



## Thule-2007 - Investigation of radioactive pollution on land

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# Thule-2007 - Investigation of radioactive pollution on land

Risø-R-Report

Sven Poul Nielsen and Per Roos  
Risø-R-1781(EN)  
October 2011



**Author:** Sven Poul Nielsen and Per Roos  
**Title:** Thule-2007 - Investigation of radioactive pollution on land  
**Division:** Radiation Research

**Abstract (max. 2000 char.):**

A survey of radioactive pollution on land in the Thule area from an airplane accident in 1968 was carried out during 2007 and 2008. The results show levels of plutonium in soil at Narsaarsuk ranging from background values around 39 Bq m<sup>-2</sup> up to levels of 1.7 MBq m<sup>-2</sup>. Local sub-areas of sizes ranging from a few hundred to a few thousands of square metres show elevated levels above 10 kBq m<sup>-2</sup> of plutonium.

Based on geostatistical analysis, the total amount of plutonium in soil at Narsaarsuk is estimated at 270 GBq (100 g).

Investigations were carried out at Narsaarsuk to determine the occurrence of radioactive particles in air. This involved collection of airborne particles with an air sampler, collection of airborne particles on sticky foils, collection of rain samples and collection of particles that could be resuspended by wind from the soil surface to the air. Small amounts of plutonium were found in air and rain samples, but the derived concentrations in air were very low corresponding to typical background levels in Europe in the range 1-10 nBq m<sup>-3</sup>. A few small particles were found on the soil surface with activities up to 1000 Bq plutonium but the air and rain samples showed no sign of resuspension of such particles from the soil.

Local areas with elevated levels of radioactive pollution in soil were found at Kap Atholl and Grønnedal 15-20 km south of Narsaarsuk. Here the levels were lower than those in the Narsaarsuk area but clearly above background.

It was not possible within this project to carry out a systematic survey of the entire region. Hence, it cannot be excluded that there may be more local sub-areas than those identified with elevated levels of radioactive pollution in soil.

Screening surveys were carried out at Thule Air Base, Moriuaq, Saunders Island and Wolstenholme Island. The results showed background levels in soil at these locations which means that no sign of radioactive pollution from the accident was found at these locations.

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# Contents

|           |  |           |
|-----------|--|-----------|
| <b>1</b>  | <b>Introduction</b>                    | <b>5</b>  |
| <b>2</b>  | <b>Scope of Investigation</b>          | <b>5</b>  |
| <b>3</b>  | <b>Sampling</b>                        | <b>6</b>  |
| 3.1       | Field campaigns                        | 6         |
| 3.2       | Air and precipitation                  | 7         |
| 3.3       | Soil                                   | 10        |
| 3.4       | Soil particles                         | 13        |
| <b>4</b>  | <b>Analytical methods</b>              | <b>14</b> |
| 4.1       | Sample preparation                     | 14        |
| 4.2       | Autoradiography                        | 14        |
| 4.3       | Radiochemistry                         | 15        |
| 4.4       | Gamma spectrometry                     | 15        |
| 4.4.1     | Laboratory measurements                | 15        |
| 4.4.2     | Field measurements                     | 16        |
| 4.5       | Analytical quality                     | 19        |
| <b>5</b>  | <b>Results</b>                         | <b>19</b> |
| 5.1       | Air                                    | 19        |
| 5.1.1     | Air filters                            | 19        |
| 5.1.2     | Sticky foils                           | 20        |
| 5.2       | Precipitation                          | 22        |
| 5.3       | Soil                                   | 23        |
| 5.3.1     | Soil samples                           | 24        |
| 5.3.2     | Soil depth profiles                    | 28        |
| 5.3.3     | Field measurements                     | 30        |
| 5.3.4     | Vacuum bags                            | 32        |
| 5.3.5     | Geostatistical analysis                | 34        |
| 5.4       | Meteorological data                    | 37        |
| <b>6</b>  | <b>Discussion</b>                      | <b>37</b> |
| <b>7</b>  | <b>Conclusions</b>                     | <b>38</b> |
| <b>8</b>  | <b>Recommendations</b>                 | <b>38</b> |
| <b>9</b>  | <b>Acknowledgements</b>                | <b>39</b> |
| <b>10</b> | <b>References</b>                      | <b>40</b> |
|           | <b>Appendix A, Meteorological data</b> | <b>44</b> |



# 1 Introduction

On 21 January 1968, a US B52 bomber carrying nuclear weapons crashed on the sea ice approx. 15 km west of the Thule Air Base in North West Greenland. The aircraft caught fire on impact and the chemical explosives of the weapons detonated causing radioactive substances to be dispersed by burning jet fuel. Flames reached heights of about 850 m and smoke even higher. Based on eyewitness information and meteorological and radar observations from the Air Base, it was concluded that smoke from the fire presumably drifted towards south and south east. It was estimated that small particles could be carried far away by the wind and later deposited on ground at low concentrations. Measurable amounts of pollution were expected to be found towards the small settlement Narsaarsuk about 8 km south of the crash site. Redistribution of radioactive material might be expected from the heavily polluted crash site during storms on 24 and 29 January 1968 carrying the pollution towards west to Saunders Island (U.S. Air Force, 1970).

During the weeks after the accident, the dispersion of plutonium was investigated by collecting and analyzing snow samples. Pollution was identified in two zones, one in a southerly direction from the crash site due to direct fallout from the smoke plume and one towards west from resuspension of pollution from the area near the crash site. Plutonium contamination along the southern coastline of Bylot Sound showed maximum concentrations near Narsaarsuk with levels of up to about 9 kBq m<sup>-2</sup>.

Since 1968, further dispersion of the radioactive pollution on land is expected to have occurred due to influence of weather conditions. Redistribution of pollution is expected to be local and due mainly to snow melt and subsequent leaching and transport of particles.

In 2003 soil samples were collected from 8 locations along the coast near Narsaarsuk in connection with a previous project on environmental plutonium at Thule (Nielsen and Roos, 2006). The top soil layers showed accident plutonium at all locations and at varying concentrations. Plutonium in the soil samples was distributed very inhomogeneously and associated with particles. The uneven distribution of plutonium in soil from Narsaarsuk was further confirmed from a preliminary investigation involving limited sampling of soil in 2006. Narsaarsuk was abandoned as a settlement years ago and the wooden shacks are now used occasionally as hunting cabins and temporary shelter for tourists.

The purpose of the present project is to evaluate possible health risks to humans at Thule and particularly at Narsaarsuk from inhalation of radioactive particles if such particles are remobilized from the ground. Resuspension of particles could occur by influence of the wind and/or dust forming activities. Inhalation of particles containing plutonium may give rise to a health risk. The risk evaluation is described in a separate report by the Danish National Institute of Radiation Protection (NIRP, 2011).

## 2 Scope of Investigation

The project has covered the following items

- A systematic survey of radioactive pollution on land in the Narsaarsuk area
- A detailed mapping of locations with particularly high levels of radioactive pollution south and east of the hunting cabins at Narsaarsuk

- Investigation of the occurrence of radioactive particles in air at Narsaarsuk
- Screening for radioactive pollution on land at Grønnedal (Green Valley) hunting area, Kap Atholl recreation area, Saunders Island, Wolstenholme Island, Moriusaq settlement and Thule Air Base
- Providing basis for evaluating radiological risks to man from inhaling radioactive particles and going about in the area

## 3 Sampling

### 3.1 Field campaigns

During summer in 2007 and 2008 an investigation including on-site gamma spectrometric screening, sampling of soil, air and precipitation was carried out in the Narsaarsuk area (Fig.1), about 15 km west of the Thule Air Base (TAB), Greenland. Apart from minor equipment used for soil sampling which was transported by plane the majority of materials and sampling equipment were sent to Thule by ship in spring 2007. However, arrival at Thule became severely delayed due to ship collision with an iceberg in Baffin Bay and subsequent repair. Furthermore, one of the boat drivers contracted for the transportation in the local area was at hospital leaving us with one boat only thus making it more difficult to split up work in teams at several locations. The initial plans of conducting the complete sampling campaign during the summer of 2007 could therefore not be carried out which made it necessary to use the following summer, 2008, for complementary sampling. Equipment and laboratory was housed at TAB while some of the basic sampling equipment for soil was stored in the shacks at Narsaarsuk.

Transportation between TAB and Narsaarsuk took place in a small boat. After arrival of the sampling equipment in 2007, transportation on land could be made using three All Terrain Vehicles (ATV's) which also permitted sampling at more distant locations; prior to this transportation was by foot. Due to the lack of a majority of the sampling gear, the initial three weeks following arrival in 2007 was spent collecting soil samples in the Narsaarsuk area as well as on the Saunders and Wolstenholme Islands which was planned sampling locations. Similarly the Inuit settlement Moriusaq was visited for soil sampling. Complementary material needed for soil sampling was obtained from the local shop and friendly staff at TAB (Greenland Contractors, GC) who granted supply to the TAB technical supply store. GC-staff also made possible soil sampling at the more far away location Grønnedal since two boats were required for safety reasons. Consequently an extra boat was placed at our disposal for this purpose. The Kap Atholl site, a former navigational station and now a recreational site for tourists and TAB staff, located at the end of a road from TAB was reached by ATV. Following delivery at Thule of the equipment container, air sampler, passive particle collectors and the rain sampler were mounted at Narsaarsuk. Initial problems with fuel supply to the generator for electricity used for the air sampler delayed start of air sampling with a few days. It was discovered that direct sunlight on the fuel tubing feeding the generator created gas bubbles which prevented flow.

Meteorological conditions at Narsaarsuk were recorded by a weather station, WeatherLink 5.7a (2006) Davies Instruments Corporation, CA, USA, (Fig. 2). The station logged temperature, wind speed, wind direction and precipitation every five minutes to a battery-powered data logger. The sampling campaign in summer 2008 included mounting of the air sampler, passive collectors and rain collector at the same positions as in 2007. Additional passive collectors were mounted on locations

eastwards of Narsaarsuk. The main part of the time in 2008 was spent on localizing areas with elevated levels of radioactivity using the portable gamma spectrometers.

Frozen soil samples were transported back to Risø laboratories by air in 2007. Remaining samples from 2007 and 2008 (except soil which was transported by air) were transported back with the equipment container by ship in 2008.

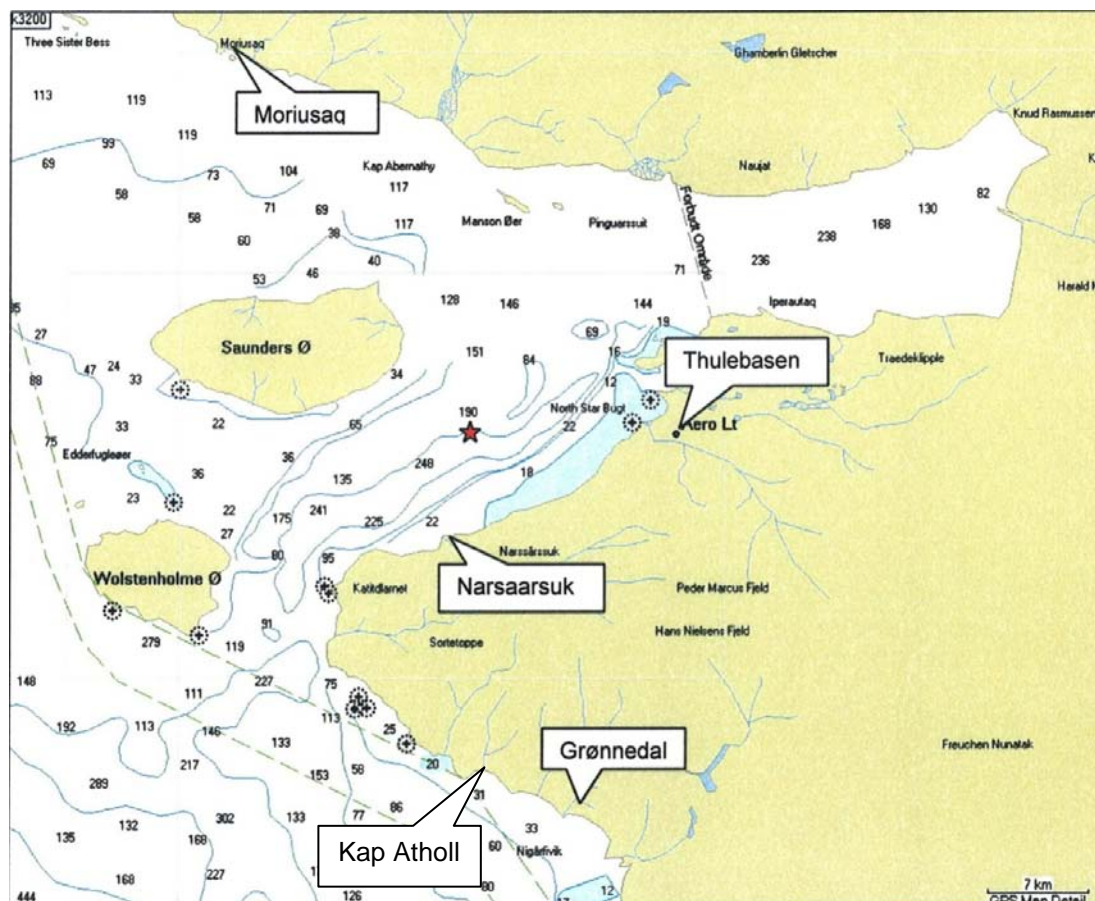


Figure 1. Overview of the Thule area. The site of the airplane crash is marked with a red star.

### 3.2 Air and precipitation

Aerosols were sampled at Narsaarsuk using a Staplex<sup>®</sup> Model TFIA-2F high volume air sampler with built-in flow adjuster. The sampler was mounted with an 8"x10" filter holder, flow-meter and constant flow controller in an outdoor aluminium housing (Fig. 3). The air-sampler was calibrated using Staplex<sup>®</sup> calibration kit for 8"x10" holders and was set to a flow rate of about  $1.5 \text{ m}^3 \text{ min}^{-1}$  before shipping. Sampling was done using glass fibre filters (Staplex<sup>®</sup> TFAGF810) with reported retention of  $0.3 \mu\text{m}$  particles better than 99.97%. The sampler was positioned and secured to the ground at the centre of Narsaarsuk. The sampler was powered by a petrol driven generator (Fig. 4). In 2007, air sampling was carried out during 1 – 13 August but not continuously, as there were frequent stops over the first days due to formation of gas in the fuel tubing supplying the generator, causing it to stop. The total volume of air sampled in 2007 was  $28575 \text{ m}^3$ . The air sampler pump was replaced in 2008 due to break down. Two air samples were collected, 3-14 July 2008 ( $22440 \text{ m}^3$ ) and 14 July – 13 August 2008 ( $51265 \text{ m}^3$ ). Mounting and removing of the filters were done using disposable gloves and showing care not to contaminate filters with any soil



present on equipment or hands. Filters were stored in double plastic bags until laboratory analysis.



*Figure 2. Weather-station sensors at Narsaarsuk recording wind speed, wind direction, temperature and precipitation.*



*Figure 3. Air sampler at Narsaarsuk*



*Figure 4. Generator used for the air sampler. Tubing from fuel tank on roof to generator protected from direct sunlight to avoid gas formation.*



*Figure 5. Passive aerosol collector placed on one of the shacks at Narsaarsuk.*



*Figure 6. Rain collector at Narsaarsuk.*

Passive aerosol collectors made of double-sided adhesive tape (sticky vinyl, size 43x54 cm) were attached to wooden frames and placed at 1-2m height above ground on the wooden shacks in Narsaarsuk (Fig. 5). In 2007 nine collectors were placed on the shacks and three on a wooden frame placed at the location with elevated levels close to Narsaarsuk. During transportation back and from Thule the adhesive surface

was protected from aerosols by a wooden plate. Exposure time for the passive collectors in 2007 was about two weeks, and in 2008 about 50 days.

Precipitation was collected through a 0.14 m<sup>2</sup> funnel placed over a 50 L polyethylene flask positioned at 1 m above ground on a tripod some 10 m away from the air sampler (Fig. 6). The flask and funnel were protected against wind by a metal cylinder connected to the tripod and secured to ground by wires. In 2007 sampling was carried out during 1–13 August and a total amount of 2.3 L rain was collected while in 2008 three individual samples were collected during 3–14 July (1 L), 14 July – 13 August (8.3 L) and 13–24 August (5.4 L). Following transportation back to Risø laboratories the samples were acidified to pH 1 using hydrochloric acid and stored for further treatment. Fig. 7 shows the set-up of the air sampler, weather station and rain sampler in Narsaarsuk.



Figure 7. View of hunting cabins at Narsaarsuk showing locations of air sampler, weather station and rain sampler.

### 3.3 Soil

Soil sampling was performed using an ordinary garden spoon and a 10 cm diameter by 30 cm long plastic-tube drilled at the edge to form a sharp saw-tooth profile. On most locations 5 individual samples (A-E) were taken in a pre-defined spiral-pattern (Fig. 7) with individual distances of 20, 60, 200 and 500 cm. This procedure was followed in order to assess spatial variability on each location. Sampling depths were largely determined by soil properties and stones but were in general between 4 and 10 cm. Photographs were taken at each soil sample hole in order to evaluate terrain type. Overview pictures were similarly taken to evaluate overall terrain. In order to better cover the entire area of Narsaarsuk a number of single soil samples were also taken. This was also done on a location known from the 2006 Narsaarsuk survey

sampling to hold significant plutonium levels. On some few locations bulk samples were collected by adding together soil collected from 5 smaller tubes (4 cm diameter). Collected soil samples were stored frozen at TAB awaiting transport back to Risø laboratories. During 2007 and 2008 a total of 534 + 84 soil samples were collected. Together with the soil samples collected during the survey in 2006 (56 samples) and during the expedition in 2003 (24 samples) a total of 698 soil samples have been collected. Each sampling site was marked with a unique number and the position taken using a GPS (Fig. 9). At a number of locations soil samples were collected by depth to provide information on depth distribution of the radioactive pollution (Figs. 10 and 11).

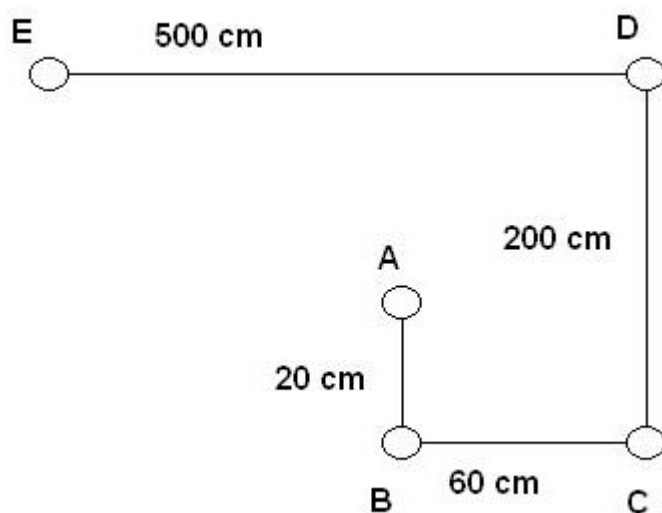


Figure 8. Schematic view of the geometry used for sampling of soil. Each location was given a unique number followed by labels A through E for the 5 samples taken.



*Figure 9. Sampling of soil and recording of GPS coordinates.*



*Figure 10. Sampling of soil for depth distribution 0-13 cm, sample no. 2008-4031/39, 11-Jul-2008.*



Figure 11. Sites for soil samples collected in 2007 and 2008 (↕) for depth distributions of Pu/Am at Narsaarsuk. Red rectangle indicates area with elevated levels on ground. Locations of hunting cabins near the coast are indicated.

### 3.4 Soil particles

In order to get information on radioactivity in small particles on the ground which might potentially be available for resuspension, a number of locations were selected in the Narsaarsuk area where an area of one square metre was vacuumed thoroughly. Fig. 12 shows a map of the Narsaarsuk area indicating the locations of the hunting cabins and the sites where vacuuming was carried out. Fig. 13 illustrates the sampling procedure.



Figure 12. Locations at Narsaarsuk for vacuuming of surface soil.

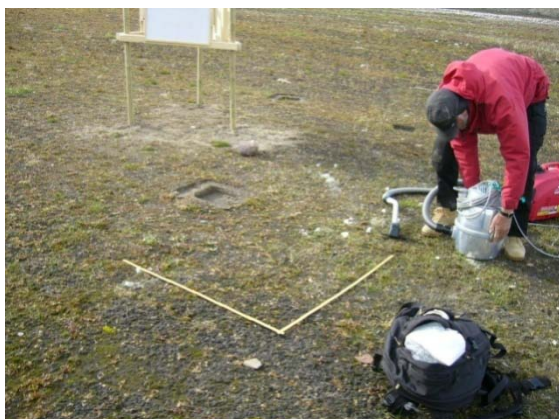


Figure 13. Sampling of particles from a square metre of surface soil using a vacuum cleaner. Sticky-foil frames and collection of soil depth profile in background.

## 4 Analytical methods

### 4.1 Sample preparation

Sample preparation was carried out to reduce volume and mass of samples prior to radiochemical and radiometric analysis. Standard pre-treatment techniques were applied including freeze drying and ashing at 450 °C. Samples were sieved to remove stones larger than 1.5 mm. Methods used for different sample types are summarised in Table 1.

During evaporation of the precipitation samples care was taken not to lose particulate material on beaker walls or other surfaces. Evaporation was done in progressively smaller beakers and finally in 200 mL containers calibrated for gamma spectrometry.

Table 1. Methods used for preparation of samples prior to analysis.

| Sample type   | Freeze drying | Sieving | Ashing | Evaporation | Chemical treatment |
|---------------|---------------|---------|--------|-------------|--------------------|
| Precipitation |               |         |        | X           | X                  |
| Sticky foil   |               |         | X      |             | X                  |
| Air filter    |               |         |        |             | X                  |
| Vacuum bag    |               | X       | X      |             |                    |
| Soil          | X             | X       | X      |             | (X)                |

### 4.2 Autoradiography

An Amersham Bioscience Storm 820 imaging system was used for autoradiography of passive aerosol collectors. Wooden plates protecting the passive collectors were removed shortly before initiating the autoradiographic measurements and 6 µm Mylar foils were placed on the sticky surfaces to allow for the phosphorus screen to be placed on the foil without adhering to it. A mounted storage Phosphor screen (Gen-

eral Purpose screens, 35x43 cm) was placed directly on the Mylar foil and the wooden protective plate put back on place to prevent light and dust from reaching the collector/screen. Collector and phosphorus screen were then inserted in a black plastic bag during the exposure time which lasted 3 – 14 days. Following exposure the screen was removed and placed in the read-out unit and the image visualized and stored using associated image software.

### 4.3 Radiochemistry

Following gamma spectrometric analysis of evaporated precipitation, the samples were transferred to glass beakers using water and acid and then evaporated to dryness. Following addition of known amounts of  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  tracers, the samples were digested on a hot plate using aqua regia under watch glass over night. Plutonium was separated out using ion exchange chromatography (Chen et al., 2001) and electroplated onto stainless steel disks. The disks were analysed for Pu isotopes using solid state alpha spectrometry. Counting times were 5-10 days. The Am-fraction of each sample was set aside for future analysis.

Glass fibre air filters were folded and pressed into 200 mL containers for gamma spectrometry and later dissolved using HF/HNO<sub>3</sub> in the presence of known amounts of  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  tracers. After removing excess HF by repeated evaporation and addition of boric acid, samples were treated as the precipitation samples above.

Passive aerosol collectors were dry ashed at 500 °C overnight and the ash leached in aqua regia in the presence of known amounts of  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  tracers and later analysed following the same procedure as precipitation samples above.

### 4.4 Gamma spectrometry

#### 4.4.1 Laboratory measurements

Gamma spectrometric analyses were applied to samples to detect gamma-emitting radionuclides including  $^{241}\text{Am}$ . The occurrence of small amounts of  $^{241}\text{Pu}$  (14.3 y half life) in the weapons plutonium from the accident has caused build up over time of the radioactive decay product  $^{241}\text{Am}$  (433 y half life). Build up of  $^{241}\text{Am}$  from  $^{241}\text{Pu}$  occurs slowly over 73 y to a maximum at 3% of the initial  $^{241}\text{Pu}$  activity levels. Therefore,  $^{241}\text{Am}$  in samples from Thule is used as an indicator of accident plutonium. Americium-241 is detectable by gamma spectrometry due to emission of 60 keV gamma rays while plutonium isotopes are not easily detected by gamma spectrometry and generally require radiochemical preparation and detection by alpha or mass spectrometry. Previous investigations during 2001-2003 of  $^{241}\text{Am}$  and plutonium isotopes in sediment and soil samples from Thule have shown activity ratios of  $^{241}\text{Am}/^{239+240}\text{Pu}$  of 0.16-0.4 (Nielsen and Roos, 2006). Therefore, the activity of  $^{239+240}\text{Pu}$  in soil is 3-6 times higher than the  $^{241}\text{Am}$  activity found.

Soil and vacuum-bag samples were analysed by gamma spectrometry after freeze drying (soil only), ashing at 450 °C and removal of stones larger than 1.5 mm. Samples were mixed and subdivided into aliquots which were all analysed to ensure that all gamma-emitting radionuclides above detection limit would be registered. Experience has shown that accident plutonium (and  $^{241}\text{Am}$ ) in soil samples from Thule are associated with particles and therefore distributed inhomogeneously across all sample sizes. So in order to minimise the risk of not detecting a high-activity particle in a sample, all sample material was analysed. This differs with standard procedures according to which a single aliquot after homogenisation is assumed to represent the entire sample. Gamma spectrometric analysis of soil thus proved to be a



rather comprehensive task of the present project involving about 600 samples collected in 2007 and 2008 and analyses of more than 3000 aliquots.

The gamma-spectrometric analyses were carried out with high-purity Ge detectors placed in 10-cm lead shields. The detectors were calibrated with mixed nuclide gamma-ray standard solutions (including  $^{241}\text{Am}$ ) purchased from Amersham plc. Sample geometries varied from vials with volumes of a few millilitres measured in well detectors to cylindrical containers with volumes of up to a hundred millilitres measured with coaxial detectors. Aliquots in cylindrical geometries had a maximum thickness of 10 mm in order to reduce effects of self absorption. The amounts of  $^{241}\text{Am}$  in aliquots determined by gamma spectrometry are assumed to be correct within a factor of 2 as determined from previous investigations of soil inhomogeneities due to high-activity particles (Nielsen and Roos, 2006).

#### 4.4.2 Field measurements

Field measurements of  $^{241}\text{Am}$  on the ground were carried out with two sets of portable equipment using 3"x3" NaI detectors (SAM-935, Berkeley Nucleonics Corporation). The equipment accumulates and stores gamma spectra from which the presence of  $^{241}\text{Am}$  is assessed by a spectrum peak at 60 keV. Measurements were carried out by placing detectors vertically either directly on ground or by holding them at 1 m above ground, Figs. 14 and 15. In order to reduce influence on detectors of gamma radiation from horizontal directions, the detectors were supplied with circular shielding made of 2 mm copper and 6 mm lead.



Figure 14. Portable gamma spectrometer for field measurement of  $^{241}\text{Am}$  with detector placed on ground.



Figure 15. Portable gamma spectrometer for field measurement of  $^{241}\text{Am}$  with detector at 1 m above ground.

The detectors were calibrated by relating count rate when carried 1 m above ground to concentration of  $^{241}\text{Am}$  by surface area. This was accomplished with a certified  $^{241}\text{Am}$  point source (400 kBq) from which the 60-keV count rate was registered along a line at 1 m distance from the detector perpendicular to the detector axis, Figs. 16 and 17. The detector response was folded with corresponding surface areas yielding calibration factor as a function of area considered, Table 2 and Fig 18. The calibration factor corresponds to an ideal case of exposing the detector to a smooth homogeneous plane source of  $^{241}\text{Am}$  which does not correspond to the field situation at Thule with uneven terrain and inhomogeneous distribution of  $^{241}\text{Am}$  on ground. However, the detector integrates and averages radiation in the field from a surface area on the ground of several square metres. Therefore, estimates of surface area concentrations based on field measurements give important data supplementing that derived from laboratory analyses of soil samples representing surface areas of 0.01  $\text{m}^2$  only. Due to the strong attenuation of  $^{241}\text{Am}$  gamma rays in just a few centimetres of soil, field gamma spectrometric measurements of  $^{241}\text{Am}$  have limited sensitivity and do not so efficiently detect the presence of very low levels of  $^{241}\text{Am}$  when present. This is the case when the source material is partly buried in soil and causes an underestimation of the area deposition of  $^{241}\text{Am}$  (and therefore Pu).

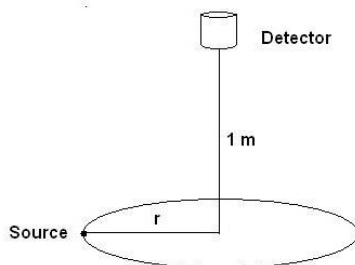


Figure 16. Calibration setup for NaI detector with point source placed at axis off-set distance  $r$ .

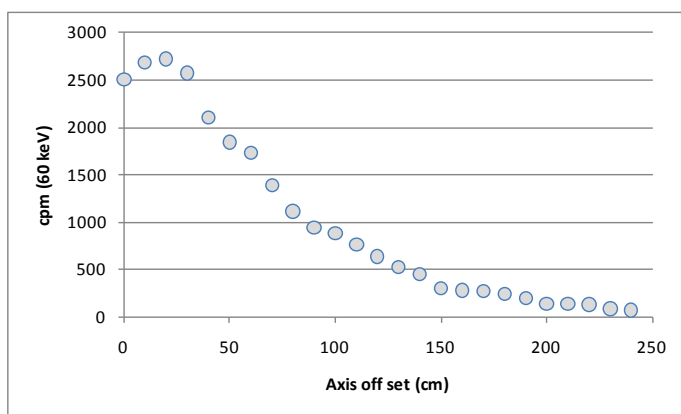


Figure 17. Response of NaI detector to 60 keV gamma rays from  $^{241}\text{Am}$  point source placed perpendicular to the detector axis on a line at 1 m distance.

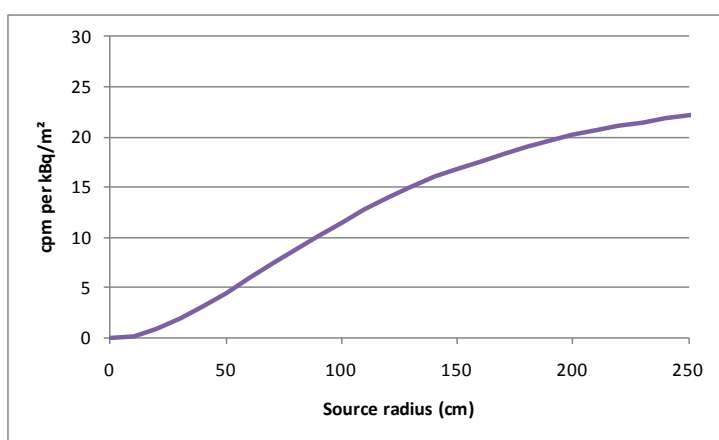


Figure 18. Calibration factor as a function of source area at 1 m distance.

Data was selected from a number of locations where soil samples were taken and field measurements made within distances less than approx. 10 m. Soil concentrations versus detector count rate (with detector on ground and at 1 m) are given in Fig. 19, which also shows values of the calibration factor of 10, 15 and 24 cpm per  $\text{kBq}/\text{m}^2$  corresponding to different source areas considered. The low correlation between concentration and count rate is due to terrain roughness and non-uniform soil concentrations. Data from locations with the detector on ground is close to the maximum theoretical value of the calibration factor ( $24 \text{ cpm per kBq}/\text{m}^2$ ). A calibration factor value of  $15 \text{ cpm per kBq}/\text{m}^2$  was chosen for converting detector count rate in the field at 1 m to surface concentration on ground of  $^{241}\text{Am}$ .

Table 2. Calibration factors for portable NaI detector to a smooth plane  $^{241}\text{Am}$  source at 1 m distance as a function of source area.

| Source radius (m) | Source area ( $\text{m}^2$ ) | Calibration factor (cpm per $\text{kBq}/\text{m}^2$ ) |
|-------------------|------------------------------|---|
| 0.9               | 2.5                          | 10  |
| 1.3               | 5                            | 15  |
| 4                 | 50                           | 24  |

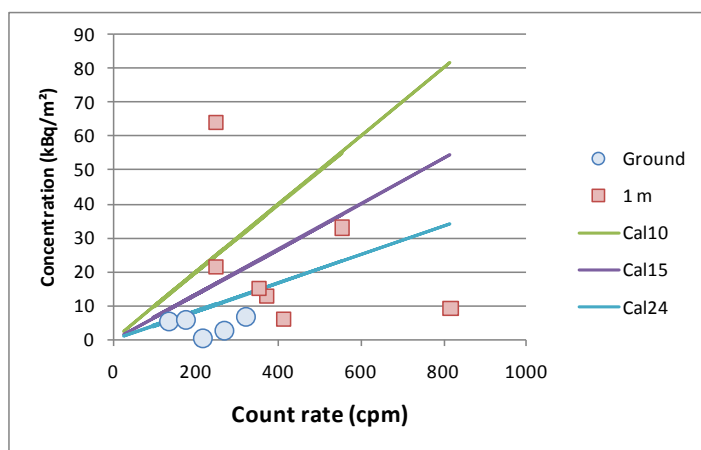


Figure 19. Concentration of  $^{241}\text{Am}$  on ground ( $\text{kBq m}^{-2}$ ) versus detector count rate (cpm) for different calibration factors compared with results from laboratory analyses of soil samples collected near sites where observations were made with detector on ground (Ground) and at 1 m above ground (1 m).

#### 4.5 Analytical quality

Reliability of Risø's analytical results on plutonium isotopes,  $^{241}\text{Am}$  and other radionuclides is tested regularly by participation in international intercomparison exercises on laboratory 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, 2000b, 2001, 2004, 2005, 2007a, 2007b, 2009a, 2009b, 2009c, 2010a, 2010b; Nielsen, 1996, 2004; NPL, 2006, 2008; Outola et al., 2008).

## 5 Results

### 5.1 Air

#### 5.1.1 Air filters

Data on plutonium from the active aerosol sampler is shown in Table 3. Plutonium was detected in filters from all three collection periods although at very low levels. The low Pu concentrations measured in the current set of air filters did not allow for detection of the low-abundance  $^{238}\text{Pu}$  isotope thus preventing the use of the isotope ratio  $^{238}\text{Pu}/^{239+240}\text{Pu}$  to tell about the origin of the plutonium. The visual appearance of the filters following collection showed very low mass loading indicating very low concentrations of resuspended material during the collection periods.

Table 3. Air concentrations of  $^{239+240}\text{Pu}$  obtained from the active aerosol sampler at Narsarsuk.

| Collection period        | Volume of air ( $\text{m}^3$ ) | $^{239+240}\text{Pu}$ ( $\text{nBq m}^{-3}$ ) | 1 sd (%) |
|--------------------------|--------------------------------|---|----------|
| 1-13 August 2007         | 28575                          | 1.4   | 40       |
| 3-14 July 2008           | 22440                          | 4.6   | 24       |
| 14 July – 13 August 2008 | 51265                          | 4.3   | 13       |

These concentrations may be compared with levels recorded from routine monitoring of plutonium in air performed by the US Environmental Measurements Laboratory at the Thule Air Base during the years 1966-1976 (NUSTL, 2011). In those years Pu-concentrations were about three orders of magnitude higher than today, in the range 0.1-10  $\mu\text{Bq m}^{-3}$  (Fig. 20), due to atmospheric nuclear weapons testing. During 1990-1998 average concentrations of  $^{239+240}\text{Pu}$  in samples of ground-level air collected quarterly in Northern Germany were observed in the range 0.4-4  $\text{nBq m}^{-3}$  (Wershofen and Arnold, 1999). Concentrations of  $^{239+240}\text{Pu}$  in surface air in Prague during 1997-2006 were found in the range 0.5-5  $\text{nBq m}^{-3}$  (Hölgye, 2008).

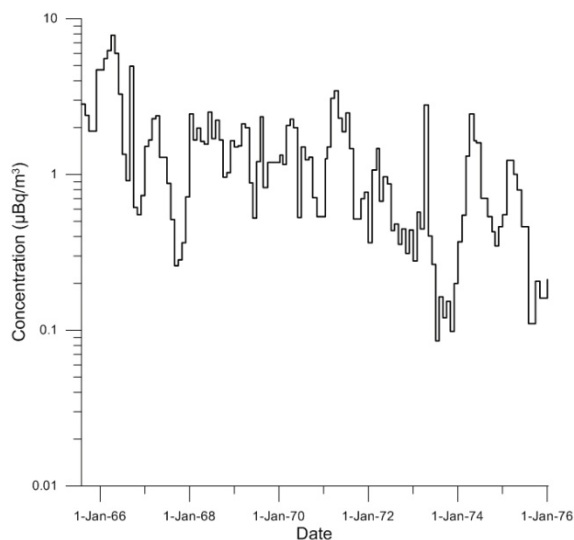


Figure 20. Plutonium-239,240 levels in air at Thule recorded by US Environmental Measurements Laboratory during 1966-1976 (NUSTL, 2011).

### 5.1.2 Sticky foils

None of the 24 autoradiographic images of the passive aerosol collectors showed any sign of high-activity particles or excess radioactivity. Variable background conditions, position on the screen, instrument scanning properties, exposure time of the phosphorus screen and integration area of the exposed screen all influence the detection limit in this method. Assuming all activity appearing as discrete particles, an approximate detection limit of 50 mBq of alpha radioactivity can be assigned to an exposure time of one day.

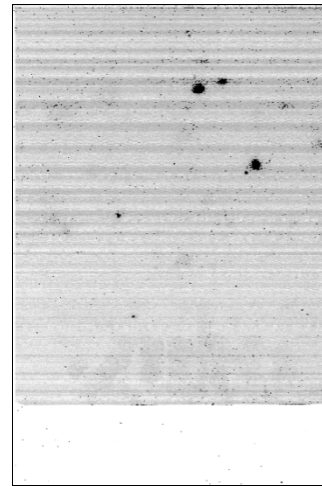
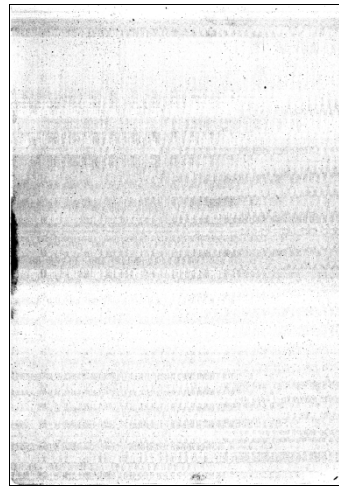


Figure 21. Autoradiography image A.

Figure 22. Autoradiography image B.

Figure 23. Autoradiography image C.

Table 4. Data on  $^{239+240}\text{Pu}$  found on passive aerosol collectors showing collector position, Pu activity found on collector, analytical uncertainty, and derived air concentration. DL in third column (1 sd) indicates that plutonium is below the detection limit the value of which is given in the second column. The fourth column shows calculated values of air concentrations assuming average wind speed of 1 m/s, constant air concentrations and that the passive collectors trap all plutonium bearing particles present in air during the exposure period (14 days in 2007 and 50 days in 2008).

| Collector position, direction and year | $^{239+240}\text{Pu}$ (mBq) | 1 sd (%) | Calculated air concentration ( $\text{nBq m}^{-3}$ ) |
|--|-----------------------------|----------|--|
| House 1 West 2007                      | 0.12                        | 11       | 0.44   |
| House 2 South 2007                     | 0.02                        | 50       | 0.06   |
| House 4 West 2007                      | 0.08                        | 38       | 0.30   |
| House 8 South 2007                     | 0.03                        | 50       | 0.11   |
| House 7 West 2007                      | 0.01                        | DL       | 0.05   |
| House 5 South 2007                     | 0.04                        | 28       | 0.15   |
| House 3 East 2007                      | 0.11                        | 15       | 0.38   |
| House 6 East 2007                      | 0.08                        | 20       | 0.29   |
| House 9 East 2007                      | 0.16                        | 12       | 0.58   |
| House 2 South 2008                     | 0.03                        | DL       | 0.03   |
| House 3 West 2008                      | 0.07                        | DL       | 0.08   |
| House 4 South 2008                     | 0.04                        | 30       | 0.05   |
| House 5 South 2008                     | 0.06                        | 20       | 0.07   |
| House 2 East 2008                      | 0.08                        | 35       | 0.10   |
| House 2 West 2008                      | 0.47                        | 14       | 0.55   |
| House 2 South 2008                     | 0.05                        | 50       | 0.05   |
| House 1 West 2008                      | 0.22                        | 20       | 0.25   |
| Hotspot-1 2007 South                   | 0.56                        | 6        | 0.65   |
| Hotspot-1 2007 Northwest               | 0.06                        | 20       | 0.07   |
| Hotspot-1 2007 East                    | 0.53                        | 12       | 0.62   |
| Hotspot-1 2008 South                   | 0.37                        | 16       | 0.43   |
| Hotspot-1 2008 Northwest               | 0.74                        | 11       | 0.86   |
| Hotspot-1 2008 Northeast               | 1.04                        | 12       | 1.20   |

Two autoradiography images of passive aerosol collectors and one autoradiography image of a Thule soil sample with two high-activity particles present are shown in

Figs. 21-23 where images A and B are from passive collectors and image C from the soil sample. Exposure times for images A and B were 4 and 6 days respectively, while image C had an exposure time of 12 h only.

Dissolution of the passive aerosol collectors followed by chemical separation and solid state alpha spectrometry showed the presence of low levels of plutonium. The highest amount of plutonium on a passive collector was 1 mBq (Table 4).

Interpretation of plutonium found on the passive collectors in terms of air concentrations is not straight forward. The intention was originally to use data on ambient stable elements like Al, Ti or Zr from both passive and active aerosol collectors in order to calculate the equivalent volume of air to which the passive collectors were exposed. The concentrations of stable elements in the passive collectors were, however, entirely made up of small amounts of blank material in the collectors themselves. The amount of resuspended material on the collectors was visibly observed to be very low or even not detectable.

Rough estimates of the air concentrations from the passive collector data were made assuming average wind speed of 1 m/s and constant air concentrations and that the passive collectors trap all plutonium bearing particles present in air during the exposure period. The activity concentrations obtained in this way are given in Table 4. A graphical summary of air concentration data from the active aerosol sampler and calculated from passive collectors are given in Fig. 24.

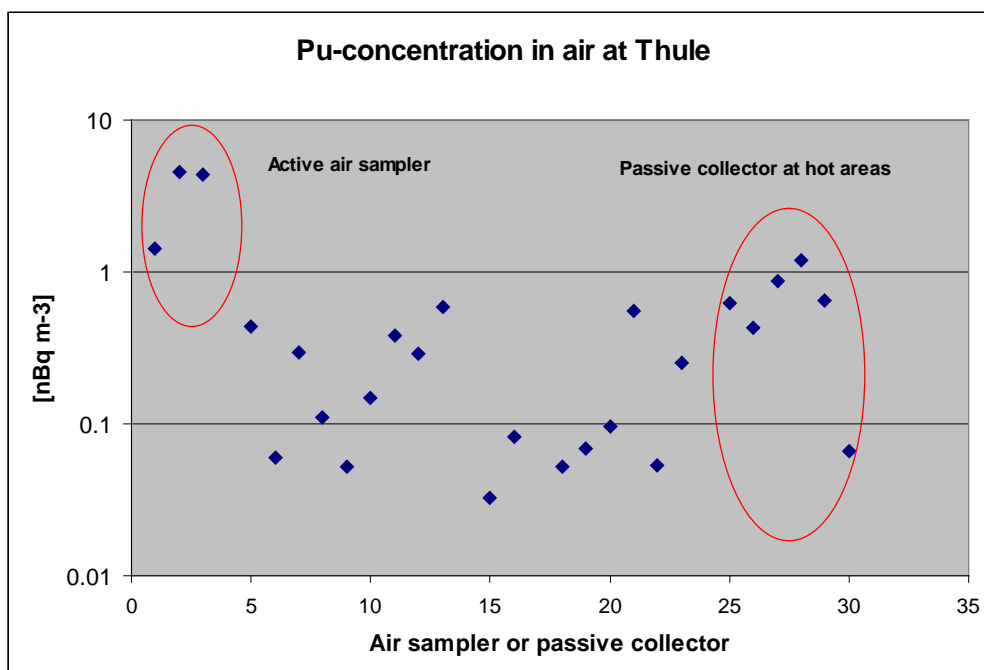


Figure 24. Summary of Pu-concentrations in air obtained from the active air sampler and concentrations calculated from passive collectors using data in Table 4.

## 5.2 Precipitation

Results of analyses of  $^{239+240}\text{Pu}$  in samples of precipitation are shown in Table 5. Data on concentrations of Pu in air and rain from samples collected simultaneously are rare. However, data is available on fallout radionuclides and on  $^7\text{Be}$  and  $^{210}\text{Pb}$  which shows that so-called washout or scavenging ratios of volume concentrations in rain to those in air are in the order of  $10^6$  (Aarkrog, 1979; McNearly and Baskaran,

2003). In the current data set we may use the  $^7\text{Be}$  data from the collected precipitation and air samples, this show a wash-out ratio of about  $2 \cdot 10^6$ . Application of this ratio to the precipitation data of plutonium yields corresponding air concentrations in the sub  $\mu\text{Bq m}^{-3}$  range which is 2-3 orders of magnitude higher than actually measured (see Fig. 24). This probably reflects a combination of the stochastic nature of collecting resuspended plutonium particles and that washout ratios for radioisotopes which are well mixed in the lower atmosphere do not apply to locally resuspended radioisotopes.

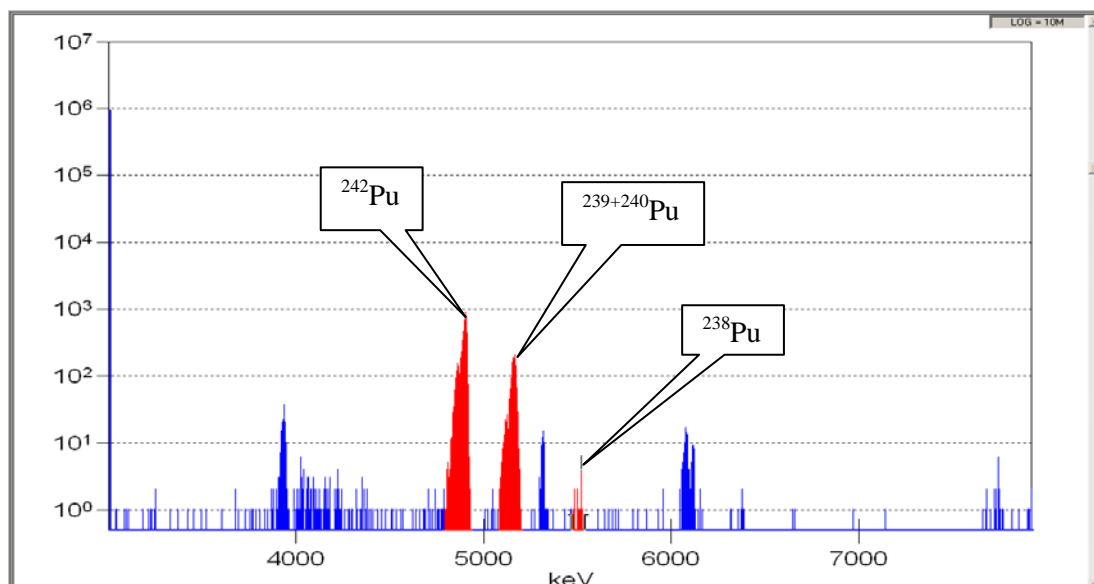


Figure 25. Plutonium alpha spectrum from rain sample collected at Narsaarsuk during 14 July – 13 August 2008.

Table 5. Concentrations of  $^{239+240}\text{Pu}$  in precipitation collected at Narsaarsuk.

| Collection period        | Volume (L) | $^{239+240}\text{Pu}$ (mBq L <sup>-1</sup> ) | 1 sd (%) |
|--------------------------|------------|--|----------|
| 1-13 August 2007         | 2.3        | 0.01   | 40       |
| 3-14 July 2008           | 1.9        | 0.51   | 8        |
| 14 July – 13 August 2008 | 8.3        | 0.80   | 6        |
| 13-24 August 2008        | 5.4        | 0.42   | 11       |

The plutonium alpha spectrum from the rain sample collected during 14 July – 13 August 2008 is shown in Fig. 25. The concentration of  $^{239+240}\text{Pu}$  in the sample was  $0.8 \text{ mBq L}^{-1}$  (total  $6.6 \text{ mBq}$ ). The relatively high concentration and the low  $^{238}\text{Pu}/^{239+240}\text{Pu}$  ratio of 0.015 indicate plutonium originating from the accident in 1968. A comparison between volumes of precipitation collected by the rain sampler (RS) and values recorded by the weather station (WS) indicates a systematic difference of 30% ( $V_{\text{WS}}/V_{\text{RS}} = 0.7$ ).

### 5.3 Soil

Results on  $^{241}\text{Am}$  in soil were obtained from field measurements and from laboratory analyses of soil samples covering surface samples and depth profiles.



### 5.3.1 Soil samples

Sampling of soil during 2003-2008 comprised 24 soil cores collected in 2003, 56 in 2006, 534 in 2007 and 84 in 2008. Soil sampling in 2007 was particularly comprehensive and covered Green Valley, Kap Atholl, Wolstenholme Island, Saunders Island, Moriusaq and Thule Air Base in addition to the Narsaarsuk area. Soil sampling in 2008 focused on areas with elevated levels. Locations for soil samples collected during 2003-2008 are shown in Fig. 26. Graphical overviews of the data on  $^{241}\text{Am}$  are given in Fig. 27 showing a histogram of the results above detection limit by year of collection and in Fig. 28 showing the data by location in Box-Whisker plots. The latter show minimum, maximum, median, lower quartile and upper quartile values for the data.

The histogram distribution of the results is bimodal with lower levels of  $^{241}\text{Am}$  in the range  $0.002\text{--}1\text{ kBq m}^{-2}$  peaking at about  $0.04\text{ kBq m}^{-2}$ , and higher levels in the range  $1\text{--}300\text{ kBq m}^{-2}$  peaking at about  $10\text{ kBq m}^{-2}$ . Summaries of  $^{241}\text{Am}$  results by area are given in Table 6 showing number of samples, minimum and maximum values, arithmetic means and standard deviations, and geometric means and geometric standard deviations.

Table 6. Summary of  $^{241}\text{Am}$  concentrations in soil by location ( $\text{kBq m}^{-2}$ ).

| Area                | Number of samples | Minimum ( $\text{kBq m}^{-2}$ ) | Maximum ( $\text{kBq m}^{-2}$ ) | Arithmetic mean ( $\text{kBq m}^{-2}$ ) | Arithmetic standard deviation ( $\text{kBq m}^{-2}$ ) | Geometric mean ( $\text{kBq m}^{-2}$ ) | Geometric standard deviation |
|---------------------|-------------------|---------------------------------|---------------------------------|---|---|--|------------------------------|
| Thule Air Base      | 29                | < 0.002                         | 0.038                           | 0.020                                   | 0.011   | 0.017                                  | 1.8                          |
| Moriusaq            | 8                 | < 0.002                         | 0.031                           | 0.017                                   | 0.008   | 0.015                                  | 1.6                          |
| Saunders Island     | 17                | < 0.002                         | 0.056                           | 0.019                                   | 0.013   | 0.016                                  | 1.9                          |
| Wolstenholme Island | 16                | < 0.002                         | 0.053                           | 0.022                                   | 0.017   | 0.017                                  | 2.1                          |
| Green Valley        | 28                | 0.004                           | 7.4                             | 0.87                                    | 1.8   | 0.16                                   | 7.3                          |
| Kap Atholl          | 40                | < 0.002                         | 19                              | 1.6                                     | 3.4   | 0.27                                   | 9.3                          |
| Narsaarsuk          | 428               | < 0.002                         | 280                             | 6.6                                     | 21  | 0.28                                   | 15                           |

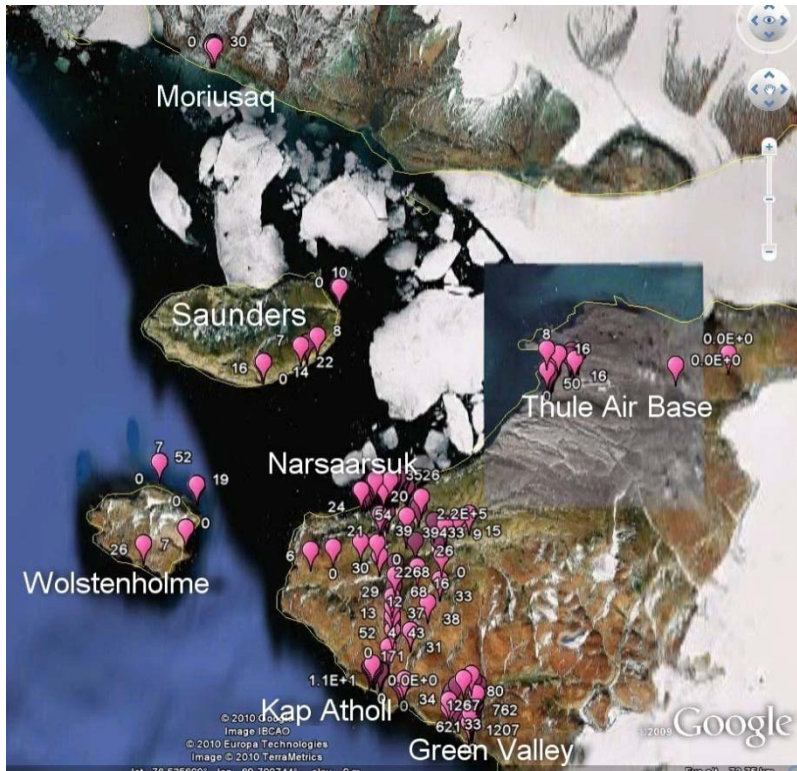


Figure 26. Locations for soil samples collected during 2003-2008.

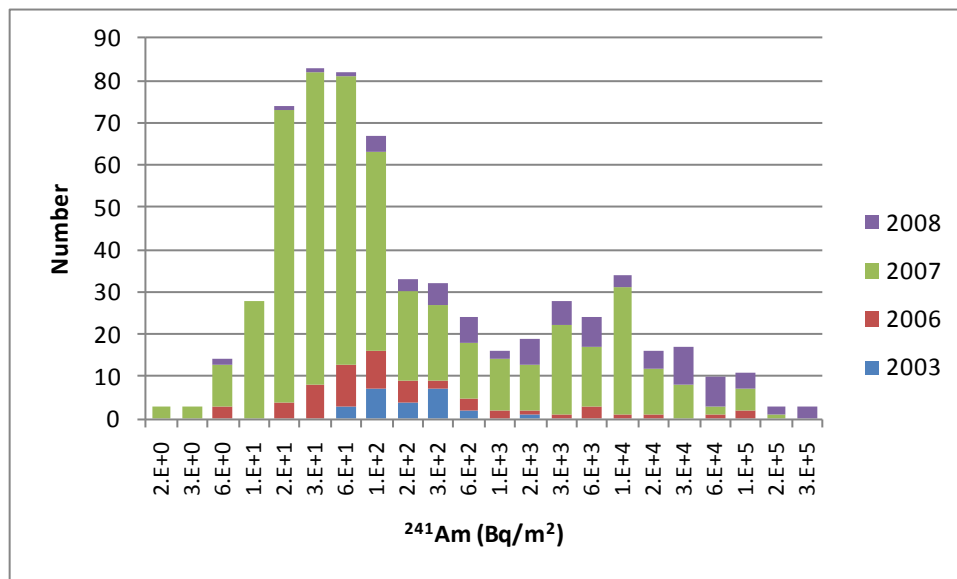


Figure 27. Histogram of  $^{241}\text{Am}$  in soil ( $\text{Bq m}^{-2}$ ) determined from laboratory analyses of soil samples collected during 2003-2008.

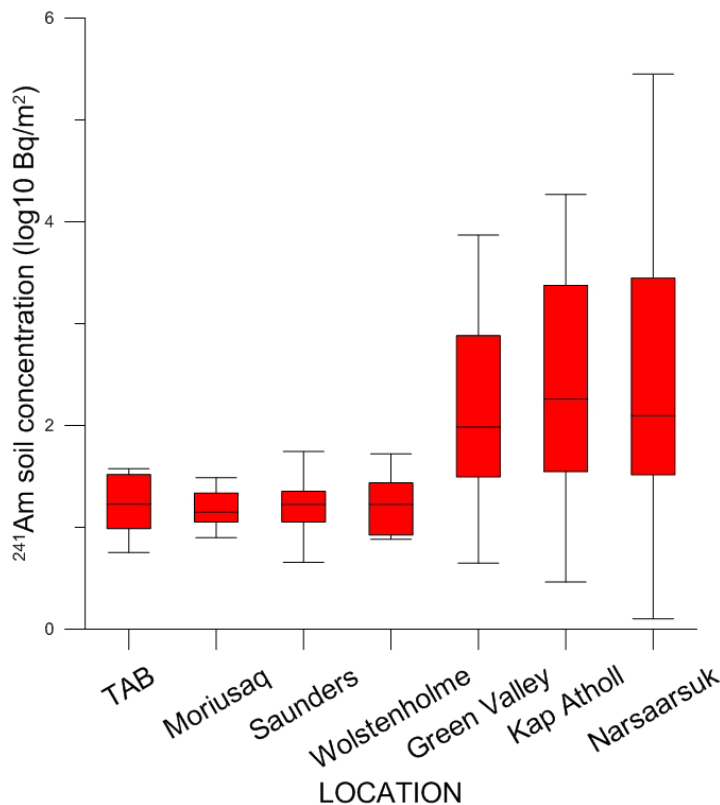


Figure 28. Box-Whisker plots of <sup>241</sup>Am soil concentrations (log<sub>10</sub> Bq m<sup>-2</sup>) by location.

Concentrations in soil of <sup>241</sup>Am at Thule Air Base, Moriusaq, Saunders Island and Wolstenholme Island are low and at background levels. Concentrations at Kap Atholl, Green Valley and at Narsaarsuk, however, are higher and clearly influenced by radioactive pollution from the accident in 1968. Highest concentrations are found at Narsaarsuk which also shows the highest variability. A number of sampling locations in the different areas showed non detectable area deposition of <sup>241</sup>Am. These locations are in principle those having very little or no vegetation (eg. sandy and/or stony areas).

Concentrations of plutonium in soil are estimated from those of <sup>241</sup>Am by taking into account variation of <sup>241</sup>Am/<sup>239+240</sup>Pu ratios from 0.16 which is characteristic for plutonium from the Thule accident (Nielsen and Roos, 2006) to 0.4 which is characteristic for background levels of global fallout (Michel et al., 2002). Radiochemical analyses of soil samples collected at Narsaarsuk in 2003 with <sup>241</sup>Am concentrations in the range 0.05-1 kBq m<sup>-2</sup> showed an average Am/Pu ratio of 0.27. Therefore, for <sup>241</sup>Am concentrations in soil below 0.05 kBq m<sup>-2</sup> we use a Pu/Am ratio of 3, for <sup>241</sup>Am concentrations in the range 0.05-1 kBq m<sup>-2</sup> we use a Pu/Am ratio of 4 and for <sup>241</sup>Am concentrations above 1 kBq m<sup>-2</sup> we use a Pu/Am ratio of 6.

The estimated concentrations of plutonium in soil are summarized by location in Table 7 and shown in Box-Whisker plots in Fig. 29. Concentrations are low and at background levels at Thule Air Base, Moriusaq, Saunders Island and Wolstenholme Island whereas concentrations at Kap Atholl, Green Valley and Narsaarsuk are significantly higher and show large variability. The integrated deposition density of <sup>239+240</sup>Pu on the northern hemisphere due to global fallout from nuclear weapons testing is 39 Bq m<sup>-2</sup> (UNSCEAR, 1982).

Table 7. Summary of  $^{239+240}\text{Pu}$  concentrations in soil by location ( $\text{kBq m}^{-2}$ ).

| Area                | Number of samples | Minimum ( $\text{kBq m}^{-2}$ ) | Maximum ( $\text{kBq m}^{-2}$ ) | Arithmetic mean ( $\text{kBq m}^{-2}$ ) | Arithmetic standard deviation ( $\text{kBq m}^{-2}$ ) | Geometric mean ( $\text{kBq m}^{-2}$ ) | Geometric standard deviation |
|---------------------|-------------------|---------------------------------|---------------------------------|---|---|--|------------------------------|
| Thule Air Base      | 29                | < 0.006                         | 0.11                            | 0.059                                   | 0.032   | 0.050                                  | 1.8                          |
| Moriusaq            | 8                 | < 0.006                         | 0.092                           | 0.050                                   | 0.025   | 0.045                                  | 1.6                          |
| Saunders Island     | 17                | < 0.006                         | 0.22                            | 0.062                                   | 0.053   | 0.049                                  | 2.0                          |
| Wolstenholme Island | 16                | < 0.006                         | 0.21                            | 0.074                                   | 0.071   | 0.054                                  | 2.3                          |
| Green Valley        | 28                | 0.013                           | 44                              | 5.0                                     | 11  | 0.62                                   | 9.2                          |
| Kap Atholl          | 40                | < 0.006                         | 110                             | 9.5                                     | 21  | 1.2                                    | 12                           |
| Narsaarsuk          | 428               | < 0.006                         | 1700                            | 39                                      | 130   | 1.1                                    | 19                           |

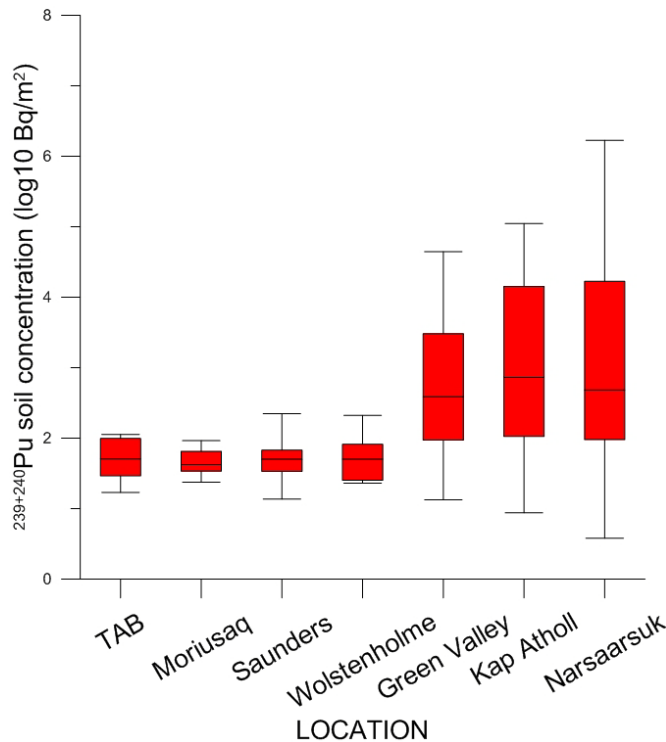


Figure 29. Box-Whisker plots of  $^{239+240}\text{Pu}$  soil concentrations ( $\log_{10} \text{Bq m}^{-2}$ ) by location.

### 5.3.2 Soil depth profiles

In 2003 soil cores were collected at 8 locations in the Narsaarsuk area to investigate depth distributions of plutonium. Results showed that accident plutonium was found in all samples and very inhomogeneously distributed with maximum concentrations in upper soil layers and  $^{239+240}\text{Pu}$  levels in the range  $0.01\text{-}7\text{ kBq m}^{-2}$  (Nielsen and Roos, 2006).

In 2007 and 2008 this was further investigated by collecting samples from one of the areas at Narsaarsuk with elevated levels of accident plutonium (Hot Spot 1 about 400 m south of the hunting cabins, Fig. 30). Soil profiles were obtained from 12 sites within 90 m from each other and with soil concentrations of  $^{241}\text{Am}$  in the range  $6\text{-}86\text{ kBq m}^{-2}$  corresponding to  $^{239+240}\text{Pu}$  concentrations in the range  $40\text{-}500\text{ kBq m}^{-2}$ . Maximum concentrations were generally found in the upper soil layers, 0-2 cm, containing up to 90% of the whole area inventory (Figs. 31-42).

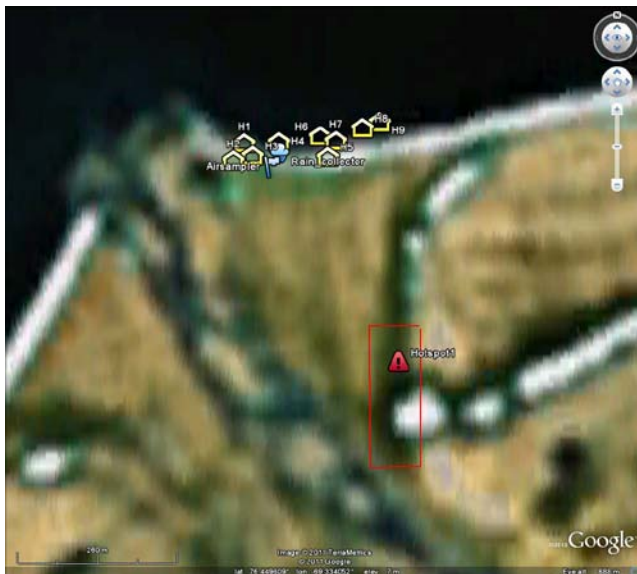


Figure 30. Hot Spot 1 (red rectangle) about 400 south of the hunting cabins at Narsaarsuk.

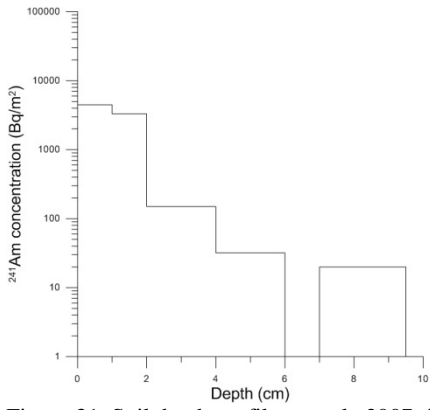


Figure 31. Soil depth profile, sample 2007-4762. Total  $^{241}\text{Am}$  concentration  $8.0 \text{ kBq m}^{-2}$ .

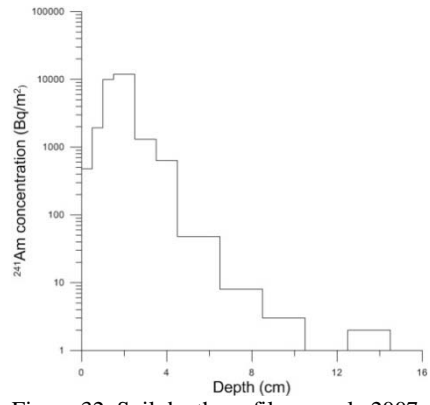


Figure 32. Soil depth profile, sample 2007-4763. Total  $^{241}\text{Am}$  concentration  $26 \text{ kBq m}^{-2}$ .

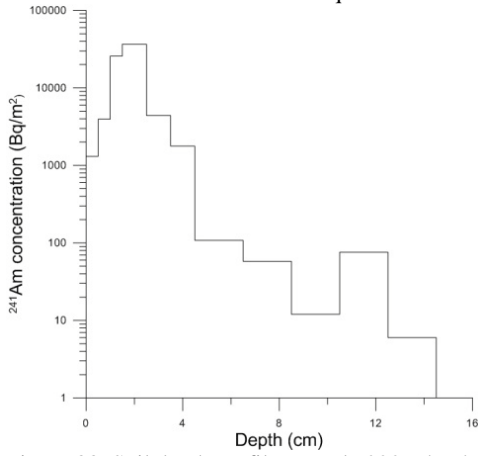


Figure 33. Soil depth profile, sample 2007-4764. Total  $^{241}\text{Am}$  concentration  $74 \text{ kBq m}^{-2}$ .

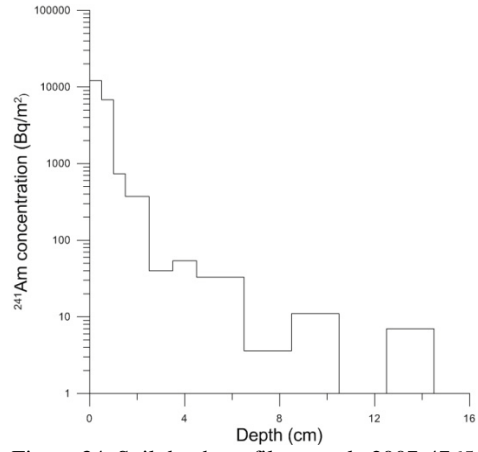


Figure 34. Soil depth profile, sample 2007-4765. Total  $^{241}\text{Am}$  concentration  $20 \text{ kBq m}^{-2}$ .

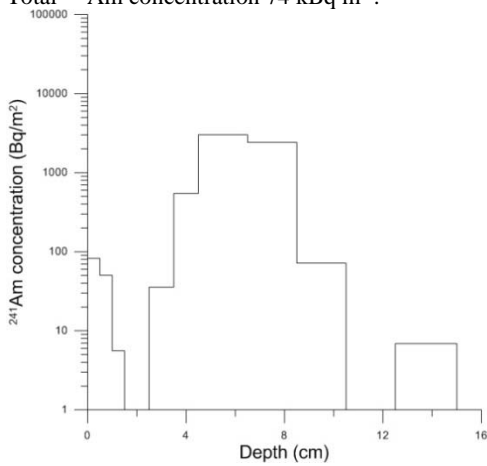


Figure 35. Soil depth profile, sample 2007-4766. Total  $^{241}\text{Am}$  concentration  $6.2 \text{ kBq m}^{-2}$ .

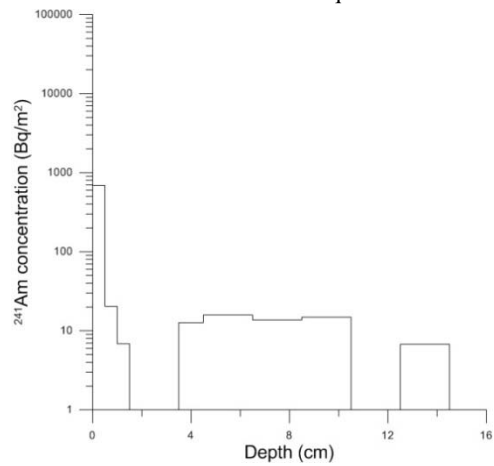


Figure 36. Soil depth profile, sample 2007-4767. Total  $^{241}\text{Am}$  concentration  $0.8 \text{ kBq m}^{-2}$ .

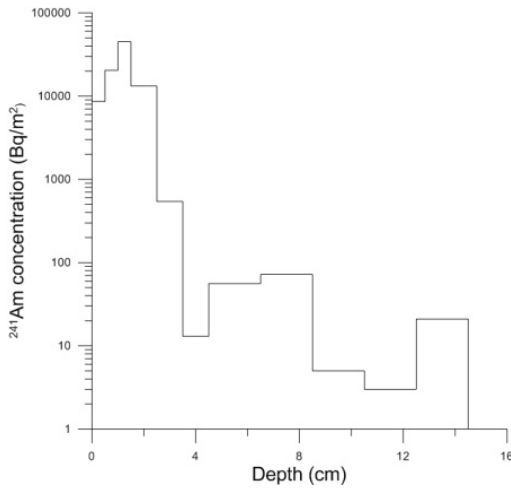


Figure 37. Soil depth profile, sample 2007-4768. Total  $^{241}\text{Am}$  concentration  $86 \text{ kBq m}^{-2}$ .

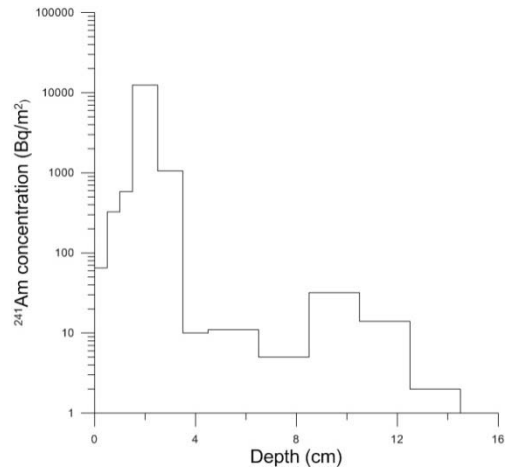


Figure 38. Soil depth profile, sample 2007-4769. Total  $^{241}\text{Am}$  concentration  $15 \text{ kBq m}^{-2}$ .

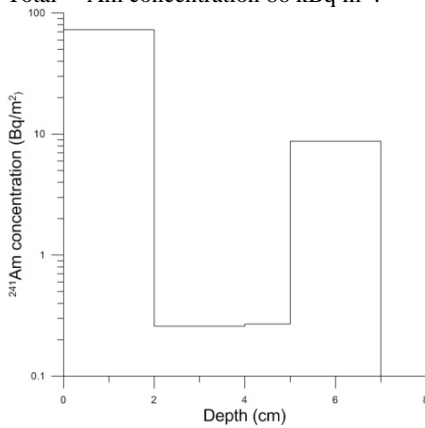


Figure 39. Soil depth profile, sample 2007-7308. Total  $^{241}\text{Am}$  concentration  $5.5 \text{ kBq m}^{-2}$ .

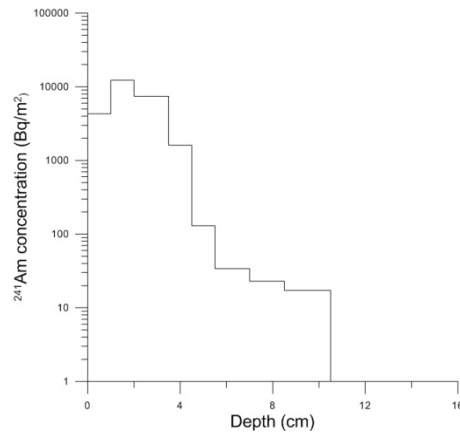


Figure 40. Soil depth profile, sample 2008-4031/39. Total  $^{241}\text{Am}$  concentration  $26 \text{ kBq m}^{-2}$ .

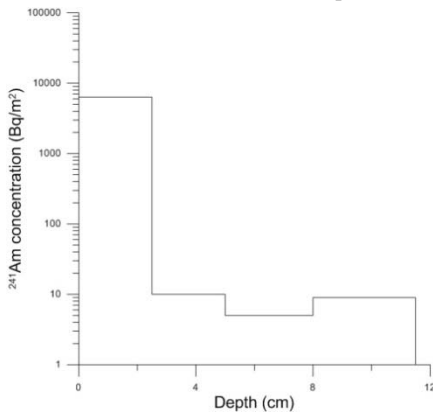


Figure 41 Soil depth profile, sample 2008-4040/43. Total  $^{241}\text{Am}$  concentration  $6.4 \text{ kBq m}^{-2}$ .

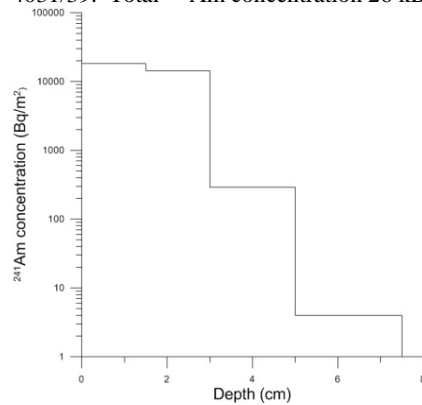


Figure 42 Soil depth profile, sample 2008-4044/47. Total  $^{241}\text{Am}$  concentration  $33 \text{ kBq m}^{-2}$ .

### 5.3.3 Field measurements

A large number of field measurements were carried out with portable equipment as a screening procedure to identify areas with elevated levels of  $^{241}\text{Am}$  on ground. Fig. 43 shows locations for field measurements made with detector at 1 m above ground.

The results are summarised in Table 8 showing number of samples, minimum and maximum values, arithmetic mean and standard deviation, geometric mean and

geometric standard deviation. The histogram distribution of the results is shown in Fig. 44. The variability of the results is lower than that from analyses of soil samples. The results of field measurements of  $^{241}\text{Am}$  on ground range between 2 and 50  $\text{kBq m}^{-2}$ .

An overview of  $^{241}\text{Am}$  levels on ground based on analyses of soil samples and field measurements is shown in the histogram in Fig. 45. The data here shows a distribution where the bimodal structure is more pronounced than from the soil data alone (Fig. 27). The reason is that the field measurements focused on areas with elevated levels of contamination.



Figure 43. Map of Narsaarsuk area showing hunting cabins and locations for field measurements of  $^{241}\text{Am}$  on ground.

Table 8. Summary of concentrations of  $^{241}\text{Am}$  in soil from field measurements.

| Number of locations | Minimum ( $\text{kBq m}^{-2}$ ) | Maximum ( $\text{kBq m}^{-2}$ ) | Arithmetic mean ( $\text{kBq m}^{-2}$ ) | Arithmetic standard deviation ( $\text{kBq m}^{-2}$ ) | Geometric mean ( $\text{kBq m}^{-2}$ ) | Geometric standard deviation |
|---------------------|---------------------------------|---------------------------------|---|---|--|------------------------------|
| 180                 | 1.5                             | 48                              | 13                                      | 8   | 10                                     | 2.0                          |



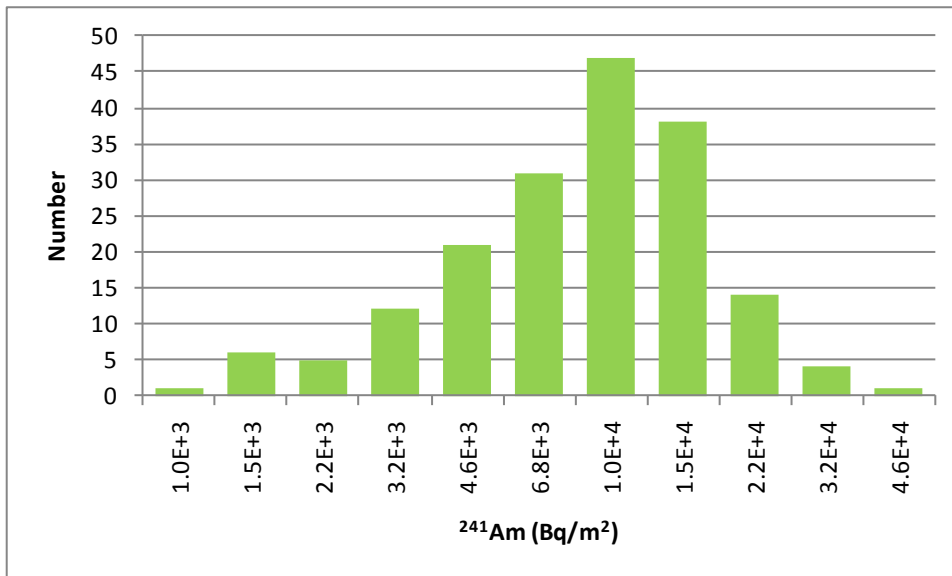


Figure 44. Histogram of data from field measurements of <sup>241</sup>Am made at 1 m above ground (Bq m<sup>-2</sup>).

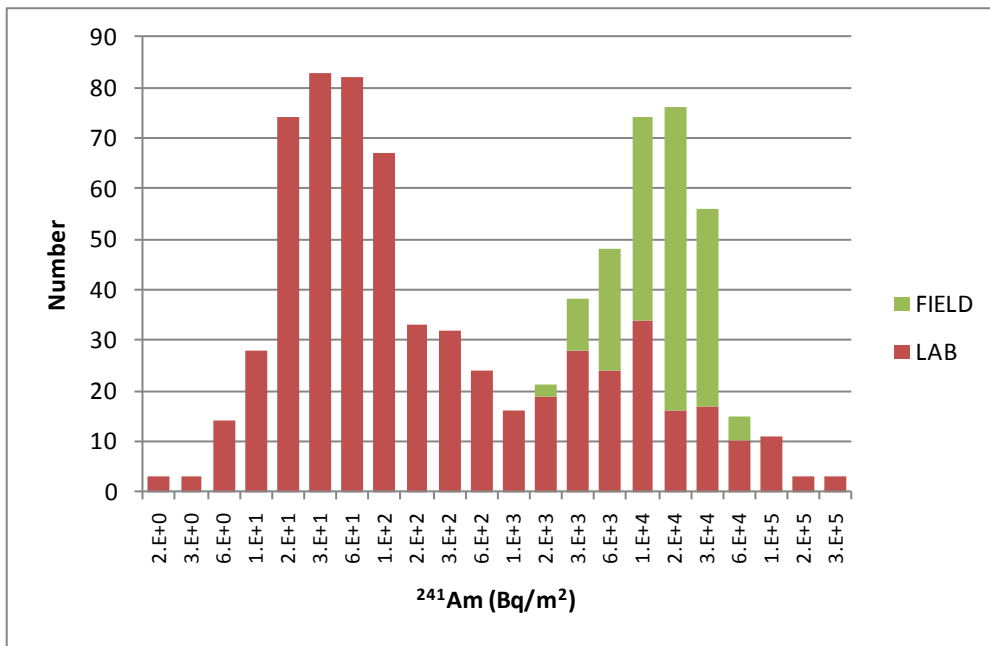


Figure 45. Histogram of data on <sup>241</sup>Am in soil (Bq m<sup>-2</sup>) determined from laboratory analyses of soil samples and field measurements.

### 5.3.4 Vacuum bags

Gamma spectrometric analyses of dust collected in vacuum bags showed presence of <sup>137</sup>Cs and <sup>241</sup>Am, but below detection limit for <sup>241</sup>Am at five locations. No radiochemical plutonium analyses were carried out for these samples. A scatter plot (Fig. 46) shows the results of <sup>241</sup>Am and <sup>137</sup>Cs versus mass of dust collected by vacuuming one square metre of soil surface. The data indicate that the levels of the two radionuclides are somewhat correlated with the mass of dust collected which means that the two radionuclides to some degree are associated with the same dust particles.

The correlations indicate that similar focusing processes are acting on the two radionuclides which have very different origin. Caesium-137 is due to fallout from atmospheric nuclear weapons tests while  $^{241}\text{Am}$  is due to local pollution from the accident in 1968. The results show two outliers where significantly more  $^{241}\text{Am}$  was found. These samples were collected a few hundred metres south of the hunting cabins and the results are due to particles with elevated levels of radioactivity. Histogram distributions of the two radionuclides are shown in Figs. 47 and 48 with geometric mean values of 0.7 and 0.2  $\text{Bq m}^{-2}$  for  $^{137}\text{Cs}$  and  $^{241}\text{Am}$ , respectively.

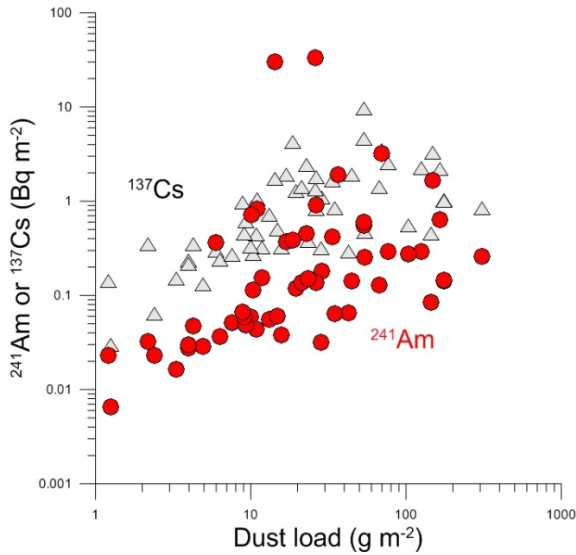


Figure 46. Scatter plot of  $^{241}\text{Am}$  (red circles,  $\text{Bq m}^{-2}$ ) and  $^{137}\text{Cs}$  (grey triangles,  $\text{Bq m}^{-2}$ ) vs. dust ( $\text{g m}^{-2}$ ) collected by vacuuming surface soil at Narsaarsuk.

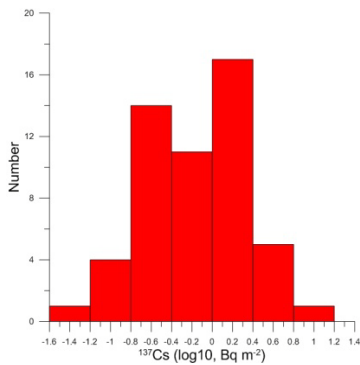


Figure 47. Histogram of  $^{137}\text{Cs}$  in samples ( $\text{Bq m}^{-2}$ ) collected by vacuuming surface soil at Narsaarsuk (geometric mean 0.7  $\text{Bq m}^{-2}$ , geometric standard deviation 3).

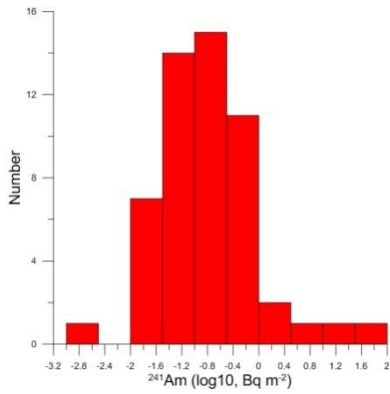


Figure 48. Histogram of  $^{241}\text{Am}$  in samples ( $\text{Bq m}^{-2}$ ) collected by vacuuming surface soil at Narsaarsuk (geometric mean  $0.2 \text{ Bq m}^{-2}$ , geometric standard deviation 6).

### 5.3.5 Geostatistical analysis

The data on  $^{241}\text{Am}$  in soil ( $\text{Bq m}^{-2}$ ) was analysed by JSA-EnviroStat (2011) using geostatistical methods which give detailed information on spatial trends and permit assessment of the total inventory of the pollution. The analysis was based on the combined data from laboratory analyses of soil samples and from field measurements. The data analysis was carried out on the  $\log_{10}$ -transformed data.

The data was split in geographical regions as shown in Fig. 49. Region 1 covered the Narsaarsuk area and southwards including Grønnedal. Region 2 was a subset of region 1 and covered the Narsaarsuk area. Region 3 covered Moriusaq, region 4 covered Saunders Island, region 5 covered Wolstenholme Island and region 6 covered Thule Air Base.

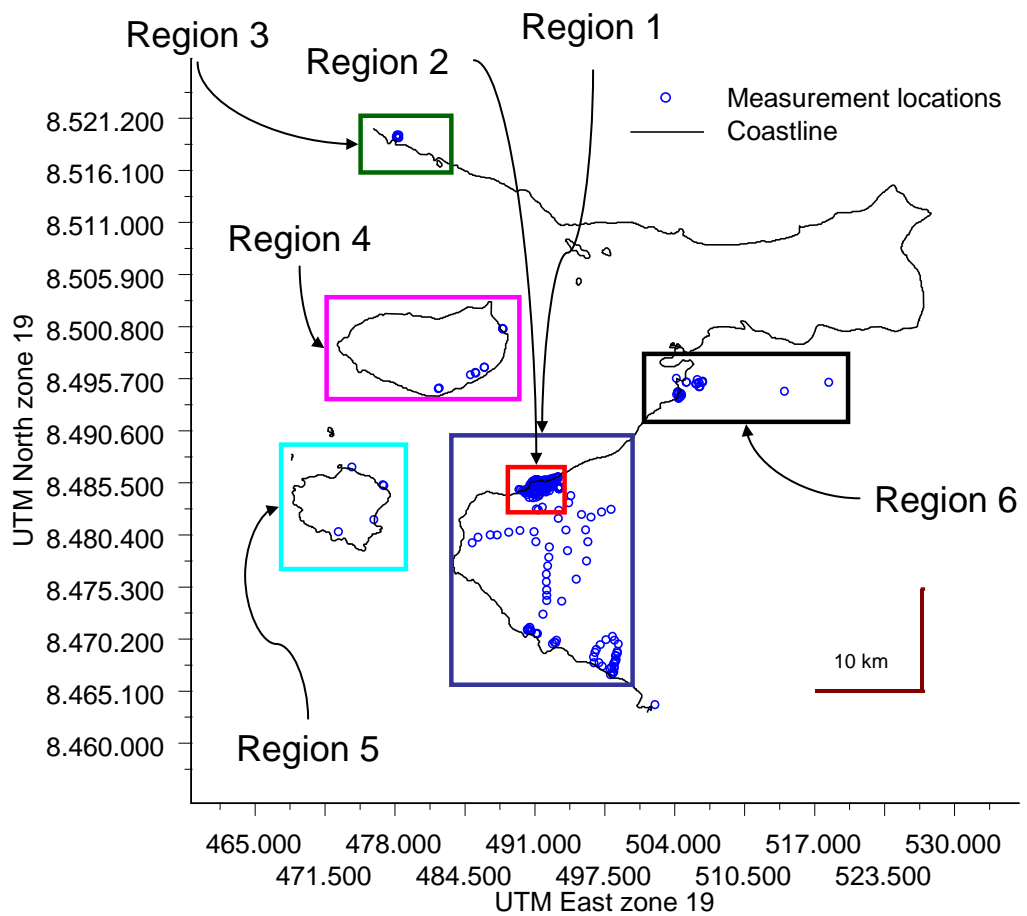
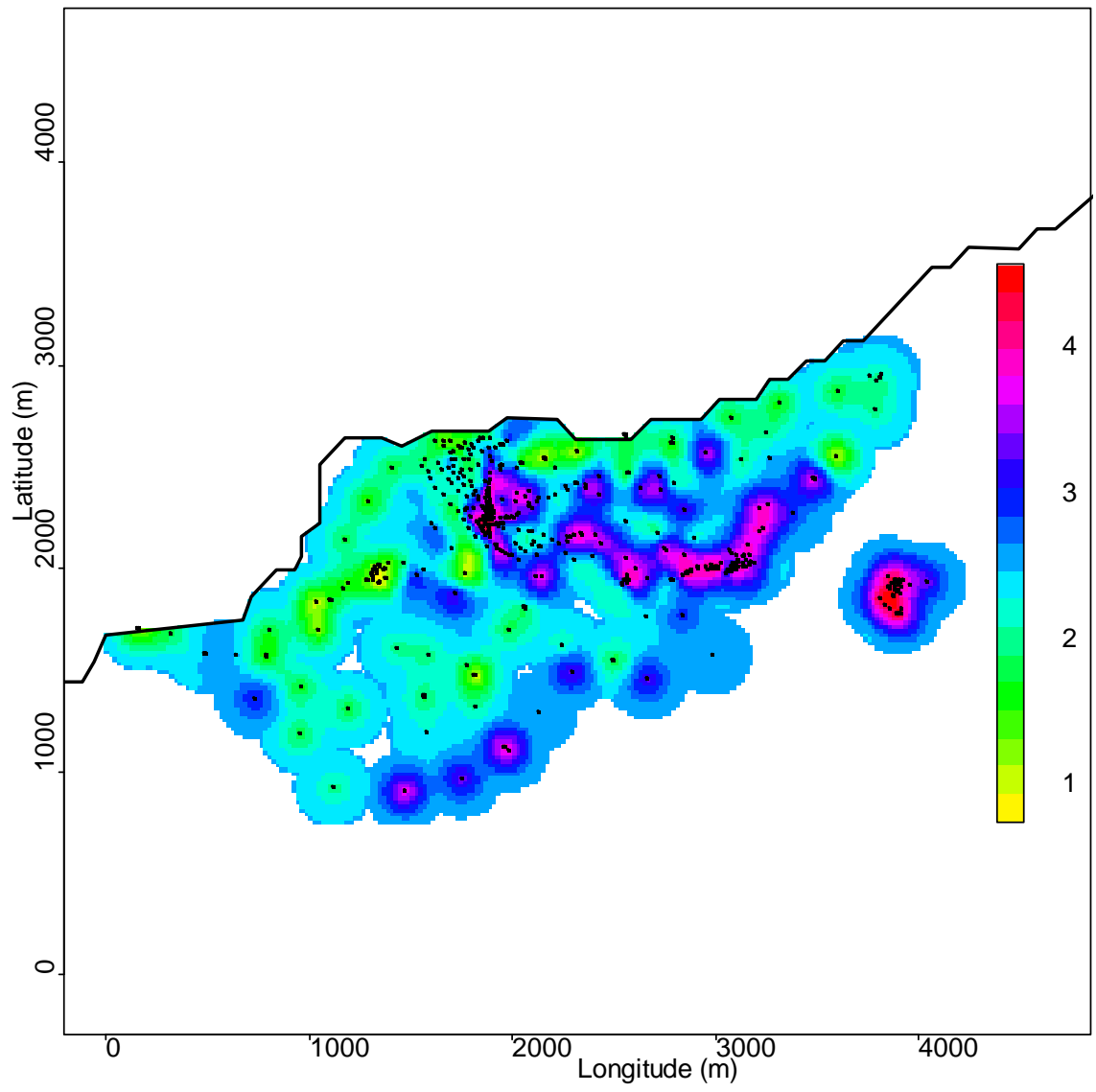


Figure 49. Subdivision of the Thule area in six regions. Measurement locations are marked with circles.

The number of observations in regions 3, 4, 5 and 6 were too small for geostatistical analysis. The analysis of data in region 1 showed a clear spatial correlation. However, due to the size of region 1 and the sampling performed, large areas are far from the nearest observations which means that estimates of levels in these areas should not be based on the observations. Further sampling would be needed to conclude anything in these areas. In addition, region 1 is sampled so sparsely that it makes no sense trying to predict the overall amounts of  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$ .



## 5.4 Meteorological data

The weather station was set up at Narsaarsuk during the field campaigns recording temperature, wind speed and direction, barometric pressure and precipitation. The data is shown in graphs in Appendix A.

## 6 Discussion

The distribution of radioactive pollution on land in the Thule area shows features not known previously. Levels of  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in soil range from low background levels to high levels caused by the accident in 1968. Background levels are due to fallout from atmospheric nuclear weapons testing. The levels at Narsaarsuk are clearly above those at Kap Atholl and Grønnedal and are relatively high in about 10 small sub-areas. These sub-areas are generally moist and wet locations facing north, indicating that the high levels are probably due to redistribution of contaminated snow during the first weeks or months after the accident followed by snow melt in the summer of 1968. Distribution of pollution on land at Narsaarsuk is very inhomogeneous and not consistent with a simple pattern of fallout from a radioactive plume extending from the accident site.

The elevated levels of radioactivity on land found at Kap Atholl and Grønnedal indicate that significant atmospheric transport of radioactive material occurred to distances of more than 20 km from the accident site, due to the 800-m high plume rise caused by the intense fire following the accident. Therefore, there may be more sub-areas with high levels of pollution than those identified near Narsaarsuk.

The results of the investigations do not indicate that radioactive particles in the air occur to an extent that give rise to concern. The investigation of radioactive particles in air has focused on locations in the Narsaarsuk area with high levels of pollution, and several sampling techniques have been employed.

Since wildlife in the area provides a local source for human food, it remains to be investigated the extent to which this food is contaminated with radioactivity. Animals foraging on locations with high levels of radioactive pollution in soil are likely to ingest amounts of radioactivity that may subsequently be transferred to man. Vegetation in the area is scarce and generally found on moist and wet locations including those with high levels of radioactivity. Investigations of radioactivity in wildlife from the Thule area were not included in the present project.

A stream runs through the area and flows into Bylot Sound at Narsaarsuk. The water flow of the stream is characterised by strong seasonal fluctuations from powerful runoff during snow melt in spring/summer to little or no flow during winter. The drainage area of the stream is more than 50 km<sup>2</sup>. Radioactive particles in the area have probably been transported by runoff to the stream and further to the sea and over the years accumulated in the sediments at the mouth of the stream. It is thus likely that significant amounts of radioactivity are present in these sediments. In order to account for all radioactive pollution in the area, and ascertain that no significant environmental impact is overlooked, it is of interest to investigate these sediments. That data together with data on possible additional sub-areas with high levels of radioactivity is relevant in relation to the amounts of plutonium from the nuclear weapons dispersed at the accident in 1968 (Christensen, 2009).

## 7 Conclusions

A survey of radioactive pollution from the airplane accident in 1968 on land in the Thule area was carried out during 2007 and 2008.

The survey in the Narsaarsuk area includes observations from more than 400 sites. The survey has included a mapping of locations with particularly high levels of radioactive pollution south and east of the hunting cabins at Narsaarsuk. The results show levels of radioactive pollution in soil ranging from background values around  $13 \text{ Bq m}^{-2}$  of  $^{241}\text{Am}$  and  $39 \text{ Bq m}^{-2}$  of  $^{239+240}\text{Pu}$  up to levels of  $0.3 \text{ MBq m}^{-2}$  of  $^{241}\text{Am}$  and  $1.7 \text{ MBq m}^{-2}$  of  $^{239+240}\text{Pu}$ . A number of local sub-areas (about ten) of sizes ranging from a few hundred to a few thousand square metres show elevated levels above  $10 \text{ kBq m}^{-2}$  of  $^{239+240}\text{Pu}$ . The occurrence of sub-areas with significantly elevated levels of plutonium in soil is probably due to redistribution and accumulation in snow of the dispersed radioactive particles.

Depth distributions of plutonium in soil were determined from one of the sub-areas with elevated concentrations. Maximum concentrations were generally found in the upper 0-2 cm containing up to 90% of the total amount.

Based on geostatistical analysis of data, the total amount of plutonium in soil at Narsaarsuk is estimated at 270 GBq (100 g) with a 95% confidence interval of from 140 to 490 GBq (54 to 190 g).

At Narsaarsuk investigations were carried out to determine the occurrence of radioactive particles in air. This involved collection of airborne particles with an air sampler, passive collection of airborne particles on sticky foils, collection of rain samples and collection of particles that could be resuspended by wind from the soil surface to the air. Small amounts of plutonium were found in air and rain samples, but the derived concentrations in air were very low corresponding to typical background levels in Europe in the range  $1\text{-}10 \text{ nBq m}^{-3}$  of  $^{239+240}\text{Pu}$ . A few small particles were found on the soil surface with activities up to 1000 Bq plutonium but the air and rain samples showed no sign of resuspension of such particles from the soil.

Local areas with elevated levels of radioactive pollution in soil were found at Kap Atholl and Grønnedal 15-20 km south of Narsaarsuk. Here the levels were lower than those in the Narsaarsuk area but clearly above background.

It was not possible within this project to carry out a systematic survey of the entire region and it can thus not be excluded that there may be more local sub-areas than those identified with elevated levels of radioactive pollution in soil.

Screening surveys were carried out at Thule Air Base, Moriusaq, Saunders Island and Wolstenholme Island. The results showed background levels in soil at these locations which means that no sign of radioactive pollution from the accident was found at these locations.

## 8 Recommendations

A relatively large number of sub-areas in the Narsaarsuk area was identified with significantly elevated levels of plutonium in soil. Further investigations would be needed in order to clarify if there are other sub-areas in the region with significantly

elevated levels of radioactivity from the accident. Due to the size of the region and the nature of the terrain an airborne survey should be carried out to provide such information.

Wild animals living in the region might become contaminated with radioactivity particularly from the sub-areas at Narsaarsuk with elevated levels. In order to determine the extent to which consumption of terrestrial wild life from the area represents a radiological risk to humans, a survey of radioactivity in wild life from the region should be carried out. The survey should include information on wild life products that are used for human consumption in the area, collection of samples from such wild life and analysis of radioactivity in the samples. Wild life of relevance might include musk ox, arctic hare and birds.

The sub-areas with elevated levels of radioactivity in the Narsaarsuk area indicate that significant redistribution of pollution from the accident in 1968 has occurred. It is likely that significant amounts of radioactive particles have been transported by water runoff to the local stream and carried to the outflow of the stream at Narsaarsuk into the sea and accumulated at the mouth in the sediments. These sediments should be investigated for content of possible high levels of radioactivity from the accident.

## **9 Acknowledgements**

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## 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., 1979. Environmental Studies on Radioecological Sensitivity and Variability with Special Emphasis on the Fallout Nuclides  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ , Risø-R-437, Risø National Laboratory, Roskilde, Denmark.
- 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  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{237}\text{Np}$ ,  $^{234,238}\text{U}$ ,  $^{228,230,232}\text{Th}$ ,  $^{99}\text{Tc}$  and  $^{210}\text{Pb}$ - $^{210}\text{Po}$  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  $\text{Ca}(\text{OH})_2$  and  $\text{Ba}(\text{Ra})\text{Cl}_2$  or  $\text{Ba}(\text{Ra})\text{SO}_4$  for the determination of radiostrontium. *Anal. Chim. Acta* **466**, 109-116
- Christensen, S.A., 2009. The Marshal's Baton. DIIS Report 2009:19. Danish Institute for International Studies, Copenhagen, Denmark.
- 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  $\alpha$ -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.

- EC, 2006. NUSIMEP 5: Uranium, plutonium and caesium isotopic abundance in a saline matrix, EUR 22286 EN, IRMM, European Commission, Belgium
- 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.
- Hölgge, Z., 2008. Plutonium isotopes in surface air of Prague in 1986-2006. J. Env. Rad. 99(10):1653.
- 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  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{241}\text{Am}$  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
- IAEA, 2007a. The IAEA-CU-2006-04 Proficiency Test on the Determination of Gamma Emitting Radionuclides, IAEA, Seibersdorf, Austria
- IAEA, 2007b. Report on the worldwide intercomparison exercise IAEA-437. Radionuclides in Mediterranean Mussels. IAEA/AL/173 IAEA/MEL/79. International Atomic Energy Agency, Vienna, Austria
- IAEA, 2009a. ALMERA Proficiency Test on Determination of Po-210 in Water, IAEA-CU-2007-09, IAEA, Vienna, Austria
- IAEA, 2009b. Interlaboratory Comparison Radionuclides in Irish Sea Water IAEA-443, IAEA/AQ/10, IAEA, Vienna, Austria
- IAEA, 2009c. ALMERA Proficiency Test on the Determination of Radionuclides in Spinach, Soil and Water, IAEA/AQ/3, IAEA, Vienna, Austria
- IAEA, 2010a. ALMERA Proficiency test Determination of Naturally Occurring Radionuclides in Phosphogypsum and Water, IAEA-CU-2008-04, IAEA, Vienna, Austria

- IAEA, 2010b. HELCOM-MORS Proficiency Test Determination of Radionuclides in Fish Flesh Sample. IAEA/AQ/13. IAEA, Vienna, Austria
- JSA-EnviroStat, 2011. Spatial statistical analysis of contamination level of  $^{241}\text{Am}$  and  $^{239}\text{Pu}$  at Thule, North-West Greenland. Gilleleje, Denmark.
- 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.
- McNeary, D., Baskaran, M., 2003. Depositional characteristics of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in southeastern Michigan. *Journal of Geophysical Research*, 108(D7), 4210.
- Michel, H., Barci-Funel, G., Dalmaso, J., Ardisson, G., Appelby, P.G., Haworth, E. and El-Daoushy, F., 2002. Plutonium and americium inventories in atmospheric fall-out and sediment cores from Blelham tarn, Cumbria (UK). *J. Environ. Radioact.* 59(2):127-137.
- Michel, H., Barci-Funel, G., Dalmaso, J., Ardisson, G., Appelby, P.G., Haworth, E. and El-Daoushy, F., 2002. Plutonium and americium inventories in atmospheric fall-out 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
- Nielsen, S.P. and Roos, P., 2006. Thule-2003 – Investigation of Radioactive Contamination, Risø-R-1549(EN), Risø National Laboratory, Roskilde, Denmark.
- NIRP, 2011. Thule-ulykken, Vurdering af stråledoser fra radioaktiv forurening af landjorden. National Institute of Radiation Protection, Herlev, Denmark.
- NPL, 2006. Environmental Radioactivity Comparison Exercise 2005. National Physics Laboratory, United Kingdom
- NPL, 2008. Environmental Radioactivity Proficiency Test Exercise 2007. National Physics Laboratory, United Kingdom
- NUSTL, 2011. EML Environmental Databases. National Urban Security Technology Laboratory, US Department of Homeland Security, USA. (<http://www.eml.st.dhs.gov/databases/>)
- Outola, I., Filliben, J. and Inn, K.G.W., 2008. Searching a robust reference value from intercomparison exercise data: Seaweed radionuclide Standard Reference Material (SRM4359), *J. Radioanal. Nucl. Chem.*, 276, 329-334.
- 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.
- SIS, 2011. Thule-ulykken, Vurdering af stråledoser fra radioaktiv forurening af landjorden. Statens Institut for Strålebeskyttelse, Sundhedsstyrelsen, Herlev (In Danish)
- 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.
- UNSCEAR, 1993. *Sources and Effects of Ionizing Radiation*. United Nations Scientific Committee on the Effects of Atomic Radiation, 1993 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.

Wershofen, H. and Arnold, D., 1999. Radionuclides in Ground-Level Air in Braunschweig – Report of the PTB Trace Survey Station for 1995, 1996 and 1997. PTB-Ra-41, Braunschweig.

## Appendix A, Meteorological data

Meteorological data recorded at Narsaarsuk during summer 2007 and 2008.

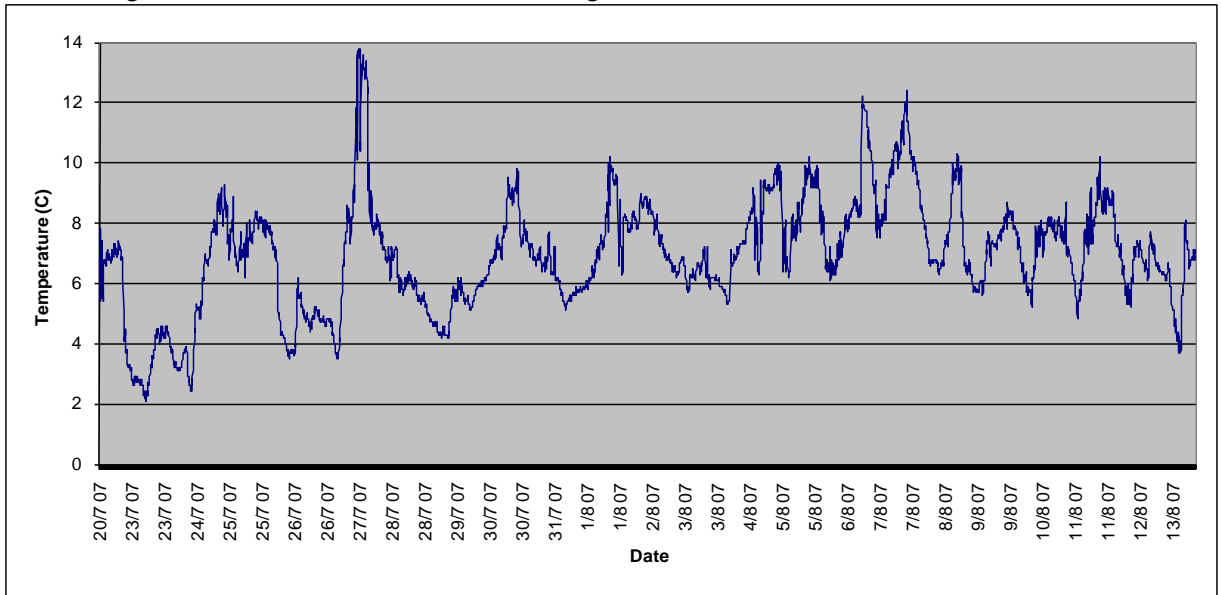


Figure 51. Temperature during field campaign at Narsaarsuk in 2007.

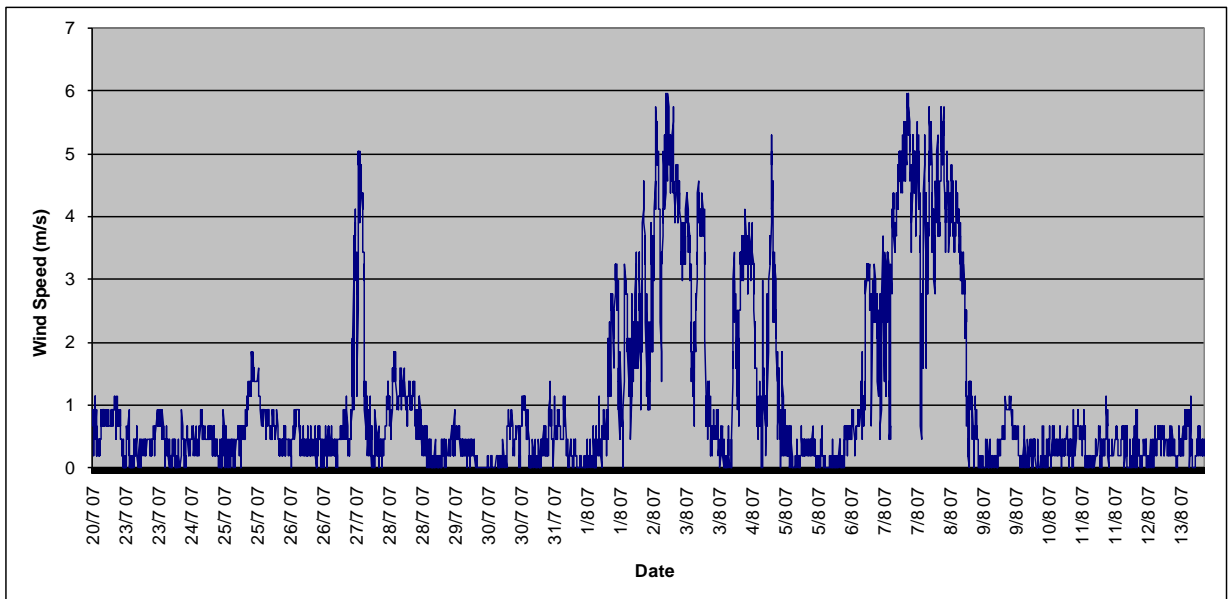


Figure 52. Wind speed during field campaign at Narsaarsuk in 2007.

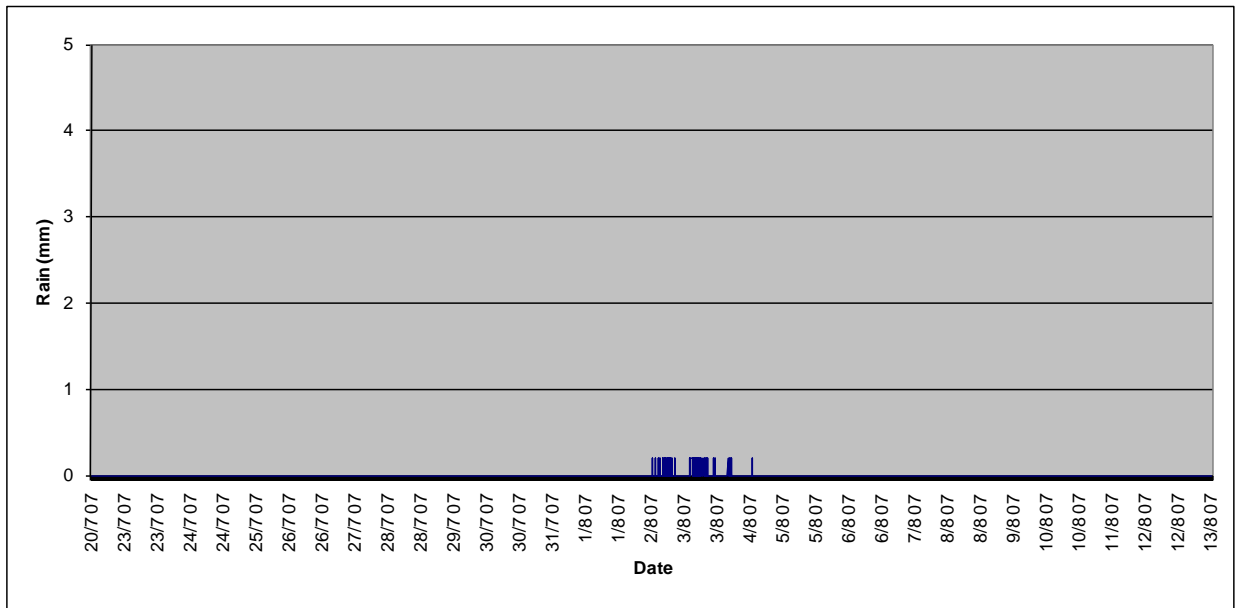


Figure 53. Precipitation during field campaign at Narsaarsuk in 2007.

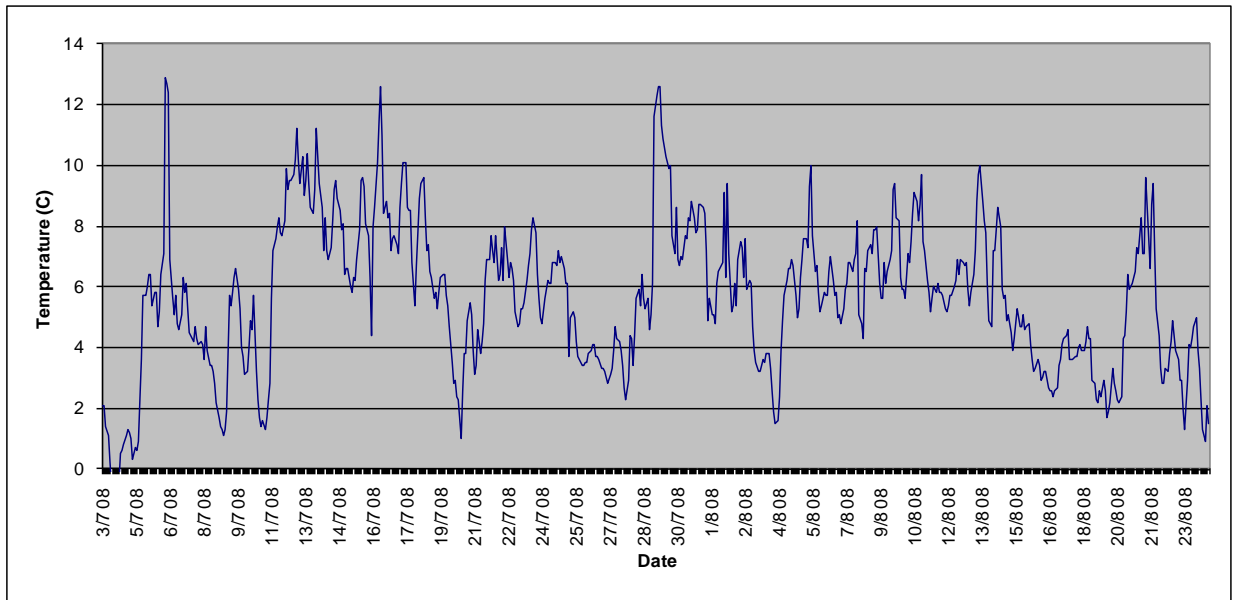


Figure 54. Temperature during field campaign at Narsaarsuk in 2008.

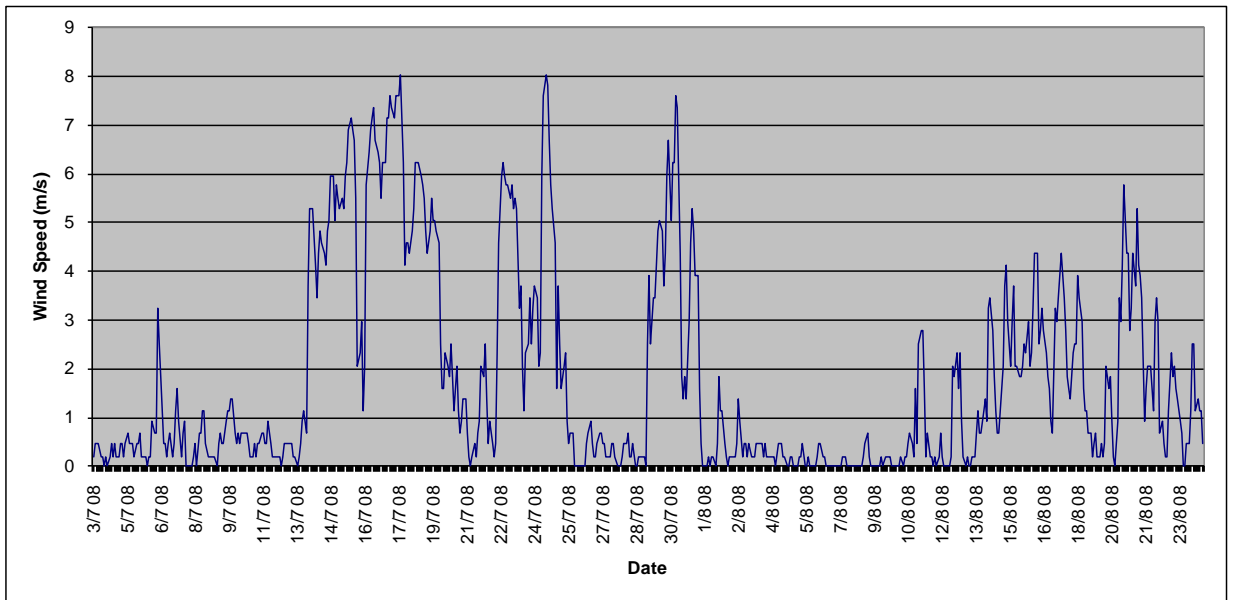


Figure 55 Wind speed during field campaign at Narsaarsuk in 2008.

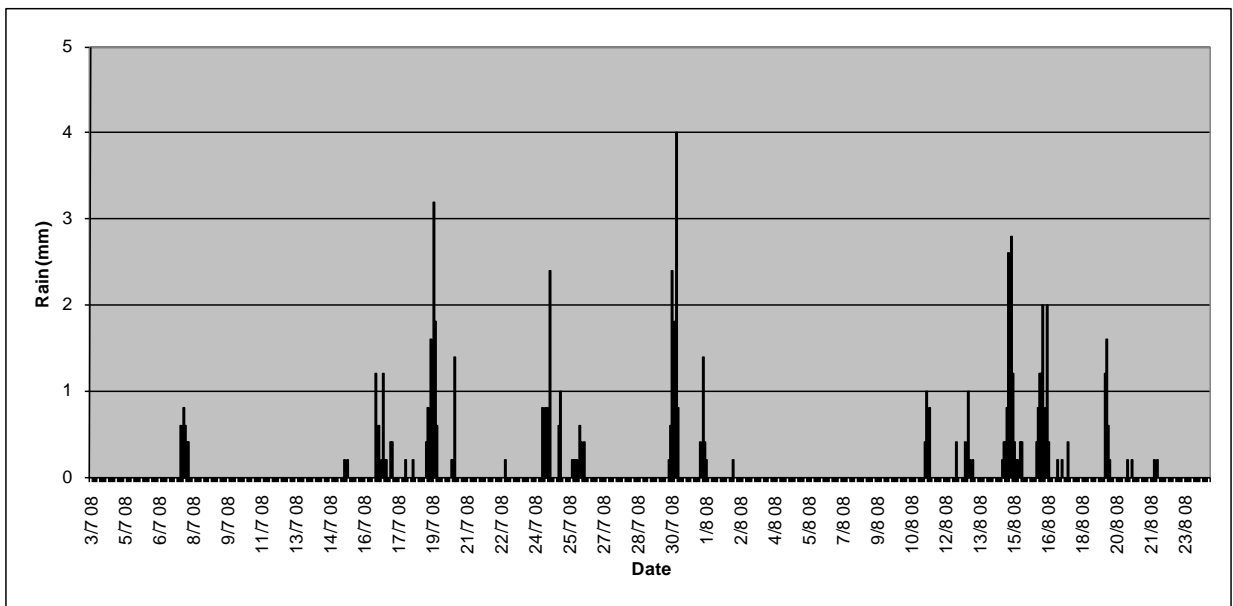


Figure 56. Precipitation during field campaign at Narsaarsuk in 2008.

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