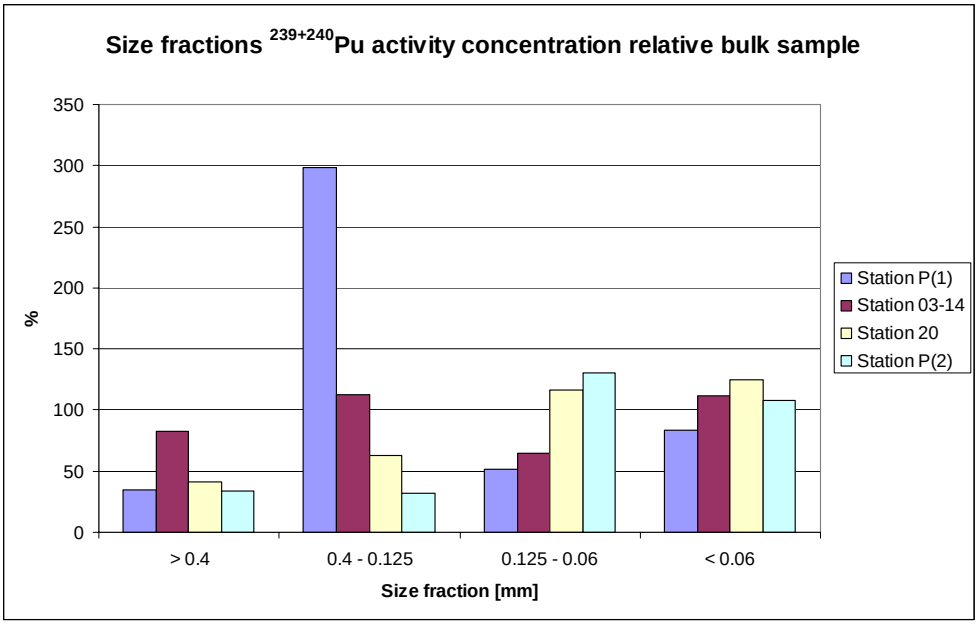


*Fig. 6.1.1.2. Distribution of $^{239,240}$Pu concentration between grain-size classes relative to concentration in bulk (unfractionated) sediment.*

Although the activity concentration does not change much over the various size classes the major part of the sediment mass is generally found in the finer fractions which therefore hold most of the Pu activity seen here. The distribution of plutonium among the sediment size classes is of importance for the resuspension of bottom

sediments and their transport away from the area. Analysis of plutonium isotope signature in the suspended matter close to the bottom in Bylot Sound indicates that the elevated levels are due to resuspended sediments containing accident plutonium; cf. Section 6.3.

The distribution of plutonium among the various size classes seen here from stations 03-14, 20 and P(2) may be representative for samples where the occurrence of larger hot particles is low. If particles with high plutonium content are present, their distribution among the sediment size classes will dominate the plutonium distribution as is the case for the sample from station P(1).

### 6.1.2 Plutonium inventory in sediments

The total inventory of $^{239,240}$Pu in the marine sediments was estimated from the results of the standard gamma and screening analyses of the sediment cores. Only activities of $^{241}$Am above the detection limit have been considered, which means that the concentrations determined from this procedure are slightly underestimated due to the relatively short measurement times for the screening analyses (about 2000 sec). The lengths of the sediment cores at some stations, notably station V2, were insufficient to collect the entire sediment depth profile, which further underestimates the total inventory somewhat. Samples collected from previous sampling expeditions at station V2 have shown depositions in the range of 50-100 kBq m$^{-2}$ of $^{239,240}$Pu while at this time only about 20 kBq m$^{-2}$ was found.

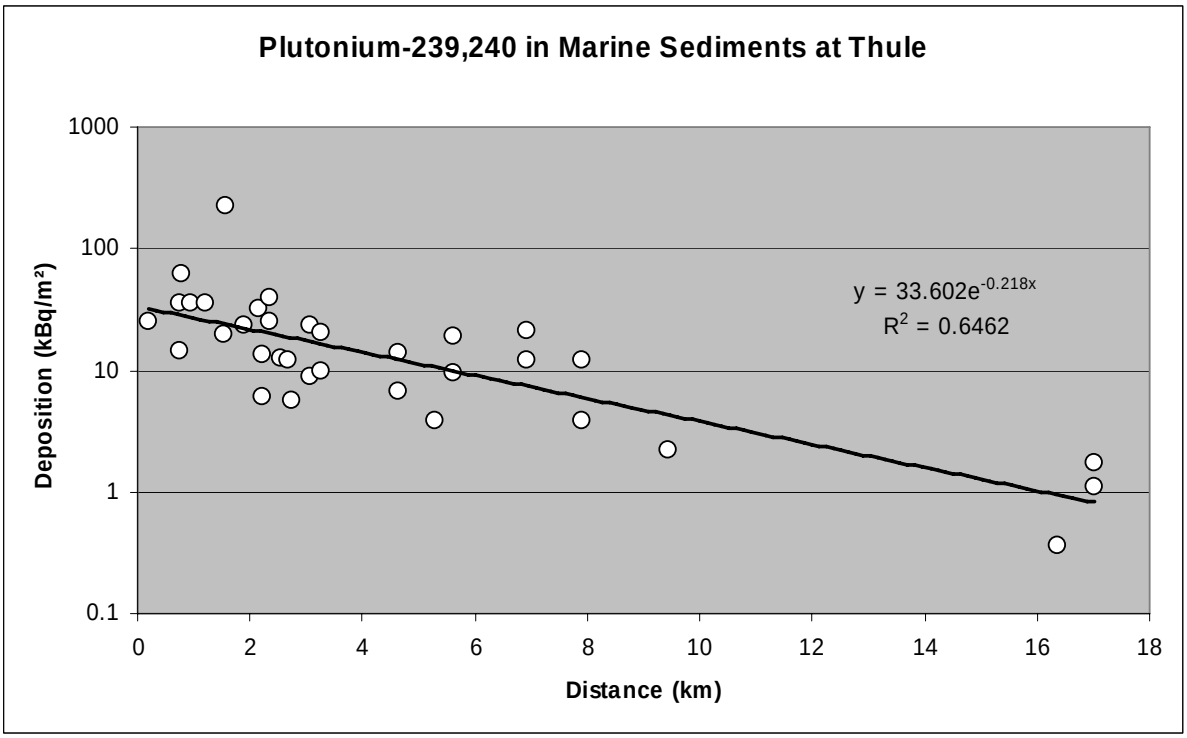


*Fig. 6.1.2.1. Deposition of $^{239,240}$Pu (kBq m$^{-2}$) at locations in Bylot Sound shown by distance from the point of impact of the B52 plane. An exponential function is fitted to the data as indicated.*

The $^{239,240}$Pu concentrations were calculated from the measured $^{241}$Am data using the ratio $^{241}$Am/$^{239,240}$Pu of 0.17 as found from analyses of hot particles. The ratio from sediment samples from Bylot Sound shows a mean value of 0.15 ±0.05 (1 SD, n=27).

The deposition of $^{239,240}$Pu in the marine sediments versus distance from the impact point in Bylot Sound may be approximated with an exponential function. The data is from Table 6.1.1 and Fig. 6.1.2.1 shows the deposition of $^{239,240}$Pu as a function of distance out to 18 km from the point of impact with a regression line fitted to the data. Integrating the exponential function over an area up to 17 km from the point of impact assuming that the distribution depends on distance only yields a total $^{239,240}$Pu inventory of 2.9 TBq. In agreement with procedures used from previous assessments it is assumed that land covers 60% of the area at distances above 7.5 km. Based on the regression fit, the 95% confidence interval for the inventory of 2.9 TBq is from 1.4 to 6.0 TBq. Alternatively, the data may be approximated with a power function but this does not improve the correlation and the calculated inventory is about the same, 2.7 TBq.

The fallout background out to 17 km from the point of impact amounts to about 0.05 TBq, which is less than 2% of the total $^{239,240}$Pu inventory. The observed distribution of plutonium in Bylot Sound is further illustrated in Fig. 6.1.2.2, showing the locations for sediment sampling and the deposition values by colour code. The circles on the map indicate distances of 4, 8, 12 and 16 km from the point of impact of the B52 plane.

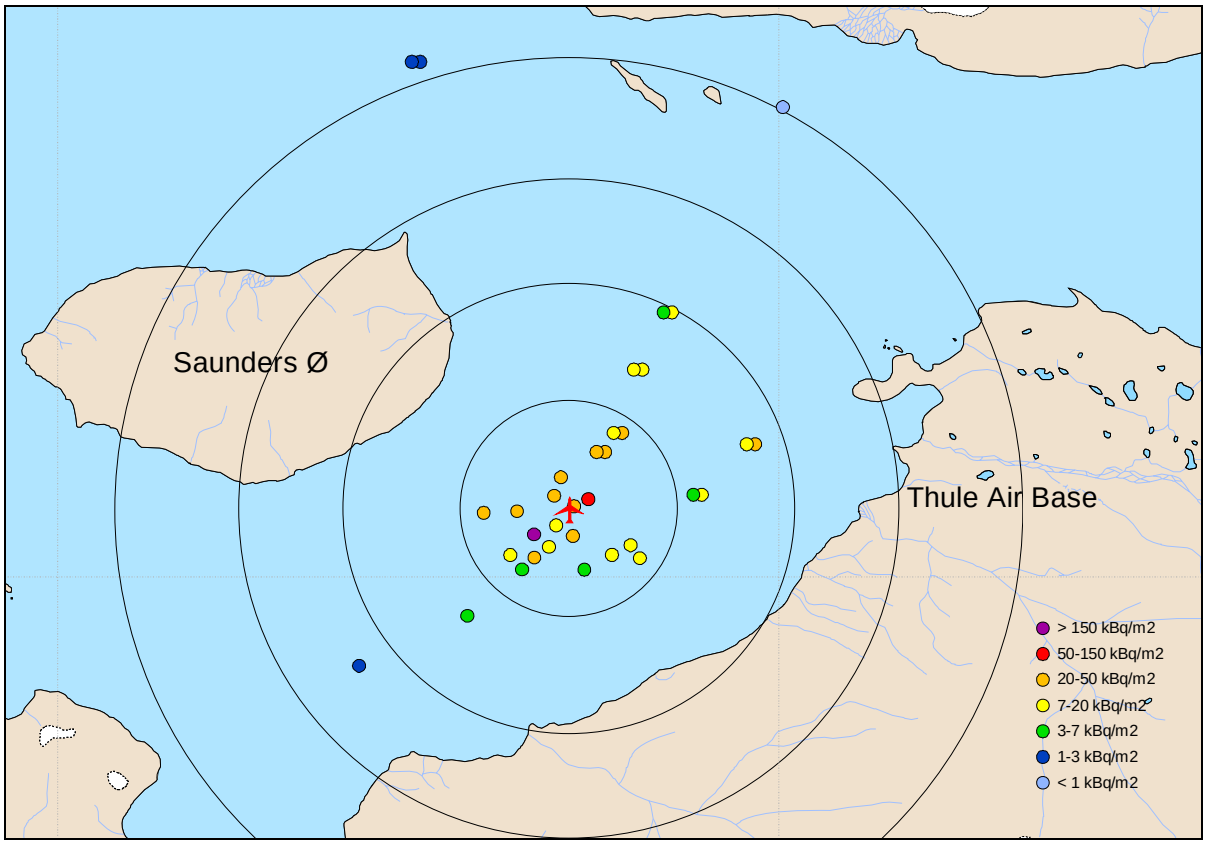


*Fig. 6.1.2.2. Locations for sediment sampling in Bylot Sound and observed values of deposition given by colour code. The circles indicate distances of 4, 8, 12 and 16 km from the point of impact of the B52 plane.*

Sediment was sampled in Bylot Sound at locations, which were selected from the acoustic mapping to be suitable for that purpose, i.e. areas with accumulation of sediment and not areas dominated by erosion. The above calculation does not consider the presence of hard bottoms dominated by erosion and with little or no accident plutonium. Therefore, the calculation tends to overestimate the inventory somewhat.

The systematic screening has covered the sediment cores collected and ensured that all hot particles in the samples have been considered for estimating the plutonium inventory. The resulting inventory estimated at 2.9 TBq of $^{239,240}$Pu supports the finding by Eriksson (2002) that previous investigations might have underestimated the total inventory in the marine sediments by not systematically considering the contribution from hot particles. Previous investigations concluded that the $^{239,240}$Pu in the marine sediments at Thule amounted to approximately 1.4 TBq, cf. Section 1. However, the inventory remains associated with considerable uncertainty due to the inhomogeneous distribution of plutonium in the sediments from which the number of samples is limited for practical reasons and in which relatively few hot particles contain an important part of the radioactivity.

## 6.2 Seawater

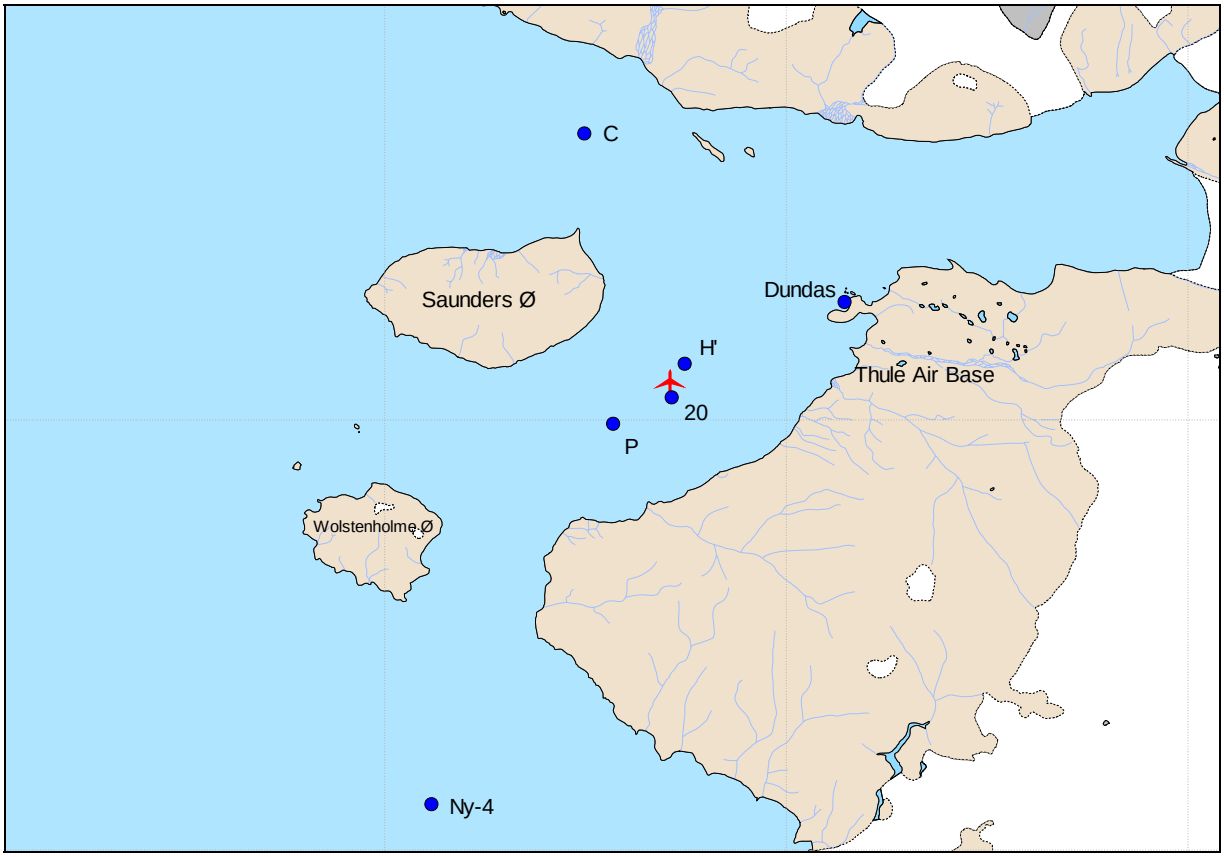


*Fig. 6.2.1. Locations for sampling of seawater in the Thule area, marked with filled circles and station names.*

Seawater samples were collected at 6 locations in the Thule area, three locations (stations P, 20 and H') in Bylot Sound, one location (station C) north-east of Saunders Island, one location (station Ny-4) south of Wolstenholme Island and one location at Dundas (station Dundas). The locations are shown in Fig. 6.2.1. Furthermore, a seawater sample was collected at Illulissat some 1000 km south of Thule.

Analyses of $^{238}$Pu, $^{239,240}$Pu and $^{241}$Am were made on samples from the stations C, H', 20, P and Ny-4 while analyses of $^{90}$Sr, $^{137}$Cs and $^{99}$Tc were made on samples from stations 20, Dundas and Illulissat. The concentrations of $^{239,240}$Pu, $^{90}$Sr, $^{137}$Cs and $^{99}$Tc are shown in Table 6.2.1, which includes the fractions of the $^{239,240}$Pu on suspended particles in the seawater.

*Table 6.2.1. Concentrations of radionuclides in seawater collected in the Thule area and at Illulissat.*

| Location (water depth) | Sampling depth (m) | $^{239,240}$Pu (mBq m$^{-3}$) | $^{239,240}$Pu (filter/total) | $^{90}$Sr (Bq m$^{-3}$) | $^{137}$Cs (Bq m$^{-3}$) | $^{99}$Tc (mBq m$^{-3}$) |
|---|---|---|---|---|---|---|
| C | 5 | 26±2 | 0.74±0.07 | | | |
| (120 m) | 85 | 14±1 | 0.28±0.04 | | | |
| | 105 | 14±1 | 0.41±0.05 | | | |
| H' | 5 | 9.3±0.8 | 0.08±0.02 | | | |
| (183 m) | 160 | 30±2 | 0.59±0.05 | | | |
| P | 5 | 9.5±0.8 | 0.05±0.01 | | | |
| (213 m) | 178 | 26±1 | 0.63±0.06 | | | |
| | 198 | 40±1 | 0.78±0.07 | | | |
| 20 | 5 | 9.0±0.8 | 0.05±0.02 | 0.81±0.08 | 2.3±0.3 | 18±4 |
| (234 m) | 199 | 34±2 | 0.68±0.06 | | | |
| | 219 | 17±1 | 0.46±0.05 | | | |
| Ny-4 | 5 | 9.2±0.9 | 0.06±0.02 | | | |
| (300 m) | 260 | 9.8±0.9 | 0.03±0.01 | | | |
| | 280 | 4.4±0.6 | 0.05±0.02 | | | |
| Dundas (30 m) | 5 | | | 0.91±0.09 | 2.2±0.2 | 20±4 |
| Illulissat | 5 | | | 0.77±0.08 | 2.0±0.2 | 53±13 |

The concentrations of $^{239,240}$Pu in seawater at the background station Ny-4 south of Wolstenholme Island are 10 mBq m$^{-3}$ and below and only a small fraction, 6% or less, is associated with suspended particles in the water. The concentrations in Bylot Sound at the locations H', 20 and P are significantly higher in near-bottom waters, up to 40 mBq m$^{-3}$ and a significant fraction of the activity, 46-78%, is associated with suspended particles. However, the samples of surface water in Bylot Sound show low concentrations corresponding to those at station Ny-4 and with a small fraction, 8% or lower, of the activity on suspended particles. The variability of the activity ratios of $^{238}$Pu to $^{239,240}$Pu on suspended particles collected on the filters is relatively small with an average value of 0.014±0.001 (1 SE, n=5), which is similar to that of accident plutonium. The variability of the isotopic ratios of $^{241}$Am to $^{239,240}$Pu in seawater is larger with an average value of 0.13±0.02 (1 SE, n=18). The samples from station C in Wolstenholme Fjord north of Saunders Island are influenced by accident plutonium from Bylot Sound throughout the water column with concentrations higher than at the background reference site Ny-4 and showing higher fractions, 28-74%, of plutonium on suspended particles.

The seawater data illustrate in a consistent manner that plutonium is remobilised from the seabed in Bylot Sound on suspended particles and transported to the north out of Wolstenholme Fjord into Baffin Bay.

The concentrations of $^{90}$Sr, $^{137}$Cs and $^{99}$Tc in seawater are low at Thule and Illulissat and compare well with levels in samples collected in 2004 along the west coast of Greenland. The radionuclides $^{90}$Sr and $^{137}$Cs are well mixed in North Atlantic seawater and show little variability in contrast to $^{99}$Tc, which shows a marked gradient due to the transport with sea currents from Europe.

## 6.3 Seaweed

Brown seaweed, *Fucus distichus*, was collected from the Thule area, 6 locations in Bylot Sound, 4 locations at Dundas, one on Wolstenholme Island and one at the Eider Islands, cf. Fig. 3.1. Sample sizes ranged from 1 to 4 kg fresh weight. The samples were analysed for gamma emitting radionuclides, $^{99}$Tc, plutonium and americium. The concentrations found for plutonium and americium are shown in Table 6.3.1. Analytical uncertainties are 10% for $^{239,240}$Pu, 20-25% for $^{238}$Pu and 10-15% for $^{241}$Am.

The seaweed concentrations of $^{239,240}$Pu at Narssarssuk in Bylot Sound (0.19-0.44 Bq kg$^{-1}$) are systematically higher than at locations outside Bylot Sound (0.10-0.16 Bq kg$^{-1}$). The plutonium levels in seaweed from Bylot Sound indicate that accident plutonium is remobilised from the contaminated sediments and transported to surface waters in the sound. This is supported by the fact that activity ratios of $^{238}$Pu to $^{239,240}$Pu in seaweed samples from Bylot Sound reflect accident plutonium of 0.01-0.02 while the activity ratios in samples from outside the sound of 0.02-0.03 are closer to the typical value of the activity ratio in global fallout of 0.04 (Perkins and Thomas, 1980).

The concentrations of $^{241}$Am in seaweed follow the same pattern as for $^{239,240}$Pu with higher concentrations at locations in Bylot Sound than outside the sound. The activity ratios of $^{241}$Am to $^{239,240}$Pu in seaweed show no significant variation by location and has a mean value of 0.17±0.01 (1 SE, n=12).

The concentrations of $^{99}$Tc and $^{137}$Cs in seaweed show no significant variation by location over the area. The mean concentration of $^{99}$Tc in seaweed is 5.0±0.1 Bq kg$^{-1}$ dw (1 SE, n=12) and the mean concentration of $^{137}$Cs is 0.39±0.02 Bq kg$^{-1}$ dw (1 SE, n=12).

*Table 6.3.1. Plutonium and americium in samples of seaweed (Fucus distichus)*

| Location | $^{239,240}$Pu (Bq kg$^{-1}$ dw) | $^{238}$Pu (Bq kg$^{-1}$ dw) | $^{241}$Am (Bq kg$^{-1}$ dw) |
|---|---|---|---|
| Bylot Sound SE, Narssarssuk 1 | 0.24 | 0.005 | 0.05 |
| Bylot Sound SE, Narssarssuk 2 | 0.18 | 0.003 | 0.03 |
| Bylot Sound SE, Narssarssuk 3 | 0.44 | 0.006 | 0.07 |
| Bylot Sound SE, Narssarssuk 4 | 0.24 | 0.005 | 0.05 |
| Bylot Sound SE, Narssarssuk 5 | 0.19 | 0.004 | 0.04 |
| Bylot Sound SE, Narssarssuk 6 | 0.19 | 0.006 | 0.03 |
| Dundas 1 | 0.15 | 0.004 | 0.02 |
| Dundas 2 | 0.13 | 0.004 | 0.01 |
| Dundas 3 | 0.10 | 0.002 | 0.02 |
| Dundas 4 | 0.11 | 0.003 | 0.01 |
| Wolstenholme Island | 0.12 | 0.003 | 0.02 |
| Eider Islands | 0.16 | 0.004 | 0.02 |

## 6.4 Benthic biota

Samples of benthic biota were collected with a Sigsbee trawl along four 500-m transects, three in Bylot Sound (T1, T2 and T3) within a few kilometres of the accident site and one north of Saunders Island (T4) about 17 km north-west of the accident site, cf. Fig 3.1. The numbering of the trawl hauls is from the east to the west with haul no. 4 north-east of Saunders Island.

The biota from each haul was sorted by species resulting in composite samples including from a single to over 300 individuals and sample amounts in the range 0.02-0.8 kg fw. Bivalves comprised the following species: *Macoma calcarea*, *Clinocardium ciliatum*, *Nuculana minuta*, *Astarte borealis*, *Serripes groenlandicus*, *Musculus niger*. Brittle stars were *Ophiura albida*, shrimp were *Crangonidae* and *Pandalidae*, and whelks were *Buccinum finmarchianum*.

Analyses were made of plutonium and americium in the biota samples, and the results are given in the Tables 6.4.1, 6.4.2 and 6.4.3 showing geometric mean concentrations for bivalves (4-5 species), whelks (1 species) and shrimp (2 species) for each trawl haul. The corresponding variation from minimum to maximum concentration across species is shown in Figs. 6.4.1, 6.4.2 and 6.4.3.

*Table 6.4.1. Geometric mean values of concentrations of $^{239,240}$Pu in benthic fauna (Bq kg$^{-1}$ fw).*

| | Bivalves | Whelks | Shrimp |
|---|---|---|---|
| Haul 1 | 0.18 | 0.086 | 0.009 |
| Haul 2 | 0.47 | 0.27 | 0.017 |
| Haul 3 | 0.76 | 0.28 | 0.044 |
| Haul 4 | 0.098 | 0.073 | 0.004 |

*Table 6.4.2. Geometric mean values of concentrations of $^{238}$Pu in benthic fauna (Bq kg$^{-1}$ fw).*

| | Bivalves | Whelks | Shrimp |
|---|---|---|---|
| Haul 1 | 0.0027 | 0.0007 | 0.0003 |
| Haul 2 | 0.0050 | 0.0046 | 0.0005 |
| Haul 3 | 0.010 | 0.0034 | 0.0027 |
| Haul 4 | 0.0015 | 0.0009 | 0.0011 |

*Table 6.4.3. Geometric mean values of concentrations of $^{241}$Am in benthic fauna (Bq kg$^{-1}$ fw).*

| | Bivalves | Whelks | Shrimp |
|---|---|---|---|
| Haul 1 | 0.069 | 0.029 | 0.003 |
| Haul 2 | 0.15 | 0.082 | 0.003 |
| Haul 3 | 0.20 | 0.15 | 0.008 |
| Haul 4 | 0.045 | 0.021 | 0.002 |

The radionuclide concentrations are generally higher in bivalves than in whelks and shrimp and highest in samples from trawl haul no. 3 and lowest in samples from haul no. 4. The highest concentration of $^{239,240}$Pu (4 Bq kg$^{-1}$ fw) was found in a bivalve sample from haul no. 3. The sample was composed of 24 individuals of *Clinocardium ciliatum* and had a total weight of 0.09 kg fw. A sample of brittle star *Ophiura albida* was collected from haul 1 (50 individuals, 0.3 kg fw) and showed concentrations above the geometric mean values of the bivalves, but within the range covered by the bivalves.

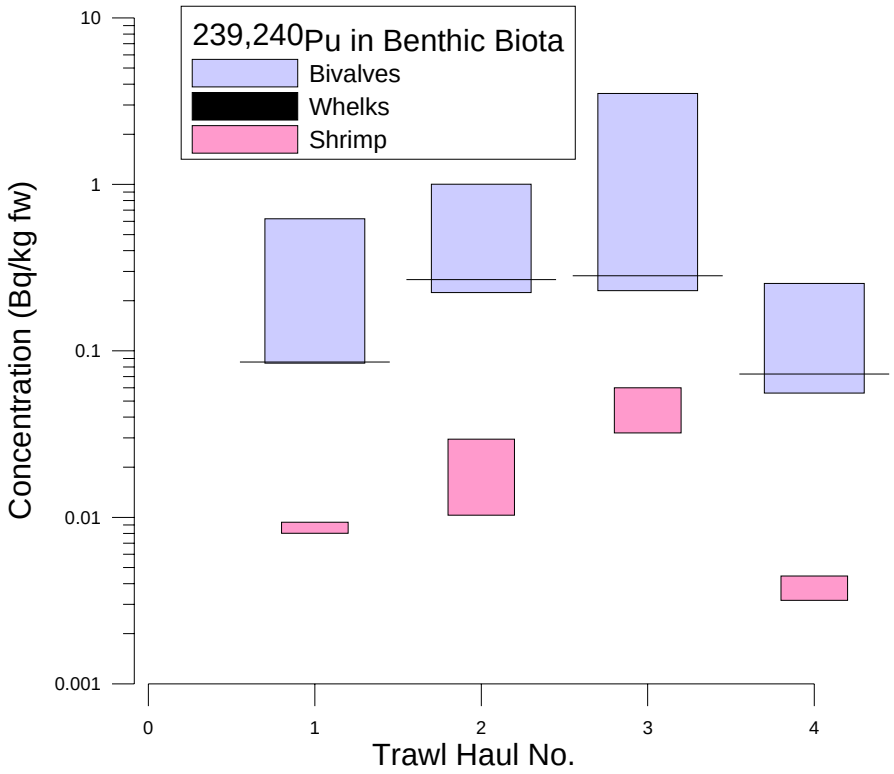


*Fig. 6.4.1. Ranges of concentrations of $^{239,240}$Pu in bivalves, whelks and shrimp.*

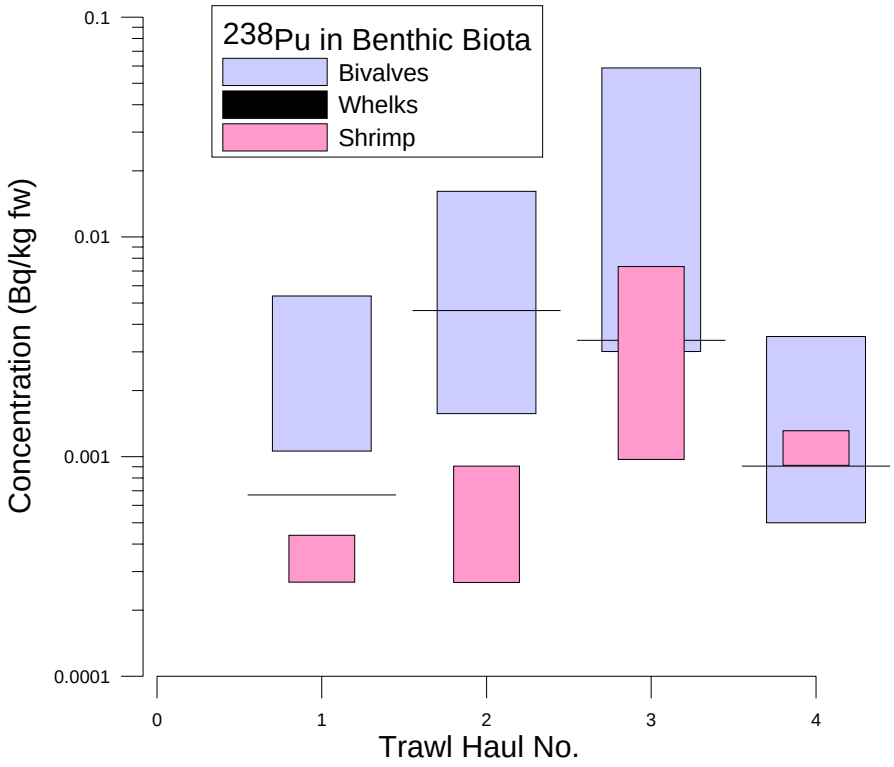


*Fig. 6.4.2. Ranges of concentrations of $^{238}$Pu in bivalves, whelks and shrimp.*

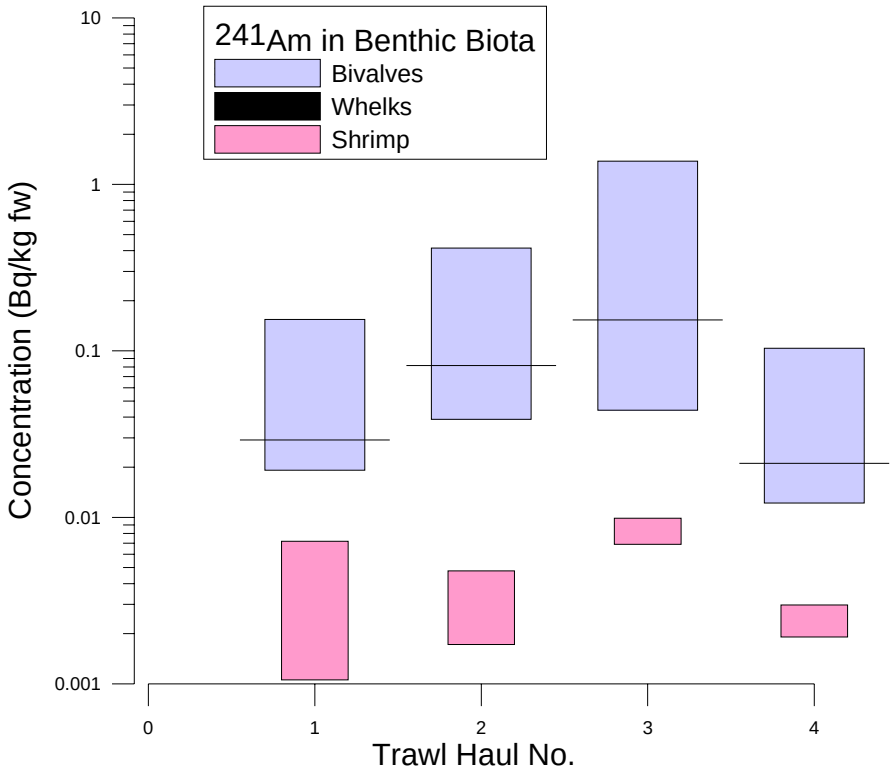


*Fig. 6.4.3. Ranges of concentrations of $^{241}$Am in bivalves, whelks and shrimp.*

The isotopic ratios of concentrations of $^{238}$Pu to $^{239,240}$Pu in bivalves do not vary across the hauls and show a mean value of 0.016±0.003 (1 SE, n=18). The corresponding ratios for shrimp and whelks are 0.034±0.005 (1 SE, n=5) and 0.012±0.002 (1 SE, n=4), respectively. The lower ratios of 0.01-0.02 are typical for the accident plutonium while the higher ratio is typical for global fallout. This indicates that shrimp feed is not of local origin in contrast to feed for bivalves and whelks.

The variation of the isotopic ratios of concentrations of $^{241}$Am to $^{239,240}$Pu in biota shows no distinct pattern across species or locations. The ratios vary from 0.13 to 0.79 with a mean value of 0.40±0.04 (1 SE, n=31).

## 6.5 Seal

A harp seal (*Phoca groenlandica*) was shot in Bylot Sound on 25 August 2003, cf. Fig. 3.1. Samples were collected of flesh (3.3 kg fw) and liver (1.9 kg fw). Analyses were carried out of plutonium, americium, $^{137}$Cs and the naturally occurring radionuclides $^{210}$Po and $^{210}$Pb. The results are shown in Table 6.5.1.

The concentrations of plutonium and americium are very low and detectable only for $^{239,240}$Pu in liver. Concentrations of $^{238}$Pu were below the limit of detection of 0.0001 Bq kg$^{-1}$ fw. The concentrations of $^{137}$Cs in seal of 0.2-0.3 Bq kg$^{-1}$ fw correspond to concentrations in fish, which have been found at 0.1-0.2 Bq kg$^{-1}$ fw in shorthorn sculpin collected in the area. The concentrations of $^{210}$Po and $^{210}$Pb are typical for seals and reflect the fact that harp seals feed primarily on fish and crustaceans.

*Table 6.5.1. Concentrations of radionuclides in flesh and liver from a seal caught in Bylot Sound (Bq kg$^{-1}$ fw). Uncertainties represent one standard deviation.*

| Radionuclide | Flesh (Bq kg$^{-1}$ fw) | Liver (Bq kg$^{-1}$ fw) |
|---|---|---|
| $^{137}$Cs | 0.20 ± 0.02 | 0.26 ± 0.03 |
| $^{239,240}$Pu | < 0.0002 | 0.0020 ± 0.0003 |
| $^{241}$Am | < 0.0001 | < 0.0001 |
| $^{210}$Po | 8.1 ± 0.8 | 43 ± 4 |
| $^{210}$Pb | 0.31 ± 0.03 | 0.25 ± 0.03 |

## 6.6 Soil

Soil samples from the Narssarssuk coastal area covered the 8 locations shown in Fig. 3.2. At each location, 3 sub-site soil cores (A, B and C) were collected down to depths of 20-30 cm within an area of one square metre. The cores were generally separated into the depth sections: 0-3 cm, 3-6 cm, 6-10 cm, 10-15 cm, 15-20 cm, 20-25 cm and 25-30 cm. The resulting 145 soil samples ranged in size from a few tens of grams to about a kilogram of dry weight sieved material. Samples of smaller size were from the peaty surface soil layers while the heavier samples were from deeper layers. The samples were analysed by gamma spectrometry for gamma emitting radionuclides and by radiochemical procedures followed by alpha spectrometry for plutonium and americium.

The radiochemical procedures and alpha-spectrometry measurements were carried out on 1-g aliquots of the samples permitting identification of $^{238}$Pu, $^{239,240}$Pu and $^{241}$Am. The resulting profiles of $^{239,240}$Pu concentration (Bq kg$^{-1}$ dw) with depth are given in Figs. 6.6.1 and 6.6.2. The concentrations vary considerably across the locations and sub sites. The maximum concentration at each location ranges from 2 Bq kg$^{-1}$ dw at location no. 7 to 80 Bq kg$^{-1}$ dw at location no. 5. In all cases the maximum concentrations occur in the surface layers of 0-3 cm or 3-6 cm. A hot particle containing 23 Bq of $^{239,240}$Pu was identified in the sample from location no. 8A at depth 3-6 cm. Fig. 6.6.2 does not include the activity of this particle, which was isolated and used for leaching experiments.

The concentrations of $^{238}$Pu in the soil samples relative to $^{239,240}$Pu have a mean value of 0.018±0.001 (1 SE, n=37) and show no significant variation between locations. The $^{238}$Pu/$^{239,240}$Pu activity ratios are all lower than the value of 0.04 typical for global fallout (Perkins and Thomas, 1980) indicating that the soil is contaminated with accident plutonium at all locations. The concentrations of $^{241}$Am in the soil relative to $^{239,240}$Pu have a mean value of 0.27±0.02 (1 SE, n=27) and show no significant variation between locations. The $^{241}$Am/$^{239,240}$Pu activity ratios are intermediate between values of 0.1-0.2 characterising accident plutonium and 0.4 characterising plutonium from global fallout (Michel et al., 2002). The $^{241}$Am/$^{239,240}$Pu activity ratios thus also indicate that the soil sampled at Narssarssuk is contaminated with accident plutonium at all locations.

Gamma emitting radionuclides in the soil were determined by standard gamma spectrometry. The analyses were carried out on sample sizes up to a few hundred grams giving information on the man-made radionuclides $^{137}$Cs and $^{241}$Am and naturally occurring radionuclides ($^{40}$K and daughters of $^{238}$U and $^{232}$Th). However, the $^{241}$Am results from these analyses may not be representative for several reasons: 1) in case of the presence of one or more hot particles in the aliquot analysed and 2) if hot particles in the soil sample were not included in the aliquot analysed. Screening analyses were made to investigate this further by sub-dividing samples down to 5-15 g size and performing short-term measurements of $^{241}$Am in these

small aliquots. Furthermore, screening analyses were made on the entire amount of soil from the top 0-10 cm layers to ensure that no hot particles had escaped detection from the standard gamma analyses.

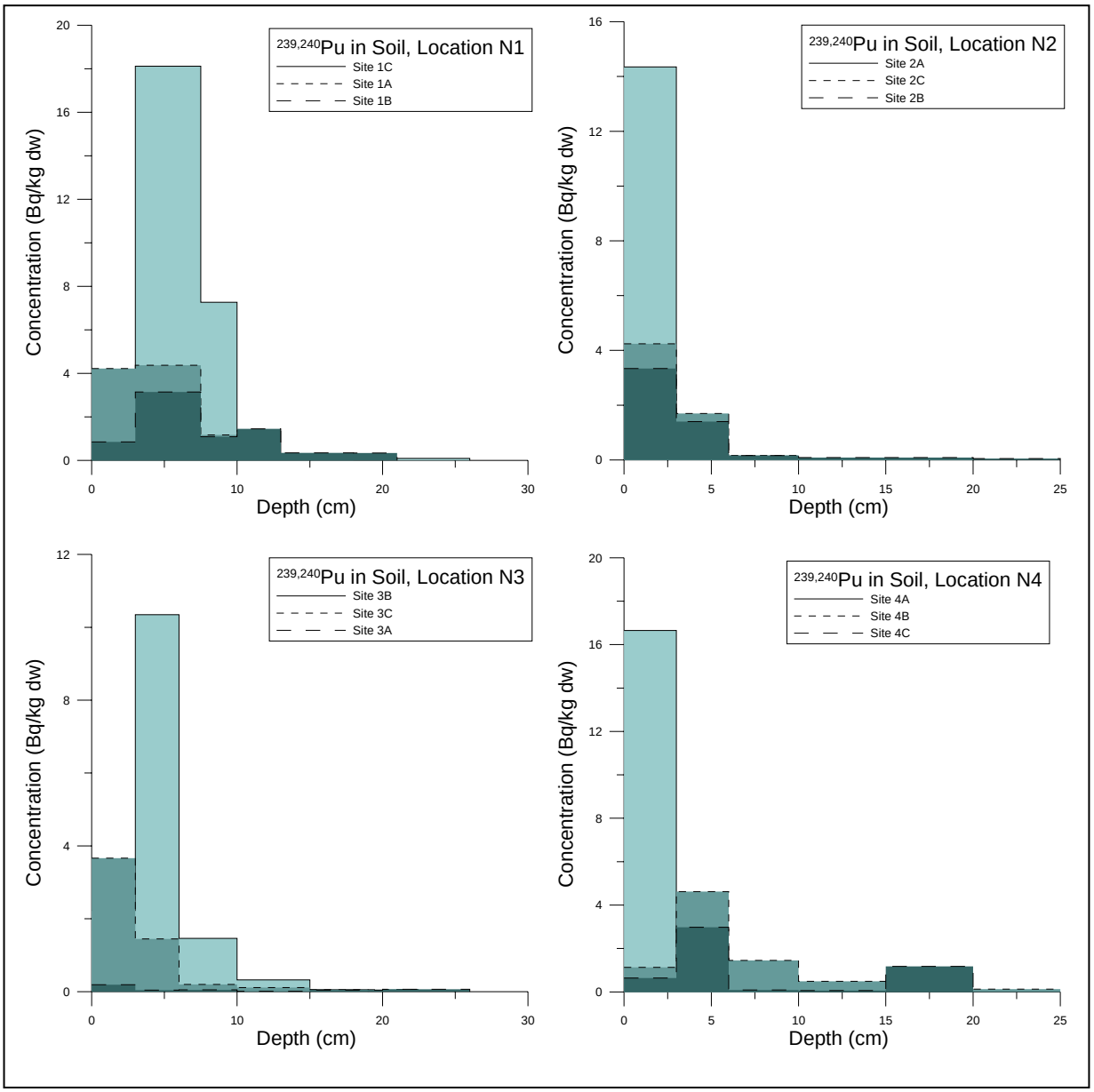


*Fig. 6.6.1. Soil concentrations of $^{239,240}Pu$ (Bq $kg^{-1}$ dw) by depth at locations no. 1, 2, 3 and 4.*

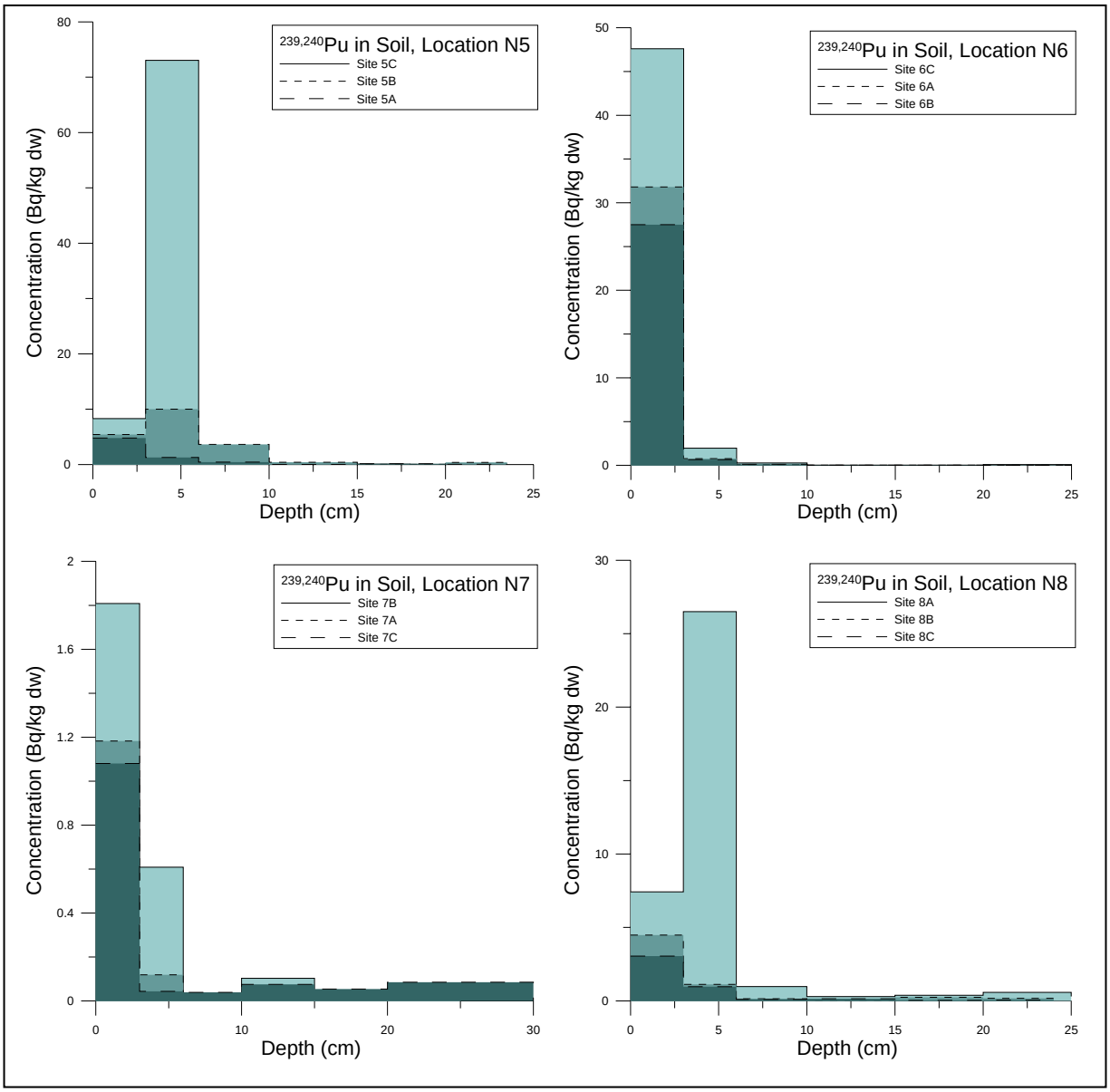


*Fig. 6.6.2. Soil concentrations of $^{239,240}$Pu (Bq kg$^{-1}$ dw) by depth at locations no. 5, 6, 7 and 8.*

A comparison between the results from the standard gamma analyses and the screening analyses is shown in Fig. 6.6.3, which shows ratios of total $^{241}$Am activities (Bq m$^{-2}$) in the soil cores determined by gamma screening relative to those determined by standard gamma spectrometry. It is noted that there are no values above 2 meaning that no very hot particles are missing from the standard analyses. Ratios of about 1.6 are found at three locations, which mean that the standard gamma analysis underestimates the $^{241}$Am activity by some 40% for these soil cores. Low ratios may be explained by the less sensitive screening analyses compared to the standard analyses. The screening analyses thus confirm that the $^{241}$Am activities

in the soil cores obtained from the standard gamma analyses are correct within a factor of 2.

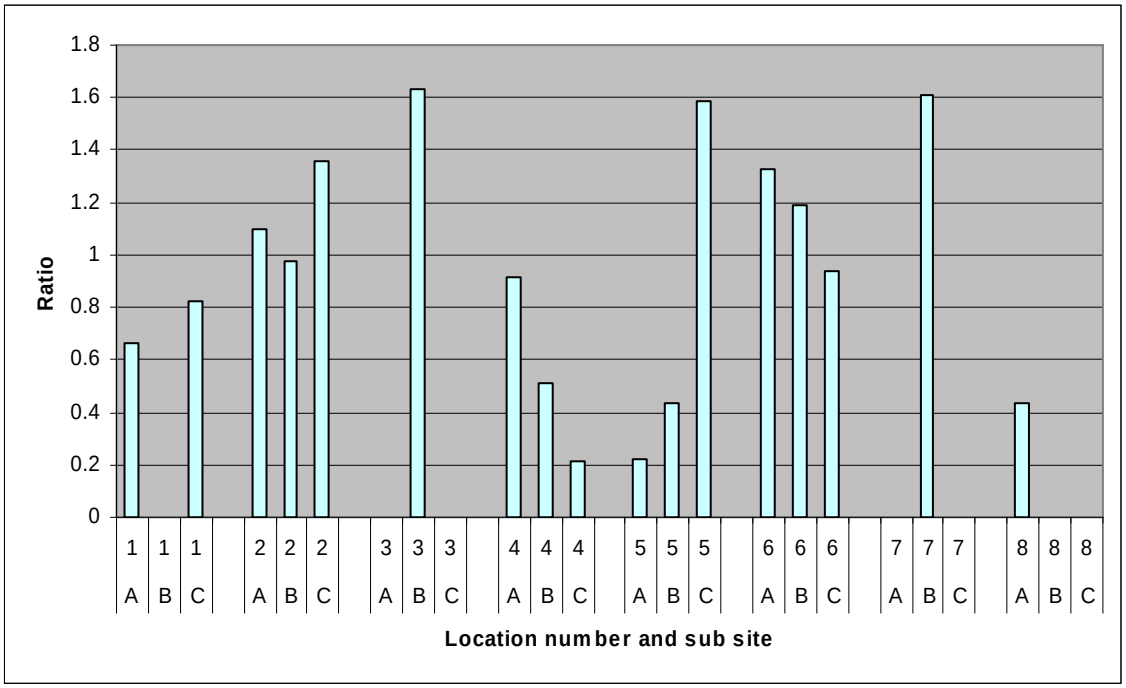


*Fig 6.6.3. Ratios of activities of $^{241}$Am in the soil cores determined by gamma screening relative to those determined by standard gamma spectrometry*

A summary of $^{239,240}$Pu in soil by area at the 8 locations estimated by two methods is shown in Fig. 6.6.4. One method is the direct determination of plutonium by radiochemical separation and alpha spectrometry of small 1-g aliquots. The other is based on standard gamma spectrometric measurements of $^{241}$Am in the samples and an assumption of an activity ratio between $^{241}$Am and $^{239,240}$Pu of 0.27 as found from the alpha spectrometric analyses. The comparison shows considerable variation between the plutonium concentrations determined by the two methods across the locations and sub sites. The ratios of the plutonium activities based on alpha analyses to those based on gamma analyses range from 0.1 to 2.8 with a mean value of 0.80±0.13 (1 SE, n=24). The variability illustrates the inhomogeneous distribution of plutonium in the soil and demonstrates that the concentrations of plutonium in the 1-g aliquots in many cases are not representative for the concentrations of plutonium in the samples from which they were taken. The plutonium results based on the gamma analyses are considered to be more representative than those based on the alpha analyses. The global fallout of $^{239,240}$Pu at Thule in 1974 was estimated at 13 Bq m$^{-2}$ by Hanson (1980). By comparison with global fallout, the 8 locations seem all affected by accident plutonium by varying degree.

The soil profiles of $^{137}$Cs obtained from the gamma analyses are shown in Figs. 6.6.5 and 6.6.6 for the 8 locations. As for plutonium, the concentrations of $^{137}$Cs vary considerably across locations and sub sites. The maximum concentration occurs generally in the layers 0-3 cm or 3-6 cm and ranges from 27 Bq kg$^{-1}$ at location no. 7 to 200 Bq kg$^{-1}$ at location no. 6.

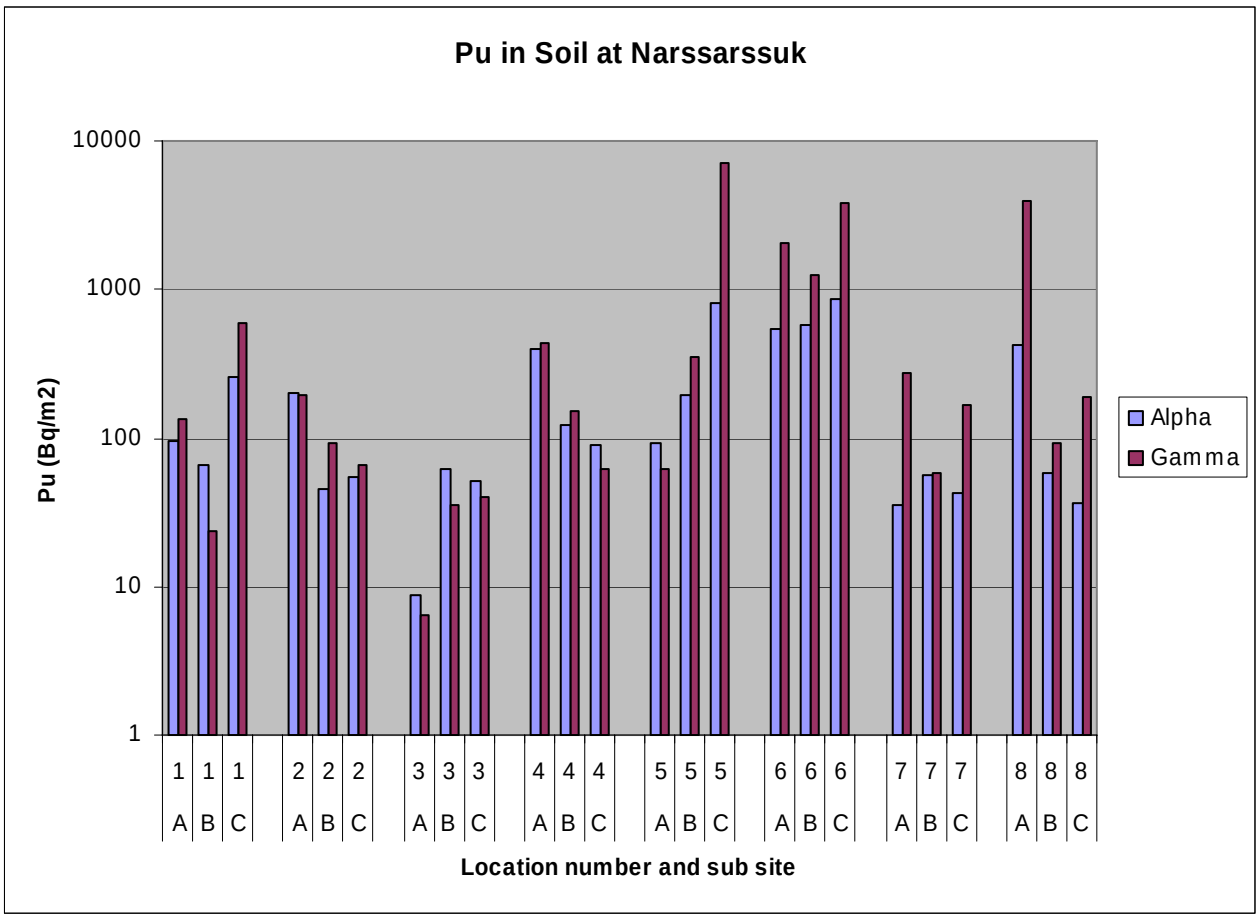


*Fig. 6.6.4. Plutonium-239,240 in soil cores (Bq $m^{-2}$) estimated from radiochemical analyses and alpha measurements of 1-g aliquots and gamma spectrometric measurements on larger samples.*

The concentrations of $^{137}Cs$ in soil by area are shown for all locations and sub sites in Fig. 6.6.7. The variability of the concentrations across the locations and sub sites is less pronounced than that of the plutonium concentrations. The $^{137}Cs$ concentrations range from a low value of 14 Bq $m^{-2}$ at site 3A to a high value of 6400 Bq $m^{-2}$ at site 6A with an average value of 1040±280 Bq $m^{-2}$(1 SE, n=24). The source of the $^{137}Cs$ in the environment at Thule is atmospheric fallout from nuclear weapons testing, which was estimated by Hanson (1980) at a value of 777 Bq $m^{-2}$ in 1974. Radioactive decay reduces this concentration to 400 Bq $m^{-2}$ in 2003, which compares quite well with the concentrations found at most of the sites. The low value at location no. 3 and the high values at the locations no. 5 and 6 may be explained by special site conditions (e.g. runoff and accumulation) that remove or enhance matter deposited on soil from the atmosphere.

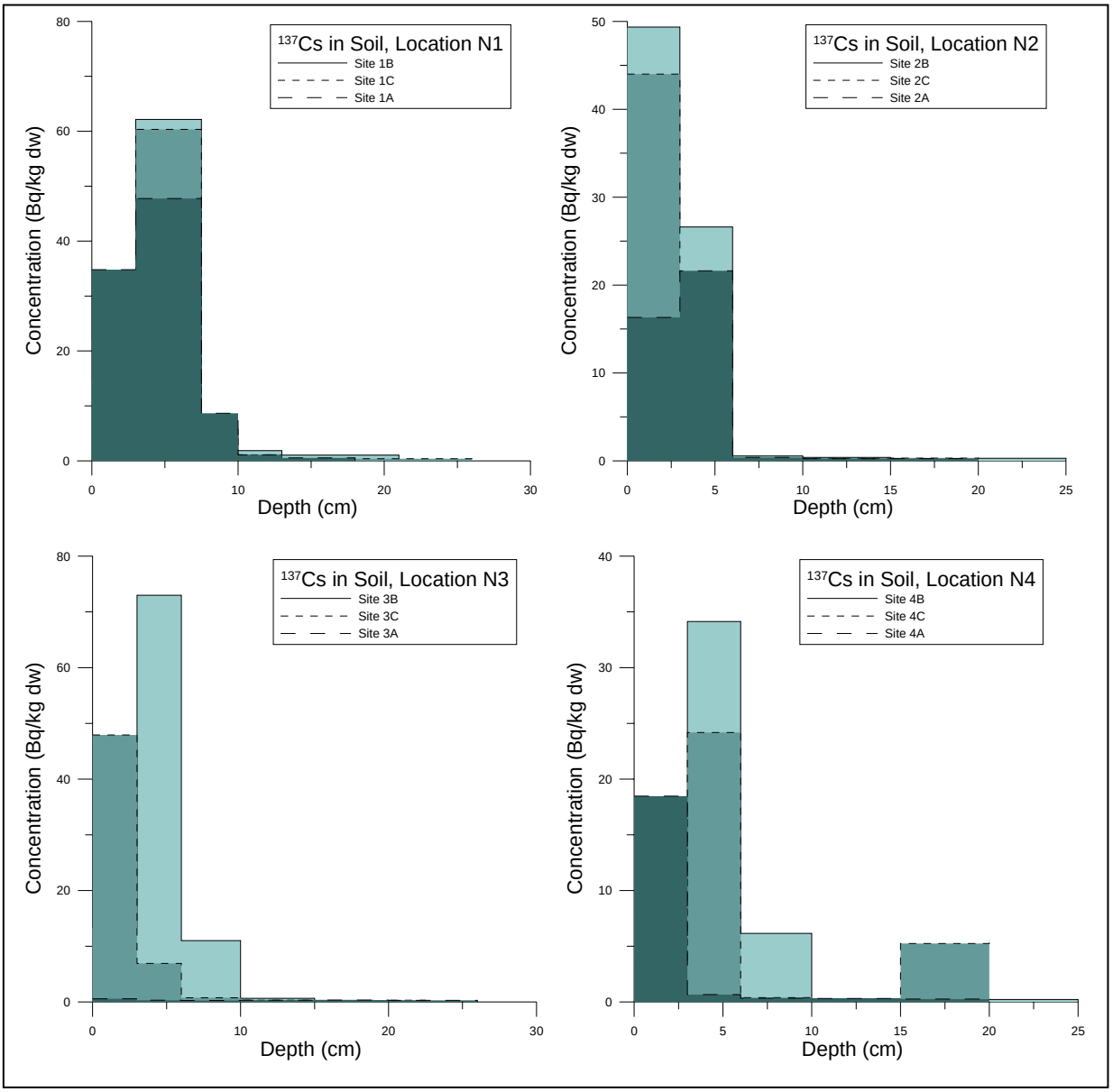


*Fig. 6.6.5. Soil concentrations of $^{137}Cs$ (Bq $kg^{-1}$ dw) by depth at locations no. 1, 2, 3 and 4.*

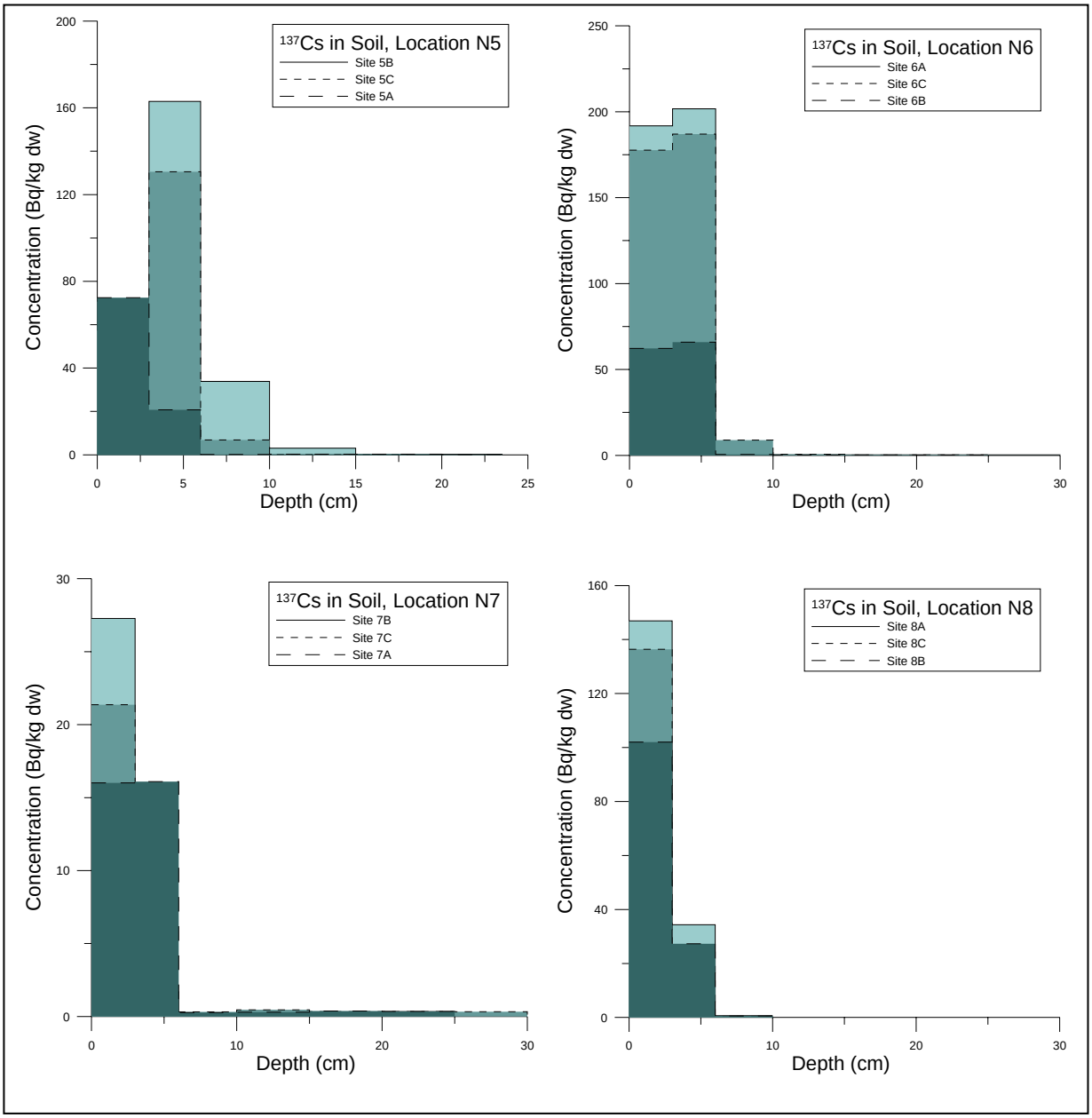


*Fig. 6.6.6. Soil concentrations of $^{137}Cs$ (Bq $kg^{-1}$ dw) by depth at locations no. 5, 6, 7 and 8.*

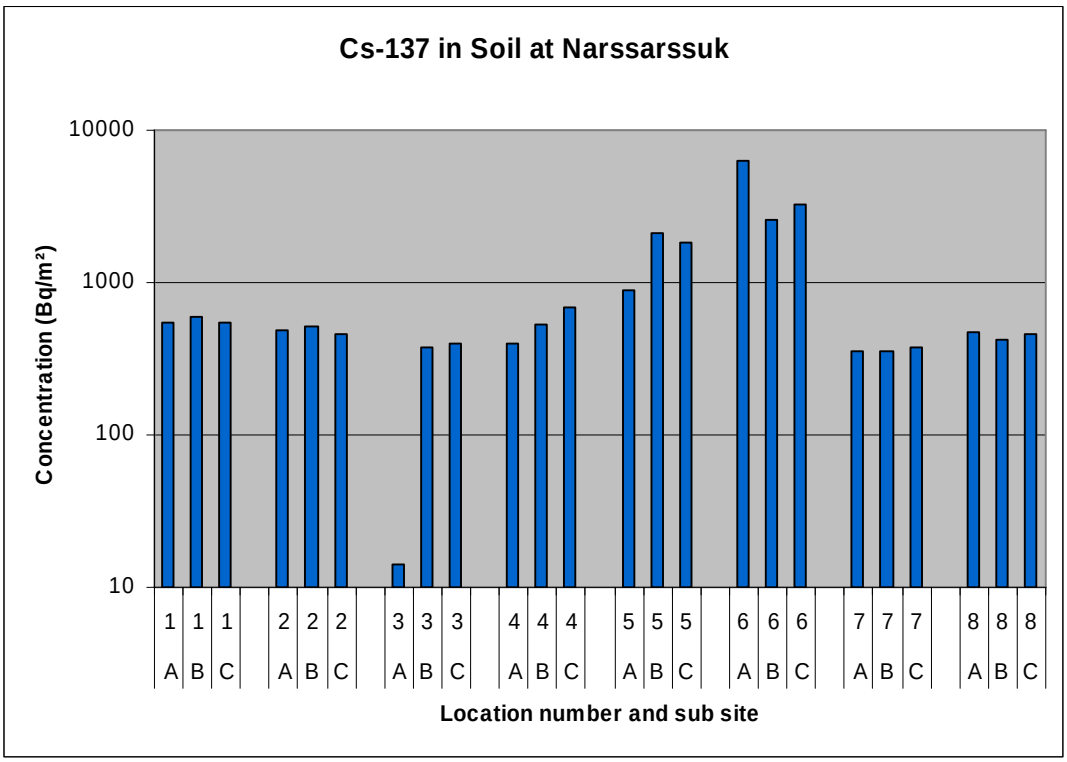


*Fig. 6.6.7. Concentrations of $^{137}Cs$ (Bq $m^{-2}$) in soil cores collected at Narssarssuk.*

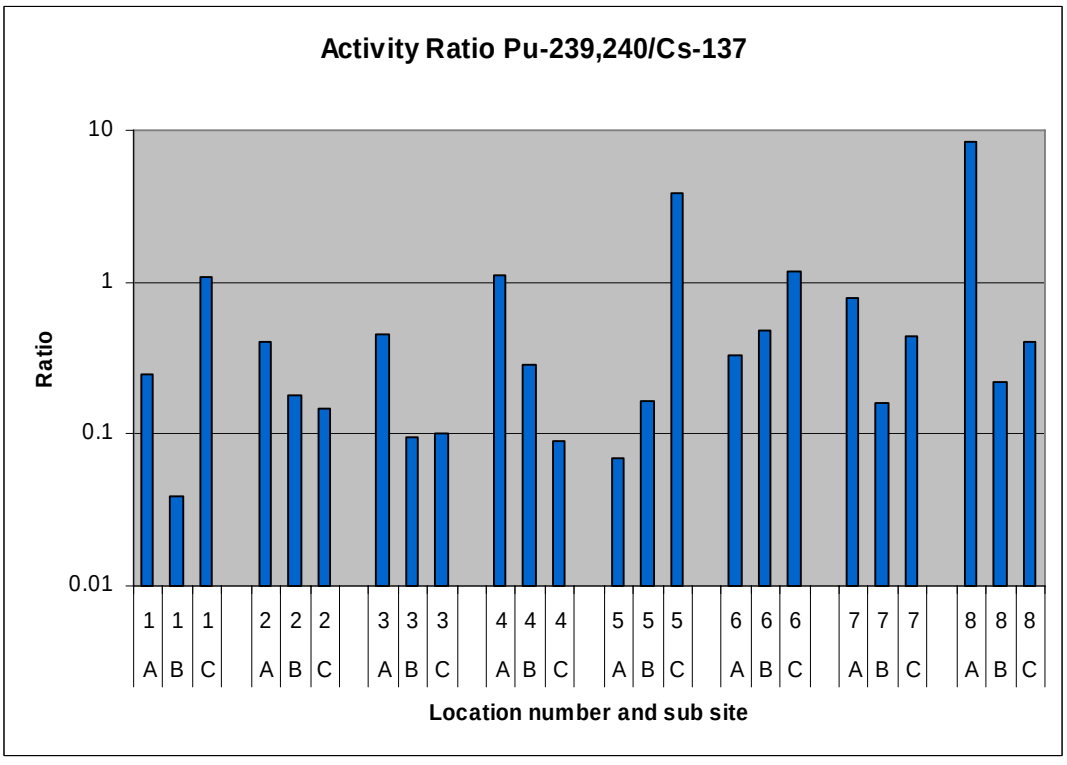


*Fig. 6.6.8. Ratios of concentrations in soil of $^{239,240}Pu$ to those of $^{137}Cs$ (Bq $m^{-2}$ per Bq $m^{-2}$) across locations and sub sites at Narssarssuk.*

The conditions at the individual sites may be taken into consideration by examining the ratio of the deposition of $^{239,241}Pu$ to that of $^{137}Cs$. These ratios are presented graphically in Fig. 6.6.8 and show considerable variability across sites from a low

value of 0.04 at site 1B to a high value of 8 at site 8A with a mean value of 0.9±0.4 (1 SE, n=24). The ratios are all higher than 0.033, which may be derived from Hanson's data of global fallout at Thule (0.033 = 13 Bq $m^{-2}$ $^{239,240}$Pu / 400 Bq $m^{-2}$ $^{137}$Cs) and illustrate that the accident plutonium is very inhomogeneously distributed across the Narssarssuk area with significantly different concentrations at the scale of kilometres (differences between locations) as well as at the scale of a metre (differences between sub sites).

## 6.7 Hot particles

The analyses of the sediment samples have shown that plutonium is very inhomogeneously distributed in the sediments due to the presence of hot particles. The systematic gamma screening of $^{241}$Am in 5-15 g aliquots of the sediment samples have indicated a geometric mean $^{239,240}$Pu activity of a few Bq with an approximate 95% confidence interval of from 0.1 to 50 Bq. The maximum $^{239,240}$Pu activity determined in a hot particle in the sediments is 1500 Bq, cf. Section 6.1.

A number of hot particles were identified in the soil samples. One of these, the sample from site 8A at depth 3-6 cm, had a mass of 54 g dw for which the analysis yielded an estimated concentration of 100 Bq $kg^{-1}$ of $^{241}$Am corresponding to about 5 Bq in the sample. Further investigations showed, however, that the activity was due mainly to a single hot particle, which was isolated by successive subdivisions of the sample material combined with gamma measurements. This particle was subject to leaching experiments to identify chemical procedures that would completely dissolve the plutonium and americium in the particle and to determine the isotopic ratios. The particle was analysed by chemical separation and alpha spectrometry and found to contain 3.6 Bq $^{241}$Am and 23 Bq $^{239,240}$Pu with the activity ratios of 0.015 for $^{238}$Pu/$^{239,240}$Pu and 0.158 for $^{241}$Am/$^{239,240}$Pu. The latter value is in good agreement with activity ratios of $^{241}$Am/$^{239,240}$Pu from five hot particles from Thule sediment samples showing a mean value of 0.169±0.002 (1 SE, n=5). These hot particles were from sediment samples collected from earlier expeditions and had $^{239,240}$Pu activities ranging from 6 to 84 Bq (Eriksson, 2002).

*Table 6.7.1. Hot particles found in soil samples collected at Narssarssuk showing sampling locations and estimated activities of $^{239,240}$Pu and $^{241}$Am.*

| Location, sub site, depth | $^{239,240}$Pu (Bq) | $^{241}$Am (Bq) | Uncertainty (%) |
|---|---|---|---|
| 5C, 3-6 cm | 150 | 25 | 10 |
| 6B, 0-3 cm | 3 | 0.5 | 20 |
| 6B, 0-3 cm | 3 | 0.5 | 20 |
| 6C, 0-3 cm | 3 | 0.5 | 20 |
| 6C, 0-3 cm | 3 | 0.5 | 20 |
| 6C, 0-3 cm | 6 | 1 | 20 |
| 8A, 3-6 cm | 3 | 0.5 | 20 |
| 8A, 3-6 cm | 23 | 3.6 | 10 |

Eight hot particles were found in the soil samples as listed in Table 6.7.1. The $^{239,240}$Pu activity in the particles are calculated from the $^{241}$Am activities given in round numbers estimated from gamma analyses except for the particle mentioned above for which the $^{239,240}$Pu activity of 23 Bq was determined by alpha spectrometry. The maximum $^{239,240}$Pu activity determined in a hot particle in the soil is 150 Bq.

# 7 Conclusions

Analyses of marine and terrestrial samples collected in August 2003 from Bylot Sound show that plutonium from nuclear weapons in the American B52 plane, which in January 1968 crashed on the sea ice and caught fire, persists in the environment at Thule.

Successful sampling of marine sediments was accomplished by selecting sampling locations based on information obtained from acoustic mapping of the seabed. The highest concentrations of plutonium are found in the marine sediments under the location where the plane crashed. The distribution of plutonium in the marine sediment is very inhomogeneous and associated with hot particles with activities found up to 1500 Bq $^{239,240}$Pu. Sediment samples collected in Wolstenholme Fjord north of the accident site show plutonium concentrations more than an order of magnitude above background levels, which illustrates the redistribution of plutonium after the accident.

The total plutonium inventory in the sediments has been assessed based on systematic analyses considering hot particles. The inventory of $^{239,240}$Pu in the sediments within a distance of 17 km from the point of impact of the B52 plane is estimated at 2.9 TBq (1 kg) with an approximate 95% confidence interval from 1.4 TBq to 6 TBq. This supports findings from the previous sampling in 1997 which found that earlier investigations might have underestimated the total inventory by not systematically considering the contribution from hot particles. Earlier estimates of the inventory were approximately 1.4 TBq $^{239,240}$Pu.

Seawater samples show increased concentrations of particle-associated plutonium in near-bottom water in Bylot Sound and seaweed samples show increased concentrations in Bylot Sound compared to locations outside Bylot Sound. These increased concentrations are due to resuspension of plutonium-containing particles from the seabed and transport further away from the area. The continuous mixing of the sediments by the benthic fauna has the effect that plutonium concentrations in the surface sediment layers are high in general and not buried under uncontaminated sediment.

Plutonium concentrations in seawater, seaweed and benthic animals in Bylot Sound are low but clearly above background levels.

All soil samples collected from Narssarssuk show accident plutonium with levels above background. Plutonium is very inhomogeneously distributed and associated with particles in the surface layers. Hot particles were found in soil with activities up to 150 Bq $^{239,240}$Pu.

Plutonium in the marine environment at Thule presents an insignificant risk to man. Most plutonium remains in the seabed under Bylot Sound far from man under relatively stable conditions. Concentrations of plutonium in seawater and animals are low and can not present any risks to man even by consumption of shellfish at the highest concentrations. However, the plutonium contamination of surface soil at Narssarssuk could constitute a small risk to humans visiting the location if radioactive particles are resuspended in the air so that they might be inhaled.

# 8 Recommendations

- A systematic mapping of accident plutonium on land at Narssarssuk should be carried out in order to clarify the extent of the terrestrial contamination
- The possible presence of radioactive particles in the air at Narssarssuk should be investigated by collecting and analysing aerosols in order to evaluate the possible risk to man from inhalation of resuspended radioactive particles
- Investigations of plutonium in the marine environment at Thule should be continued for scientific reasons and in order to document the radioactive pollution of the area.

# 9 Acknowledgements

The authors wish to thank the staff of the Radioecology Programme in the Radiation Research Department for their conscientious work on samples and analyses. The investigation vessel Adolf Jensen belonging to Pinngortitaleriffik (Grønlands Naturinstitut) in Nuuk, Greenland, collected the marine samples at Thule, and GEUS (Geological Survey of Denmark and Greenland) carried out the acoustic mapping of sediments in Bylot Sound.

The authors gratefully acknowledge the financial support from the Ministry of Environment to this investigation. The conclusions in this report do not necessarily reflect the views of the Ministry.

# 10 References

Aarkrog, A. (1971). Radioecological investigation of plutonium in an arctic marine environment. Health Phys., 20, 31-47.

Aarkrog, A. (1977). Environmental behaviour of plutonium accidentally released at Thule, Greenland. Health Phys., 32, 271-284.

Aarkrog, A., Dahlgaard, H., Holm, E., Hansen, H., Lippert, J. & Nilsson, K. (1981). Environmental Radioactivity in Greenland in 1980. Risø-R-449. Risø National Laboratory, Roskilde, Denmark.

Aarkrog, A., Dahlgaard, H. & Nilsson, K. (1984). Further studies of plutonium and americium at Thule, Greenland. Health Phys., 46, 29-44.

Aarkrog, A., Boelskifte, S., Dahlgaard, H., Duniec, S., Holm, E. & Smith, J. N. (1987). Studies of transuranics in an Arctic marine environment. J. Radioanal. Nucl. Chem. Articles, 115, 39-50.

Aarkrog, A., Buch, E., Chen, Q. J., Christensen, G. C., Dahlgaard, H., Hansen, H., Holm, E. & Nielsen, S. P. (1988). Environmental Radioactivity in the North Atlantic Region Including the Faroe Islands and Greenland. 1986. Risø-R-550. Risø National Laboratory, Roskilde, Denmark:.

Aarkrog, A., Chen, Q. J., Clausen, J., Christensen, G. C., Dahlgaard, H., Ellis, K., Hansen, H., Holm, E., Joensen, H. P,, Nielsen, S. P. & Strandberg, M. (1997). Environmental Radioactivity in the North Atlantic Region Including the Faroe Islands and Greenland. 1992 and 1993. Risø-R-757. Risø National Laboratory, Roskilde, Denmark

Chen, Q.J.; Aarkrog, A.; Nielsen, S.P.; Dahlgaard, H.; Lind, B.; Kolstad, A.K.; Yu, Yixuan, 2001. Procedures for determination of $^{239,240}$Pu, $^{241}$Am, $^{237}$Np, $^{234,238}$U, $^{228,230,232}$Th, $^{99}$Tc and $^{210}$Pb-$^{210}$Po in environmental materials. Risø-R-1263(EN), Risø National Laboratory, Roskilde, Denmark

Chen, Q.J.; Hou, X.L.; Yu, Y.X.; Dahlgaard, H.; Nielsen, S.P., 2002. Separation of Sr from Ca, Ba and Ra by means of $Ca(OH)_2$ and $Ba(Ra)Cl_2$ or $Ba(Ra)SO_4$ for the determination of radiostrontium. Anal. Chim. Acta 466 , 109-116

Dahlgaard, H., Chen, Q. J., Stiirup, S., Eriksson, M., Nielsen, S. P. & Aarkrog, A., 1999a. Plutonium Isotope Ratios in Environmental Samples from Thule (Greenland) and the Techa River (Russia) measured by ICPMS and a-Spectrometry. *International Symposium on Marine Pollution, Monaco,* 5-9 October 1998, IAEA-SM-3541109 (pp. 254-259). Vienna, Austria: IAEA.

Dahlgaard, H., Eriksson, M,, Ilus, E., Ryan, T., McMahon, C. A. & Nielsen, S. P., 1999b. Plutonium in an Arctic marine environment 29 years after the Thule accident. *Nordic Society for Radiation Protection, 12th ordinary meeting,* August 23-27, 1999, Skagen, Denmark.

Eriksson, M., 2002. On Weapons Plutonium in the Arctic Environment (Thule, Greenland). Risø-R-1321(EN) , Risø National Laboratory, Roskilde, Denmark

Fogh, C.L., 2000. NKS 1999 intercomparison of measurements of radioactivity. NKS-19. Nordic nuclear safety research, Roskilde, Denmark

Fogh, C.L., Nielsen, S.P. and Keith-Roach, M., 2002. Intercomparison of Radionuclides in Environmental Samples 2000-2001. Nordic nuclear safety research, Roskilde, Denmark

Hanson, W.C. (1980). Transuranic elements in Arctic Tundra Ecosystems. In W.C. Hanson (Ed.), Transuranic Elements in the Environment, DOE/TIC-22800. Springfield, VA: National Technical Information Service.

IAEA, 1999. Report on the intercomparison run and certified reference material IAEA-381, Radionuclides in Irish Sea Water. IAEA/AL/118; IAEA/MEL/66. International Atomic Energy Agency, Vienna, Austria

IAEA, 2000a. Summary report of the Proficiency Test for $^{239}$Pu, $^{241}$Pu and $^{241}$Am Measurement in a Mineral Matrix (PT002). International Atomic Energy Agency AQCS, Seibersdorf, Austria

IAEA, 2000b. IAEA-384, Radionuclides in Fangataufa lagoon sediment. IAEA/AL/126; IAEA/MEL/68. International Atomic Energy Agency, Vienna, Austria

IAEA, 2001. Report on the intercomparison run for the determination of radionuclides in soils, IAEA-326 and IAEA-327. IAEA/AL/100. International Atomic Energy Agency, Vienna, Austria

IAEA, 2004. Report on the worldwide intercomparison IAEA-414, Radionuclides in Mixed Fish from Irish Sea and the North Sea. IAEA/AL/145; IAEA/MEL/73. International Atomic Energy Agency, Vienna, Austria

IAEA, 2005. Report on the Worldwide Intercomparison Exercise IAEA-385, radionuclides in Irish Sea Sediment. IAEA/AL/151; IAEA/MEL/76. International Atomic Energy Agency, Vienna, Austria

Jensen, J.B., 2003. Akustisk kortlægning af havbundssedimenterne i Bylot sund, Thule. GEUS Rapport 2003/103. Danmarks og Grønlands Geologiske Undersøgelse, Miljøministeriet.

McMahon, C. A., Lecin Vintro, L., Mitchell, P. I. & Dahlgaard, H. (2000). Oxidation-state distribution of plutonium in surface and subsurface waters at Thule, northwest Greenland. *Appl. Radiat. Isot.*, 52, 697-703.

Michel, H., Barci-Funel, G., Dalmasso, J., Ardisson, G., Appelby, P.G., Haworth, E. and El-Daoushy, F., 2002. Plutonium and americium inventories in atmospheric fallout and sediment cores from Blelham tarn, Cumbria (UK). J. Environ. Radioact. 59(2):127-137.

Nielsen, S.P., 1996. An Intercomparison Exercise on radionuclides in Sediment Samples. Risø-R-914(EN). Risø National Laboratory, Roskilde, Denmark

Nielsen, S.P. (2004) An Intercomparison on Radionuclides in Environmental Samples. Baltic-Danish Co-operation Project on radiation Protection 2001-2003. Risø-R-1467(EN). Risø National Laboratory, Roskilde, Denmark

Perkins, R.W. and Thomas, C.W., 1980. World wide fallout. In W.C. Hanson (Ed.), Transuranic Elements in the Environment, DOE/TIC-22800. Springfield, VA: National Technical Information Service.

Outola, I., 2005. Seaweed SRM-4359 NIST Intercomparison. In press.

UNSCEAR, 1982. Ionizing Radiation: Sources and Biological Effects. United Nations Scientific Committee on the Effects of Atomic Radiation, 1982 Report to the General Assembly, United Nations, New York.

U.S. Air Force, 1970. Project Crested Ice. USAF Nuclear Safety, 65, 1-97.

Vibe, C., 1950. The marine mammals and the marine fauna in the Thule district (Northwest Greenland) with observations on ice conditions in 1939-41. Meddelelser om Grønland, 150, 1-117.

Risø's research is aimed at solving concrete problems in the society.

Research targets are set through continuous dialogue with business, the political system and researchers.

The effects of our research are sustainable energy supply and new technology for the health sector.