

Assessment of risk from plutonium isotopes in connection with the proposed onshore mining activities at Moriusaq, Thule, Northwest Greenland.

The Thule accident

In January 1968 an American B52 bomber carrying four thermonuclear weapons caught fire during a mission in northwest Greenland. The pilot tried reaching Thule airport but the airplane crashed on the Bylot Sound ice, approximately 15km west of the airbase, following bailout of the crew. At impact the chemical explosives in the weapons detonated and dispersed the weapons content of plutonium into the surrounding environment. The smoke pillar carried dust and debris to several hundred meters where the prevailing winds were towards south. Apart from a heavily contaminated area on the Bylot Sound ice finer dust were initially carried over land to the south. During the following days severe snowstorms redistributed some of this material in a more irregular pattern. A thorough clean-up operation was undertaken in the next months to remove the debris and contaminated ice around the crash site. Several radioecological investigations have been conducted in the area over the years, mainly focusing on the marine life due to the inuite food habits. The distribution of plutonium in sediments in Bylot Sound has been mapped during several occasions over the years. While the normal background deposition of plutonium over the world is in the order of 10-50 Bq per square meter the corresponding area concentrations in Bylot Sound sediments range from background levels to 10^6 Bq m⁻². An example of the results from the last survey (2003) is shown in the figure below. Due to the low solubility of plutonium, concentrations in the water are only slightly higher than background values. This is also due to the specific nature of the contamination being in the form of very fine particles (sub micrometer to several tens of micrometers).

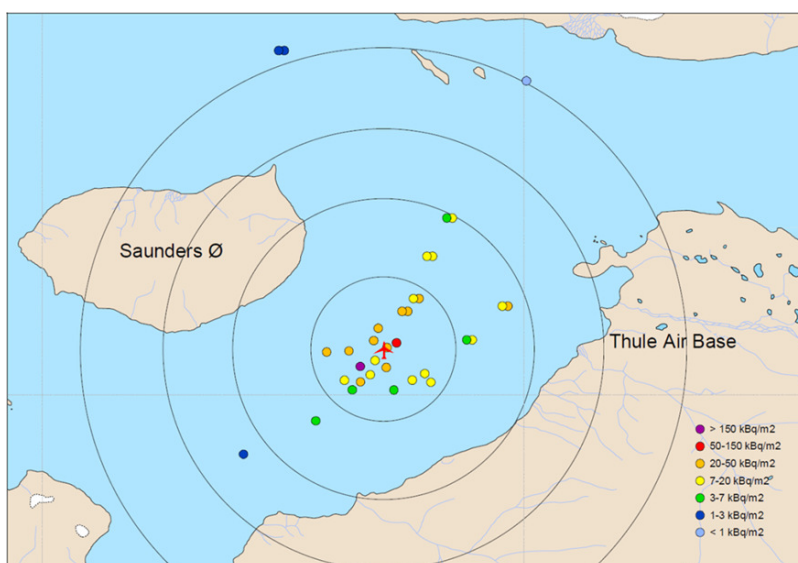


Figure 1

Last survey for Pu-isotopes in the marine environment around the Thule crash site conducted in 2003. From Ref [1].

Focus on the terrestrial contamination started in 2003 and mapping of the area south of the crash site was conducted in 2006-2008. During this time also Thule Air Base and some other more remote locations were briefly scanned (fig 2). The selection of these sites was based on the combined probability of having contaminated ground and the density of people. The primary reason for including Moriusaq was thus to relax the worries of remaining residents and the fact that a substantial part of the Moriusaq residents with sledge dogs and other equipment played a central role in the early clean-up of the crash site in 1968. Although the Moriusaq area may not have been directly exposed to any debris from the crash, potentially contaminated material could have reached the village during the clean-up work. The results of the screening work conducted in Moriusaq during 2008 are shown in figure 3 together with results from the other sites screened. The data in figure 3 are given as concentrations of ^{241}Am , a daughter isotope to one of the Pu-isotopes. To convert to Pu the data should be multiplied with a factor of 6. The ^{241}Am enables a faster screening of contaminated areas but is much less sensitive than when specific Pu-analysis is performed. Later Pu-analysis of selected soil samples collected in Moriusaq during 2008 confirmed a Pu-isotopic signature originating from the B-52 weapons but the total concentrations found were of no concern to health. It must be kept in mind however that the Thule contamination pattern is extremely variable and is associated with small particles.

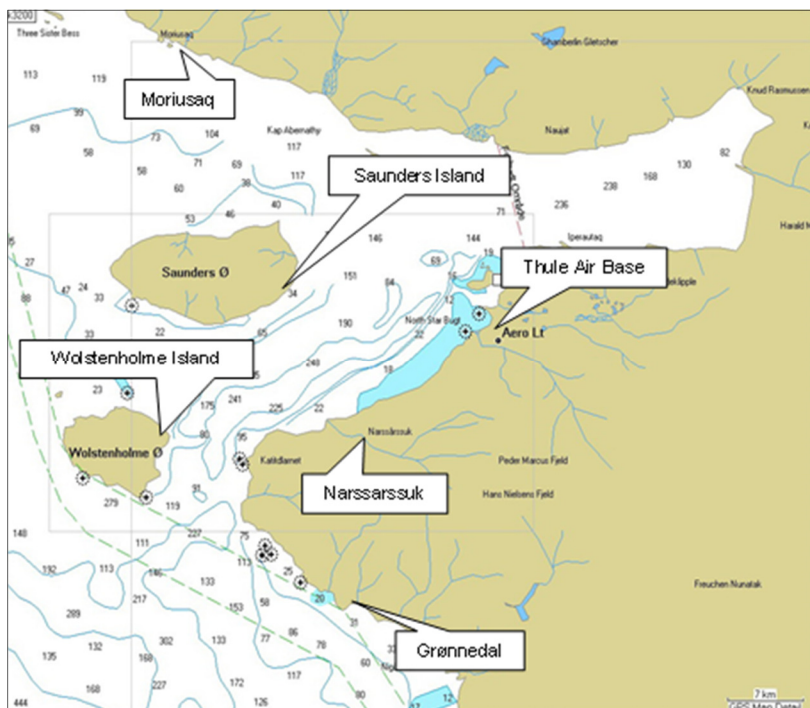


Figure 2.
Areas investigated in the 2006-2008. From ref [2].

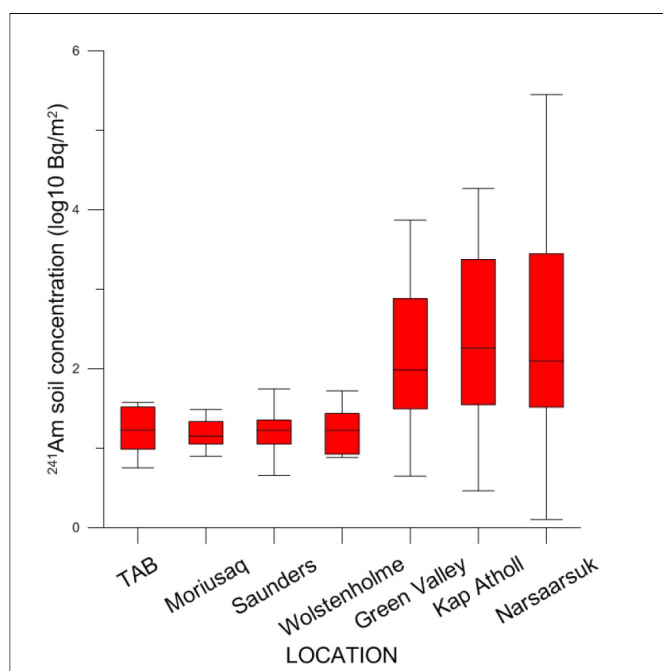


Figure 3

Summary of results from the analysis of soil samples for ^{241}Am in 2006-2008. The activity ratio $\text{Pu}/^{241}\text{Am}$ is approximately 6. From ref [2].

Relevant contamination pathways

The primary radioisotopes of concern at Thule are ^{239}Pu , ^{240}Pu and ^{241}Am . These all decay through alpha emission. Gamma emission is of very low intensity (except for ^{241}Am) and with low photon energies. The radiological risk from external irradiation is therefore negligible. All risks are related to transfer of contaminated material inside the body through inhalation, open wounds and to a lesser extent ingestion. Given the nature of the present project the main route of exposure would be through inhalation of resuspended soil and secondly through transfer to blood from soil contaminated wounds. The typical size of Pu particles found in the Thule area is in the 1-5 μm range. Although not known for sure these particles are likely attached to larger pieces of soil material so a significant reduction in inhalation dose could be obtained with masks having retention size much higher. Larger particles, 20-100 μm , are known to appear as single objects.

Analysis of soil samples in the Moriusaq area

Dundas Titanium A/S proposes to develop a project, which will extract ilmenite concentrate with high titanium oxide content from the black heavy mineral sand deposits found along the coastline of Steensby Land around Moriusaq (Figure 4). A surface miner will be used to excavate the mineral sands. The risk assessment in this study are based on previous data from 2008 and the result of analyses of plutonium in soil samples collected in 2016, 2017 and 2018 from four sites within the planned project area plus a location on Saunders Island (Figure 4). The results of the 15 samples are shown in Table 1.

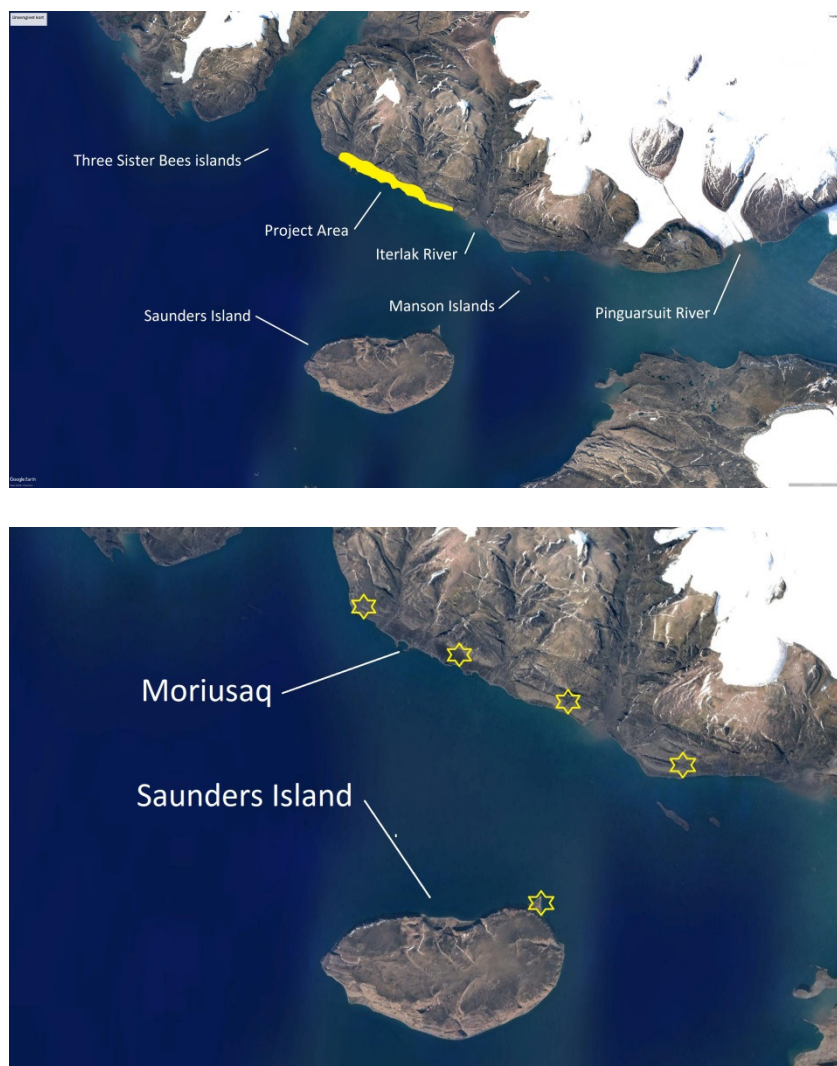


Figure 4
Map of the project area and the 5 locations where soil sampling was conducted in 2016, 2017 and 2018.

The soil samples were analyzed by gamma spectrometry as well as ICP-MS specifically for Pu-isotopes. The samples were freeze-dried and a mass of 200-400g were packed in plastic containers and analyzed on large (50-60% relative efficiency) HPGe-detectors placed in low-background lead shields. About 2g of the material was dissolved and Pu isolated using radiochemical methods and analyzed using a Triple Quad Agilent-8800 ICP-MS.

DTU-nr	Sample ID	Sample date	Location	Station	Latitude	Longitude	²⁴¹ Am [Bq/kg]	¹³⁷ Cs [Bq/kg]	Pu- ²³⁹ + ²⁴⁰ [Bq/kg]
8002	59406	42949		EBS4	76.67N	69.13W	<1.7	1.9 +/- 0.2	0.11
8003	59410	42950		EBS5	76.60N	69.50W	<1.2	3.3 +/- 0.2	0.10
8004	59425	42952		EBS1	79.79N	69.98W	<1.3	0.4 +/- 0.09	0.04
8005	59427	42953		EBS3	76.71N	69.46W	<0.5	1.8 +/- 0.1	0.10
8006	59440	42956		EBS2	76.73N	69.68W	<1.1	33 +/- 2	1.63
8007	59601	43335		EBS001	76.7614N	69.9653W	<0.7	2 +/- 0.2	0.12
8008	59602	43335		EBS002	76.7334N	69.6892W	<0.4	7 +/- 0.3	0.19
8009	59626	43336		EBS003	76.7109N	69.4571W	<0.4	15 +/- 0.6	0.66
8010	59634	43337		M5	76.6008N	69.50W	<1.5	5.1 +/- 0.3	0.28
8011	59645	43338		EBS004	76.673N	69.1571W	<1.6	11.3 +/- 0.7	0.20
8012	59703	42596	Moriusaq	M1	76.7614N	69.9253W	<0.7	4.6 +/- 0.2	0.16
8013	59717	42597	Moriusaq	M2	76.7334N	69.6892W	<0.5	0.3 +/- 0.06	0.04
8014	59723	42598	Moriusaq	M3	76.7109N	69.4571W	<0.4	0.9 +/- 0.07	0.12
8015	59729	42598	Moriusaq	M4	76.673N	69.1571W	<1.5	11 +/- 0.6	0.55
8016	59746	42601	Sanders Island	M5	76N 36.033	69W 03.138'	<1.5	7.6 +/- 0.4	0.34

Table 1
 Results of analyzed soil samples. Plutonium concentrations are presented as the sum of the two main isotopes ²³⁹Pu and ²⁴⁰Pu. Uncertainty for the Pu-analysis is around 20%.

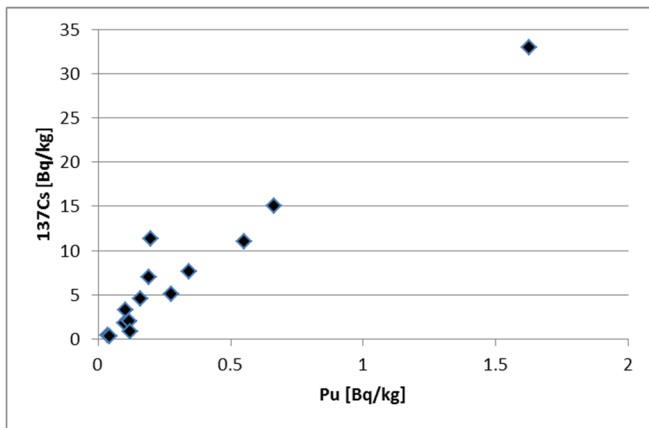


Figure 4
 Graph showing the correlation between ¹³⁷Cs and Pu in the analyzed soil samples.

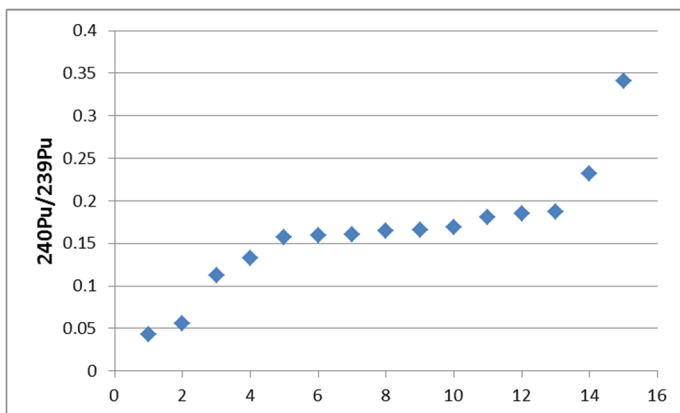


Figure 5
 Graph showing the ²⁴⁰Pu/²³⁹Pu atom ratios in the analyzed soil samples.

As already mentioned above, analyses of soil samples collected at the village Moriusaq in 2008 showed concentrations of plutonium that were of no concern to the environment or human health. The new data presented here clearly shows that the impact from the Thule accident is insignificant. Pu-concentrations in the analyzed soil samples are all low (a factor 10^3 - 10^6 lower than at the contaminated areas south of the crash site) and similar to what is found on other areas in the north hemisphere only exposed to nuclear weapons test fallout. The variability in the Pu-concentrations in the samples is best explained by the different retention of the soils. This is best seen in the correlation with ^{137}Cs , an isotope not originating from the Thule weapons crash but from nuclear weapons test fallout. The Pu/ ^{137}Cs ratio in this fallout found in soil samples today is around 2-4% but depends on the retention properties of the soils. The ratios found in the soil samples analyzed here points to a Pu-origin which mainly is from the weapons test fallout. Further indications for this come from the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios obtained from the ICP-MS analysis. The Thule Pu-signature is a ratio of around 0.05 while from test fallout it is about 0.2. Although with large uncertainties for the ^{240}Pu results the ratios as seen in figure 5 again indicate mainly Pu with a weapons test fallout signature.

Dose calculations

Concentrations of plutonium in the analyzed samples do not motivate any specific dose calculations from plutonium exposure to be done. Inhalation doses would be the most likely way of exposure but natural radioactivity (U and Th-series radioisotopes) are an order of magnitude more abundant and would provide the dominating inhalation dose from a given dust load. Based on the results and on previous findings in the area there are no relevant needs to perform extensive monitoring during the proposed activities. However, due to the conclusions being based on relatively few samples it would be justified to include analysis of Pu-isotopes in a potentially coming surveillance program particularly for air monitoring.

References

[1] Nielsen, S.P. and Roos, P., 2006. Thule-2003 – Investigation of Radioactive Contamination. Risø-R-1549(EN). Forskningscenter Risø, Roskilde.

[2] Nielsen, S. P. and Roos, P. Thule-2007 - Investigation of radioactive pollution on land. Risø-R-1781, October-2011.