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# ENVIRONMENTAL BEHAVIOUR OF PLUTONIUM ACCIDENTALLY RELEASED AT THULE, GREENLAND

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**Abstract**—The environmental contamination resulting from the B-52 accident in 1968 at Thule was studied by scientific expeditions in 1968, 1970 and 1974. The contamination was mainly confined to the marine environment, where plutonium was preferentially located in the sediments and the benthic fauna. Plutonium concentrations down through the sediment layers decayed exponentially with a half-depth of 1–2 cm. The horizontal distribution of the plutonium may be described by an exponential expression: mCi $^{239,240}$Pu km$^{-2}$ = 460e$^{-0.28R}$ or by a power function: mCi $^{239,240}$Pu km$^{-2}$ = 370 $R^{-1.2}$, where $R$ is the distance in km from the point of impact. The inventory of $^{239,240}$Pu in the marine environment from the accident was estimated at 25–30 Ci. The amount of $^{238}$Pu was ~0.5 Ci. The bottom animals, such as worms and molluscs, showed a horizontal distribution of radioactivity similar to that of the sediments. From 1968 to 1970 the $^{239,240}$Pu concentrations in the biota decreased by an order of magnitude, since 1970 the decrease has been less evident. In 1970 and 1974 there were no indications of increased plutonium concentrations in surface seawater or in sea plants or zooplankton. Higher animals such as fish, seabirds and marine mammals have shown no tendency to increasing plutonium levels since the accident.

## INTRODUCTION

SINCE the beginning of the Nuclear Age several cases of environmental plutonium contamination have been reported. The most widespread and well-studied contamination is the global fallout from nuclear weapons testing which has dissipated (325 ± 36) kCi plutonium in the biosphere (Har73). However, more concentrated contamination has been that from the reprocessing plant at Windscale, which has discharged approx ~$10^4$ Ci $^{239,240}$Pu into the Irish Sea during the past 15 yr (He75). In comparison, the release from the B-52 accident at Thule Airbase, Greenland, in January 1968, when ~25 Ci were dissipated in the marine environment (Aa71a) is modest contamination. Although the Thule incident may thus seem trivial as regards the amount of plutonium involved, it is felt that the special circumstances such as the Arctic environment, the particulate nature of the plutonium, and the point source character of the release have made it worthwhile to monitor the environmental plutonium levels at Thule. The aims of the studies described herein have been to identify the area of distribution of the contamination and to elucidate a possible radioecological propagation of the plutonium.

The initial levels were established by the scientific expedition in 1968 (Aa71a); increased plutonium concentrations were measured as far as 15 km from the point of impact, and sediments and bottom animals in the impact zone showed plutonium levels 2–3 orders of magnitude above the Pu fallout background. The next scientific expedition took place in 1970. A summary of the results from that expedition (Aa71b) showed that the contamination had moved as far as 30 km from the point of impact; that the integrated plutonium level in the biomass of bottom animals had decreased by a factor of ~4; and that higher animals such as fish and seals did not show significantly higher levels than in 1968.

The third scientific expedition took place in August 1974; besides four Danish participants, Mr. David E. Robertson from Batelle Northwest Laboratories and Dr. Wayne C. Hanson from Los Alamos Scientific Laboratory were members of the party. This paper reports the results from the marine sampling aspects of the 1974 expedition and some of

the unpublished data from the 1970 expedition. To elucidate the time trend in the plutonium levels, a short summary of some of the 1968 data is included for comparative purposes.

## MATERIAL AND METHODS

The environmental characteristics of the Thule locality have been described earlier (Aa71a) and are not repeated here. As previously the marine sampling was carried out by the Greenlandic ship "Aglanta." In comparison with 1970 and 1968, we extended the sampling in 1974 further away from the point of impact, the most distant sampling location being at a distance of 45 km (Fig. 1).

The 1974 expedition emphasized the collection of bottom sediments and bottom dwelling animals because sampling in 1968 and 1970 had shown that the highest plutonium levels were encountered in those types of samples. Two kinds of sediment samplers were used: a surface sampler, the PK sampler; and a bottom corer, the HAPS sampler that was developed by the Marine Biological Laboratory at Elsinore (Ka73). The PK sampler was also used in the earlier expeditions; it scrapes the uppermost layer of the sea bottom to a depth of ~1 cm over an rectangular area of 0.1 $m^2$ (Aa70). Experience showed that the sample depth varied with the nature of the bottom sediments; in muddy material the sample depth exceeded 1 cm and was somewhat shallower on a sandy bottom. The HAPS is in principle a corer with a top valve and a core catcher supported by a frame; it yielded well-defined samples and increased the effective sampling depth. The coring tube of stainless steel has a sampling diameter of 135 mm corresponding to a sampling area of 143 $cm^2$. When the core catcher closes, it may carry off a little surface material which contaminates the bottom layer of the sample; to prevent this, the lowest 1–2 cm of the sample core was discarded. The remaining part of the core is pushed out of the sampling tube by a piston and divided into 3 cm thick sections. The HAPS corer operated satisfactorily in Thule sea depths of >200 m provided that the bottom material was not stony.

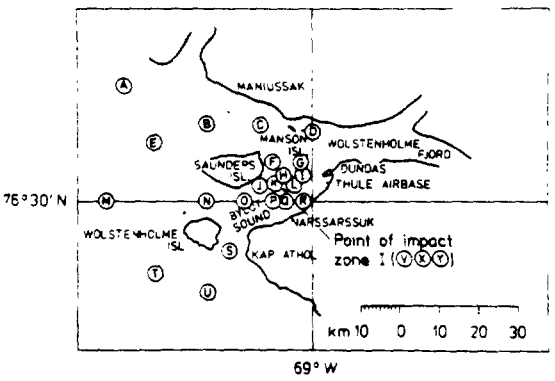


FIG. 1. The sample locations at Thule, Greenland (Location 19 is located in Bylot Sound at 76°27'8N, 69°37'5W).

Sediment samples were collected by both the PK-sampler and the HAPS corer at a number of locations. This comparison made it possible to empirically convert the surface data obtained by the PK-sampler to the total integrated level in the sediment column. The plutonium inventory in the uppermost cm of the sediments was estimated to contain one-third of the total plutonium content of the sediment layer.

As previously, the biological samples were kept frozen until processed in the laboratory. The radiochemical plutonium analysis followed the classical procedure (Ta71). The chemical yields were determined by $^{242}Pu$ or $^{236}Pu$ spikes supplied by Dr. John H. Harley, U.S.E.R.D.A. Health and Safety Laboratory, New York. The average yield of all sample types was 55%, and the minimum detectable activity for 4000 min counts was ~10 fCi of $^{239}Pu$. The analyses of the samples that contained plutonium fallout background levels, were usually performed on 5–10 g ash, while the samples collected near the point of impact were normally analyzed in 0.5–1 g aliquots. Samples that contained <3 fCi $^{239}Pu$ per g of ash were generally below the detection limits of the procedures applied in this study.

To avoid cross-contamination, both sample collecting and laboratory processing were carried out on samples taken far from the point of impact before proceeding to those taken close to.

Due to the particulate nature of the Thule

contaminations the plutonium was very inhomogeneously distributed in the samples especially the sediment. Those samples were therefore analyzed as duplicates and in some cases even as triplicates.

The analytical errors, including counting error, were usually 10–20%, depending on the activity content. Samples with a counting error of more than 33% were denoted by ~0.

## RESULTS AND DISCUSSION

### *Seawater*

Only six 50-l. samples of unfiltered water were analyzed from the 1970 and 1974 samplings (Table 1) because 1968 plutonium levels in the Thule seawater was essentially that of the fallout background (~1 fCi $^{239,240}$Pu l.$^{-1}$), apart from a few samples that probably contained particulates stirred up from the bottom sediments. In 1970, six samples collected off the west Greenland Coast from Godthåb (~64°N) to Thule (~76°N) showed a mean level of 1.6 ± 0.3 (S.E.) fCi $^{239,240}$Pu l.$^{-1}$. Thus, neither in 1970 nor in 1974 did the Thule water contain levels significantly above fallout background, apart from the bottom sample from location G, sampled in 1974. Analysis of large-volume seawater samples collected in 1974 (Robertson, pers. comm.) showed that the surface water at Thule was typical of fallout background levels, whereas the near-bottom waters showed elevated plutonium levels of particulate nature due to resuspension of bottom sediments. Our samples of sea plants and zooplankton living in the surface water confirmed the fallout background concentrations of dissolved plutonium in the Thule water.

The low $^{239,240}$Pu concentrations in Thule seawater were ascribed to the fact that the contamination at Thule consisted of highly insoluble plutonium oxide. Even in cases of releases of soluble plutonium to the aquatic environment, it has been general experience that the bulk of the plutonium is found in the sediments and not in the water masses. The Windscale studies (He75) have thus shown that only a small percentage of the total discharge of plutonium remains in the water, and similar results were obtained from studies in Lake Michigan of plutonium from fallout (Wa75), where ~96% of the fallout plutonium was transported to the sediment.

To what extent the plutonium in the Thule sediments dissolves has not yet been clarified, but leaching experiments and studies of interstitial water in the sediment cores (Robertson, pers. comm.) indicate some degree of dissolution.

*Table 1. Plutonium in seawater from Bylot Sound collected in 1970 and 1974*

| Sample year | Location (Fig. 1) | Sample depth in m | fCi $^{239,240}$Pu l$^{-1}$ |
|---|---|---|---|
| 1970 | C | 112 | 0.6 |
| 1970 | C | Surface | 3 |
| 1970 | V | Surface | 2 |
| 1974 | G | 190 | 50 |
| 1974 | P | 240 | 1.5 |
| 1974 | V | Surface | 1.0 |

Detection limit 0.3 fCi l$^{-1}$

### *Sea-bottom sediments*

All but one of the sediment samples collected in 1968 and 1970, were surface sediments taken by the 0.1 m$^2$ PK-sampler. The exception was a 1970 core sample collected at the point of impact down to a depth of 10 cm by a Burke open-tube gravity corer, kindly placed at our disposal by Dr. V. T. Bowen of the Woods Hole Oceanographic Institute. This sample was divided into 3 sections of equal thickness. The analysis showed that ~$\frac{1}{3}$ of the total plutonium activity of the core were contained in the upper 3–4 cm. Assuming an exponential distribution of the plutonium concentration in the core, we estimated that the uppermost cm of the sediments contained ~$\frac{1}{4}$ of the total activity. Hence we could transform the results of the PK-samplings to total integrated plutonium in the sediment layers, under the assumption that the vertical distribution of Pu in all sediment samples collected both in 1968 and 1970 followed the pattern observed in the core sample collected at the point of impact in 1970. It was, of course, not satisfactory to base such sweeping conclusions on a single sample, and it was important to obtain a more comprehensive collection of core samples. This was made possible in 1974 by the HAPS sampler.

As shown in Fig. 2, the plutonium concentration in the sediment cores decreased exponentially with increasing sampling depth. The

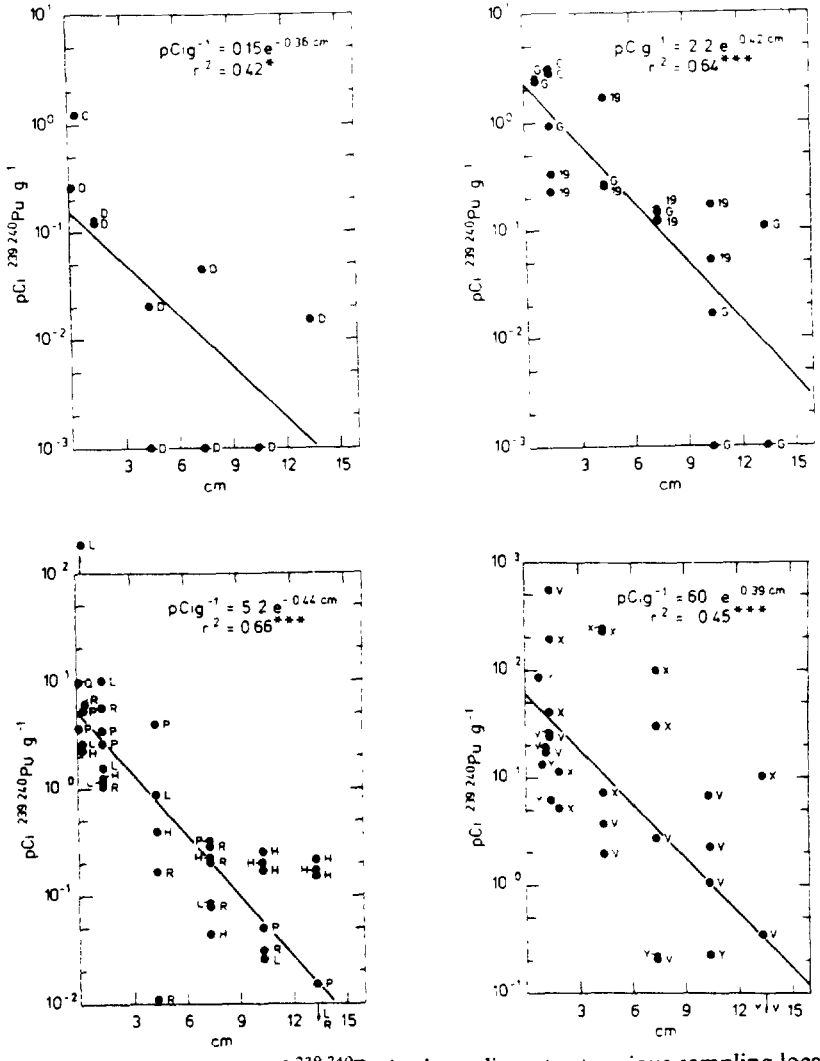


FIG. 2. The vertical distribution of $^{239,240}$Pu in the sediments at various sampling locations at Thule. The abscissae are the depth in cm of the sediments; $r^2$ is the correlation coefficient between observed data and values calculated from the exponential equation (Significance levels: *: $P \leq 0.05$, **: $P \leq 0.01$, ***: $P \leq 0.001$).

four groups of sample locations, arranged according to decreasing distance from the point of impact, showed similar vertical distributions. Location H results were considered to be anomalously high, probably due to cross-contamination during sampling. The half-depth for the plutonium varied between 1.6 and 1.9 cm. The plutonium concentration $C_{RX}$ in the sediments $R$ km from the point of impact and in the sample depth $x$ cm may be described by:

$$C_{RX} = K_R e^{-0.40x} \text{ pCi g}^{-1}, \quad (1)$$

where

$$K_R = 40e^{-0.33R} \text{ pCi g}^{-1}, \quad (2)$$

as shown in Fig. 3.

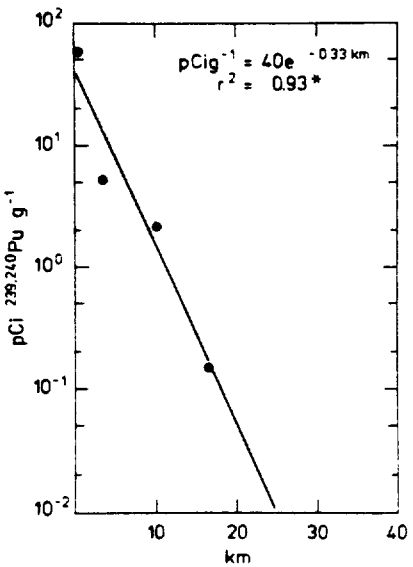


FIG. 3. The $^{239,240}$Pu concentration in the sediment surface (0 cm depth) related to the distance (in km) from the point of impact.

The Windscale studies (He75) have also demonstrated an exponential activity decrease down through the sediments. The half-depths in the Irish Sea were, however, considerably greater (4.6–8.6 cm) than in the present study, presumably because of the continuous nature of the releases from Windscale. In the studies of fallout plutonium in Lake Michigan (Ed75), plutonium was detectable only down to a depth of 6 cm, and there was no evident exponential decrease in plutonium concentrations down through the sediments; this was partly due to redistribution of previously deposited plutonium and partly to the dependence on the fallout rate.

The vertical distribution of the plutonium in sediments has received great attention and several explainations have been put forward. It has been proposed that the presence of plutonium in the deeper layers was due either to molecular diffusion and sorption/desorption taking place in the bulk sediment, or to biological reworking of the deposits, or to a combination of these processes (No72). The nearly constant relations between two so chemically different elements as Cs and Pu, which have been observed by several authors (No72; He75; Ed75), however, suggest, that physico-chemical mechanisms are less important.

In the Thule environment the strong tidal currents at the bottom stir up the sediments, as demonstrated by the particulate nature of plutonium found in the bottom water (Robertson, pers. comm.). This may result in both a horizontal and vertical dispersal of plutonium. Furthermore, we know that the worms contain plutonium in similar concentrations (per g of ash) as the upper sediment layers. Therefore we assume that these worms and other bottom animals may displace plutonium to greater depths in the sediments through their biological activities. Bowen (Bo75) suggested such a biological transport by fastmoving worms such as *Arenicola* and *Nereis*.

The inventory of plutonium in the Thule sediments may be estimated from equations (1) and (2). From the HAPS samples we calculated the average density in the upper 15 cm of the sediments at the 10 stations shown in Table 2 to be $(7.55 \pm 0.46) \cdot 10^9$ g cm$^{-1}$ km$^{-2}$. There is no land area within 7.5 km from the point of impact and beyond 7.5 km land constitutes ~60% of the surface; thus, the inventory is calculated from integration of equations (1) and (2) to be: 25 Ci.

We may estimate the inventory more directly from the mCi km$^{-2}$ data in Table 2, where the horizontal distribution of the plutonium deposit may be described by:

$$\text{mCi } ^{239,240}\text{Pu km}^{-2} = 460e^{-0.276R} \qquad (3)$$

where $R$ is the distance in km from the point of impact. The correlation coefficient ($r^2$) between observed and calculated values was 0.60 and significant ($P \leq 0.01$). Analogous to the above calculation, we get: 29 Ci. The latter estimate is slightly greater because the density increases down through the sediments. which means that the deposition of plutonium (mCi km$^{-2}$) decreases more slowly with depth than the plutonium concentration (pCi g$^{-1}$).

Instead of the exponential expression (3) we may obtain a better description of the situation by a power model:

$$\text{mCi } ^{239,240}\text{Pu km}^{-2} = 370\, R^{-1.21}. \qquad (4)$$

*Table 2 The vertical and horizontal distribution of plutonium at Thule in 1974. All values are in mC. $^{239,240}$Pu km$^{-2}$ in sediment sample layers*

| Location (Fig. 1) | Distance from point of impact (km) | HAPS sample section 0–3 cm | 3–6 cm | 6–9 cm | 9–12 cm | 12–15 cm | PK samples Sample depth in cm | PK samples Inventory down to sample depth | Integrated activity in total sediment layer calculated from HAPS and PK |
|---|---|---|---|---|---|---|---|---|---|
| C | 17 | — | — | — | — | | 0.8 | 4.8 | 16 |
| D | 16 | 3.0<br>3.1 | ~0<br>0.5 | 1.2<br>~0 | ~0 | 0.4 | 0.2 | 0.4 | 2.5 |
| G | 9 | 37<br>11<br>34 | 5.4 | 3.0 | 0.4<br>~0 | 2.4<br>~0 | 1.2 | 11.6<br>12.1 | 30 |
| H | 3 | 25 | 11 | 1.5<br>7.7 | 7.2<br>4.9<br>5.6 | 5.3<br>6.5<br>4.9 | 0.6 | 9.7<br>165 | 97 |
| K | 1.4 | — | — | — | — | — | 1.1 | 39 | 118 |
| L | 4 | 18<br>156<br>26 | 16 | 1.6 | 0.5 | ~0 | 0.6 | 7.4<br>440 | 106 |
| P | 4 | 51 | 78 | 6.1 | 1.2 | 0.3 | 0.7 | 13<br>18 | 94 |
| Q | 3 | — | — | — | — | — | 0.3 | 4.8<br>13 | 44 |
| R | 4 | 107<br>49<br>20 | 4.0<br>0.2<br>16 | 2.0<br>4.9<br>8.7 | 0.8 | ~0 | 1.0 | 37 | 61 |
| 19 | 11 | 8.3<br>10.8 | 46<br>7.4 | 4.0<br>4.7 | 1.6<br>5.4 | — | — | — | 37 |
| V | 0 | 5800<br>260<br>270 | 75<br>39 | 4.3<br>57 | 25<br>54 | 8.5<br>1.4 | 2.4 | 165<br>153 | 2300 |
| X | 0.8 | 2600<br>540 | 4500<br>4400 | 2200<br>640 | 170 | 300 | 4.3 | 96<br>47 | 7900 |
| Y | 0.8 | 77 | 146 | 5.1 | 5.4 | 0.4 | 1.8 | 100<br>640 | 2300 |

Comments ~0 means below detection limit (<0.1 mCi km$^{-2}$)

The correlation coefficient ($r^2$) between observed and calculated values was 0.73 and highly significant ($P \leq 0.001$). The integrated plutonium level out to a distance of 45 km, where we measured levels not significantly different from the fallout background, becomes: 32 Ci. Power functions have also been used in inventory estimates by other investigators, such as around the Savannah River reprocessing plant (Mc75).

A comparison of the present estimates of the plutonium inventory in Thule with the previous ones of 17 Ci in 1968 and 24 Ci in 1970 (Aa71b) seems fairly satisfactory when we consider the great uncertainties of the earlier estimates because they were almost exclusively based on surface sediment samples. We conclude that the inventory of $^{239,240}$Pu still present in the Thule sediments is 25–30 Ci, or ~400–500 g.

### *Seaweed*

*Laminaria* and *Fucus* species were collected along the coasts of Bylot Sound in 1970 and 1974. The geometric mean levels were 2.1 pCi $^{239,240}$Pu kg$^{-1}$ wet weight (13 pCi kg$^{-1}$ dry weight) in 1970 (7 samples) and 2.4 pCi $^{239,240}$Pu kg$^{-1}$ wet weight (16 pCi kg$^{-1}$ dry weight in 1974 (3 samples) (cf. Table 3).

Concentration factors of the order of $3 \cdot 10^3$

*Table 3 Plutonium in brown algae collected at Thule, 1970 and 1974*

| Sampling year | Locality (cf. Fig. 1) | Species | pCi $^{239,240}$Pu kg$^{-1}$ fresh weight | pCi $^{239,240}$Pu kg$^{-1}$ dry weight |
|---|---|---|---|---|
| 1970 | Narssarsuk | *Laminaria* sp | — | 6 |
| 1970 | Saunders Island SW | *Fucus* sp | — | 24 |
| 1970 | Wolstenholme Island N | *Laminaria* sp | — | 5 |
| 1970 | Dundas | *Fucus* sp | — | 22 |
| 1970 | 5 km south of Manson Island | *Fucus* sp | — | 35 |
| 1970 | Wolstenholme Island SE | *Fucus* sp | — | 9 |
| 1970 | Manson Island | *Laminaria* sp | — | 12 |
| 1974 | Narssarsuk | *Laminaria* sp | 0.42 | 3 |
| 1974 | Dundas | *Fucus* sp | 8, 4 | 55, 25 |

were found for brown algae in Thule in 1968 (Aa71a). From this we may estimate the seawater level to be of the order of 1 fCi $^{239,240}$Pu l.$^{-1}$, which is compatible with the observations in Table 1.

*Zooplankton*

Mixed samples of zooplankton were collected in Bylot Sound both in 1970 and 1974. The mean level of three samples in 1970 was $1.1 \pm 0.3$ (S.E.) pCi $^{239,240}$Pu kg$^{-1}$ fresh weight; in 1974 we found 0.4 pCi $^{239,240}$Pu kg$^{-1}$ fresh weight.

In the plutonium studies in Lake Michigan (Wa75) it was observed that the concentration factor (CF) for plutonium followed the expression: CF = 200 (% ash)$^{1.4}$ for phytoplankton as well as zooplankton. In 1970 the mean ash content of the samples from Thule was 2.2% and in 1974 3.2%. If we assume that the Lake Michigan equation also applies to the marine environment at Thule, the expected concentration factors should be: $0.6 \cdot 10^3$ in 1970 and $1 \cdot 10^3$ in 1974 and the seawater levels: 1.8 fCi l$^{-1}$ and 0.4 fCi l.$^{-1}$, respectively. Table 1 shows that these estimates are of the right order of magnitude.

*Brittlestars* (Ophiura) *and starfish* (Asterias)

Echinodermata (generally brittlestars) were present in most bottom samples and contained from 1 to nearly 6000 pCi $^{239,240}$Pu kg$^{-1}$ fresh weight. Although the highest plutonium concentrations were usually found closest to the point of impact, it was not possible to demonstrate any significant variation in the levels. Figure 4 shows a log–log plot of Pu concentrations in echinoderms found in 1970 and 1974 at various distances from the point of impact. The data indicates no time trend.

*Molluscs*

Bivalves were the most extensively sampled marine biota at Thule. Plutonium concentrations in their soft parts (Fig. 5) were related to the sampling distance from the point of impact and showed a marked decrease in the Pu values from 1968 to 1970, followed by a much slower decline to 1974 values. The decreased rate of change is presumably due to the dispersal and dilution with distance of the Pu. A

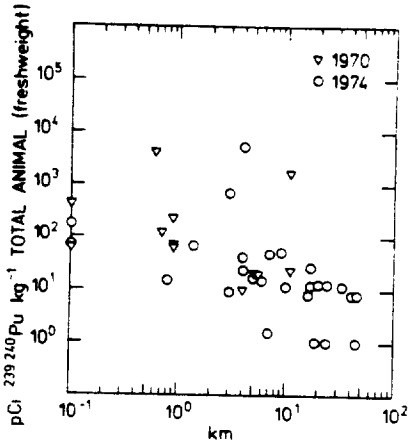


FIG. 4. The $^{239,240}$Pu concentration in brittlestars and seastars collected at Thule in 1970 and 1974 related to the distance from the point of impact.

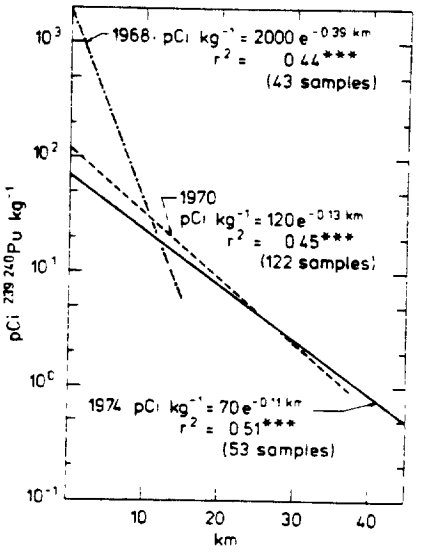


FIG. 5. The $^{239,240}$Pu concentration in soft parts of bivalves collected in 1968, 1970 and 1974 as a function of distance from the point of impact; $r^2$ is the correlation coefficient between observed data and the values calculated from the exponential equations. (Significance level: * * *: 0,001, i.e. highly significant).

*Table 5 Plutonium in shells of molluscs collected in Thule 1974*

| Location (Fig 1) | Distance from point of impact in km | Species | pCi $^{239,240}$Pu g$^{-1}$ ash |
|---|---|---|---|
| A | 45 | *Macoma calcarea* | 0.01 |
| | | *Clinocardium ciliatum* | ~0 |
| B | 24 | *Serripes groenlandia* | 0.05 |
| | | *Clinocardium ciliatum* | ~0 |
| C | 17 | *Macoma calcarea* | 0 07 |
| | | *Macoma calcarea* | 0.02 |
| D | 16 | *Hiatella sinaia* | 0 02 |
| | | *Astarte montagui* | 0 03 |
| | | *Hiatella sinaia* | 0 05 |
| E | 33 | *Macoma calcarea* | 0 01 |
| | | *Musculus laevigatus* | ~0 |
| | | *Clinocardium ciliatum* | ~0 |
| | | *Macoma calcarea* | 0 06 |
| F | 7 | *Chlamys islandica* | 0 05 |
| | | *Clinocardium ciliatum* | 0.03, 0 02 |
| | | *Astarte montagui* | 0 04 |
| G | 9 | *Macoma calcarea* | 0 10 |
| | | *Macoma calcarea* | 0 19 |
| I | 6 | *Clinocardium ciliatum* | 0 08 |
| | | *Chlamys islandica* | 0 04 |
| | | *Hiatella sinaia* | 0 06 |
| M | 40 | *Astarte montagui* | 0 01 |
| P | 4 | *Clinocardium ciliatum* | 0 88 |
| R | 4 | *Astarte montagui* | 0 03 |
| V | 0 | *Macoma calcarea* | 0 59 |
| | | *Clinocardium ciliatum* | 1 56 |
| X | 0 8 | *Macoma calcarea* | 0 48 |
| Y | 0 8 | *Hiatella sinaia* | 0 22 |

Comment ~0 means below detection limit (<0 005 pCi g$^{-1}$ ash)

*Table 6 Plutonium in soft parts of snails (Buccinum) collected at Thule 1974*

| Location (Fig 1) | Distance from point of impact in km | pCi $^{239,240}$Pu kg$^{-1}$ fresh weight |
|---|---|---|
| B | 24 | ~0 |
| E | 33 | ~0. 5 |
| F | 7 | 2 |
| I | 6 | 6 |
| N | 19 | 0 4 |
| O | 10 | 5 |
| S | 20 | 2 |
| V | 0 | 37 |

Comment ~0 means below detection limit (<0 1 pCi kg$^{-1}$) 1 kg fresh weight ~3" g ash ~300 g dry matter

concentration in the soft parts of snails to be 4–5 times lower than that of bivalves.

*Worms*

As the plutonium contamination in the sediments approximately decreased exponentially with distance from the point of impact, it was obvious also to relate the plutonium levels in the bottom animals to the distance. Worms (Polychaeta) contain sediments in their digestive tracts, and a rather close relationship to the radioactivity concentrations in sediments was to be expected. Figure 6 shows a comparison. A three-way ANOVA of data of surface sediments and the worms from 1970 and 1974, grouped according to distance from the point of impact, showed no interaction between sample species and years, nor between years and locations or locations and

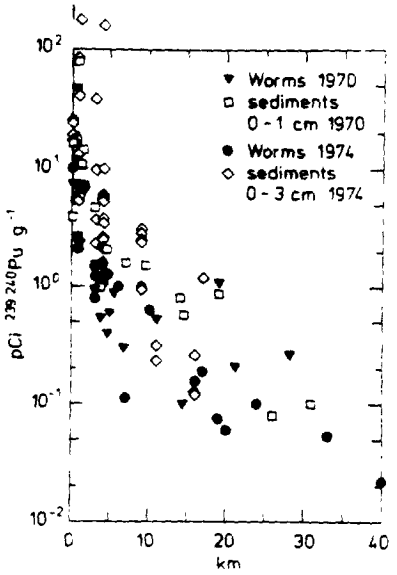


FIG. 6 The $^{239,240}$Pu concentrations in ash of worms and surface sediments samples collected in 1970 and 1974 related to the distance from the point of impact.

species: neither was there any significant difference between sampling years. The plutonium concentration in the ashed worms was 2.8 times less than the concentration in sediments, and the values decreased exponentially with distance from the point of impact. It is calculated that, as regards the plutonium concentration in pCi g$^{-1}$ of ashed material, a worm sample in the present material corresponds to a sediment sample collected down to a depth of 8.4 cm. According to equation (1) we may estimate the sample Pu content to be 96.5% of the total plutonium of the sediment column. From the ten HAPS cores collected we estimate the ash weight per m$^2$ of the 0–8.4 cm sediment layer to be 58.8 kg.

The plutonium content of the combined worm material collected in 1970 and 1974 (Fig. 6) follows the expression:

$$\text{pCi g}^{-1} = 2.47 \cdot e^{-0.13R}$$

or

$$\text{mCi km}^{-2} = 151e^{-0.13R}$$

The integrated plutonium level becomes ~31 Ci which is in agreement with the estimates based on sediments.

We may conclude that worms, due to their ingestion of sediments, may be considered to

be "sediment equivalents" for the purpose of inventory determinations after an appropriate "calibration."

*Shrimps* (Arcturus)

Both in 1970 and in 1974 shrimps were collected from the point of impact out to a distance of ~40 km (Fig. 7). Similar to the echinoderms, there was a slight decrease in the plutonium levels with distance, but less definitive than in the case of worms and bivalves. The explanation may partly be the greater mobility of the crustaceans (and echinoderms), partly the more pelagic nature of these animals.

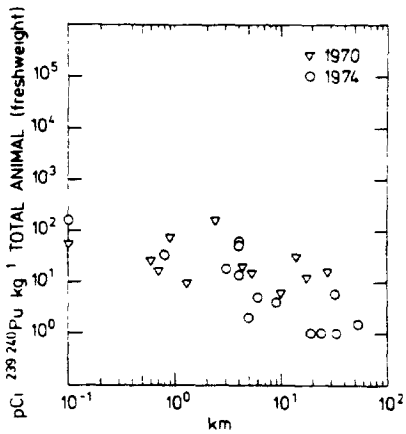


FIG. 7. The $^{239,240}$Pu concentration in shrimps collected at Thule in 1970 and 1974 related to the distance from the point of impact.

In a few cases shells and flesh of shrimps were analyzed separately; the mean ratio between the plutonium concentrations in the two parts was 6; in 1968 the mean ratio of five samples was 15 (Aa71a). Experimental uptake studies (Fo75) have shown a concentration ratio between shells and soft parts of 3.3. The shells of shrimps from Thule have probably been contaminated by particulate plutonium from adhering sediments, which fact may have increased the ratios observed.

*Fish*

Eleven samples of fish (sea scorpion, Polar cod, *Agonida*, *Lycodida*) caught in Bylot Sound in 1970 and three samples from 1974 were analyzed for plutonium. The geometric mean levels of the two samplings (Table 7) were 4 and 1 pCi $^{239,240}$Pu kg$^{-1}$ fresh weight, respectively, being an order of magnitude less than values determined in 1968.

*Table 7 Plutonium in fish samples caught in Bylot Sound in 1970 and 1974*

| Sampling year | Species | Sample type | pCi $^{239,240}$Pu kg$^{-1}$ fresh weight |
|---|---|---|---|
| 1970 | Polar cod | Total fish | 2.5, 5 |
| 1970 | Polar cod | Total fish | 400 |
| 1970 | *Lycodida* | Total fish | 20 |
| 1970 | *Lycodida* | Total fish | 2 |
| 1970 | *Lycodida* | Total fish | 4.6 |
| 1970 | *Lycodida* | Total fish | 2 |
| 1970 | *Agonida* | Total fish | 5 |
| 1970 | *Agonida* | Total fish | 9 |
| 1970 | *Agonida* | Total fish | 4 |
| 1970 | Sea scorpion | Flesh | 0.2 |
| 1970 | Sea scorpion | Liver | 0.8 |
| 1974 | Polar cod | Total fish | ~0 |
| 1974 | Sea scorpion | Total fish | 10 |
| 1974 | Sea scorpion | Liver | 0.3 |
| 1974 | Unidentified | Total fish | 1.5 |

Comments ~0 means below detection limit (<0.1 pCi kg$^{-1}$)
Total fish 1 kg fresh weight ~44 g ash ~220 g dry matter

*Birds*

Only a few samples of seabirds (guillemots) were analyzed in 1970 and 1974. The results are shown in Table 8. As in the case of fish, there was no evidence of any increase of plutonium concentrations since 1968.

*Table 8 Plutonium in samples of seabird, shot at Thule in 1970 and 1974*

| Sampling year | Species | Sample type | pCi $^{239,240}$Pu kg$^{-1}$ fresh weight |
|---|---|---|---|
| 1970 | Black guillemot | Entrails | 3.5 |
| 1970 | Brünnich's guillemot | Liver | 128 |
| 1970 | Brünnich's guillemot | Entrails | ~0 |
| 1970 | Brünnich's guillemot | Liver | ~0 |
| 1970 | Brunnich s guillemot | Entrails | ~0 |
| 1974 | Brünnich s guillemot | Entrails | 0.3 |

Comments ~0 means below detection limit (<0.1 pCi kg$^{-1}$)

*Walrus and seal*

The last link in the marine foodchain at Thule before man (and polar bear) is occupied by marine mammals. Table 9 shows that neither in 1970 nor in 1974 did the sampling (mostly entrails) of seal and walrus show any general indication of increased plutonium levels, as compared with the observations in 1968 (Aa71a). The geometric mean of all samples was 0.5 pCi $^{239,240}$Pu kg$^{-1}$ both in 1970 and 1974. The higher levels were mostly found in the undigested contents of intestine and stomach. We therefore conclude that no significant amount of plutonium from the Thule accident has been transferred to marine mammals.

*Table 9 Plutonium in marine mammals shot at Thule in 1970 and 1974*

| Sampling year | Species | Sample type | pCi $^{239,240}$Pu kg$^{-1}$ fresh weight |
|---|---|---|---|
| 1970 | Ringed seal (*Phoca hispida*) I | Entrails | ~0 |
| 1970 | Ringed seal II | Entrails | 1.1 |
| 1970 | Ringed seal III | Entrails | 0.1, 3 6 |
| 1970 | Greenland seal (*Phoca groenlandica*) | Entrails | ~0 |
| 1970 | Seal I (unspecified) | Entrails | ~0 |
| | (unspecified) | Entrails | 2 2 |
| 1970 | Ringed seal II | Contents of intestine | 0.8 |
| 1970 | Ringed seal I | Liver | ~0 |
| 1970 | Ringed seal III | Liver | 0.6 |
| 1970 | Seal II (unspecified) | Liver | 1.0 |
| 1970 | Greenland seal | Liver | 3.9 |
| 1974 | Walrus I (*Odobenus rosmarus*) | Contents of intestine | 3.3 |
| 1974 | Walrus II | Contents of intestine | 5 5, 1 6 5 5 |
| 1974 | Walrus I | Intestine | 0 1 |
| 1974 | Walrus II | Intestine | 0 7 |
| 1974 | Walrus III | Intestine | ~0 |
| 1974 | Walrus I | Stomach contents | 1 6 |
| 1974 | Walrus III | Stomach contents | 1 4 |
| 1974 | Walrus I | Stomach | 0.2 |
| 1974 | Walrus II | Stomach | 0.04 |
| 1974 | Walrus III | Stomach | 0 8 |
| 1974 | Walrus I | Liver | 0.17 |
| 1974 | Walrus II | Liver | 0.20 |
| 1974 | Walrus III | Liver | 0 03 |

Comment ~0 means below detection limit (<0 1 pCi kg$^{-1}$)

The Eskimos occasionally collect bivalves from the walrus stomachs and thus have a direct access to the more contaminated links of the marine foodchain. The high degree of discrimination against plutonium by the gastrointestinal tract, however, reduces the importance of this type of exposure; this was also demonstrated by the low Pu levels found in walrus although they may have fed on contaminated bivalves.

## *Summary*

The plutonium concentrations determined in environmental samples collected in 1970 and 1974 at Thule can be used for an estimate of the inventories in the various compartments of the benthic food-chain. The sediments contain practically all the plutonium released as a result of the accident in 1968. The benthos contains less than 1‰ of the total Pu-inventory (Table 10). Molluscs are estimated to contain the major part of the plutonium in the biota. The transfer-coefficient to the molluscs is an order of magnitude greater than the transfer-coefficient to brittlestars and shrimps. If, however, we compare the transfer-coefficients based on concentrations (Ci · g$^{-1}$ · yr pr. Ci released), there is no significant difference between molluscs (soft parts), brittlestars and shrimps. We may thus

*Table 10 Estimates of $^{239,240}$Pu inventories and transfer-coefficients in the environment of Thule*

| Sample | Estimated biomass (Vi50) g m$^{-2}$ | Inventory in Ci 1970 | Inventory in Ci 1974 | Transfer from the release of 1 Ci in Ci yr |
|---|---|---|---|---|
| Molluscs | | | | |
| Soft parts | $10^2$ | $2.4 \cdot 10^{-3}$ | $1.9 \cdot 10^{-3}$ | $1.5 \cdot 10^{-3}$ |
| Shells | $10^2$ | $6 \cdot 10^{-3}$ | $5 \cdot 10^{-3}$ | $4 \cdot 10^{-3}$ |
| Brittlestars (total animal) | 3 10 | $3 \cdot 10^{-3}$ | $8 \cdot 10^{-4}$ | $6 \cdot 10^{-4}$ |
| Shrimps (total animal) | 2 10 | $1 \cdot 10^{-3}$ | $2 \cdot 10^{-4}$ | $2 \cdot 10^{-4}$ |
| Worms (total animal) | 10 | $5 \cdot 10^{-4}$ | $5 \cdot 10^{-4}$ | — |
| Sediments | — | $3 \cdot 10$ | $3 \cdot 10$ | — |

The inventories of the molluscs were taken from the text. Figures 4 and 7 were used for the estimates of brittlestars and shrimps. The inventory in worms was calculated from the exponential equation for worms

expect nearly the same Pu concentrations in these animal groups from a given plutonium release to the sea. In Table 10 it has been assumed that the decay of the Pu-inventories observed from 1970 to 1974 would continue as single, exponential decays to infinity. The infinite integral of the respective exponential expressions calculated for the various sample types divided by 30 thus corresponds to the transfer-coefficient for the sample in question in the Thule environment (Ci · yr pr. Ci released).

The estimates are preliminary, and it is probably an over-simplification to expect the decay of the plutonium concentrations in the biota at Thule to follow a single, exponential decay. From 1968 to 1970 we observed a faster decay in molluscs than from 1970 to 1974, and we can expect the decay rates to decrease with time a factor, which will increase the transfer-coefficients for the various biota. Furthermore, it should be noted that the estimates of the biomass of the various animal groups in the Thule area only indicate the order of magnitude.

As regards the transfer of plutonium from the sediments to the seawater we may calculate $K_d$ values [= (pCi g$^{-1}$ surface sediments/pCi ml$^{-1}$ sea water)] from the data given in Table 1 and Fig. 2. The geometric mean of these $K_d$ values was ~$10^6$ ml g$^{-1}$ (range: $4 \cdot 10^4 - 6 \cdot 10^7$).

In a study of $^{239,240}$Pu in groundwater and groundwater particulates at Enewetak Atoll (No75) $K_d$ values between $10^4$ and $10^6$ ml g$^{-1}$ were observed. In a laboratory experiment with seawater and contaminated soil from

Enewetak Atoll a $K_d$ mean value $8 \cdot 10^4$ ml $g^{-1}$ was measured. The high $K_d$ values at Thule were found near to the point of impact, where we expect plutonium particulates to have largest grain size. We therefore conclude that the plutonium from the Thule accident probably shows higher $K_d$ values, and thus lesser solubility, than the fallout plutonium found at Enewetak Atoll.

*Plutonium-238*

In a number of the samples collected near the point of impact $^{238}$Pu was determined together with $^{239,240}$Pu. The mean percentage of $^{238}$Pu as compared with $^{239,240}$Pu was 1.9 ± 0.1 (SE), equal to the mean observed in 1968 (Aa71a). It has been suggested (Han75) that some physical separation process perhaps due to radioactive decay, may account for an apparently increased biological availability of $^{238}$Pu. Although Table 11 shows a mean ratio in bivalves of 2.1, as compared to 1.7 in sediments and 1.8 in worms, the data were too few to prove that the $^{238}$Pu/$^{239,240}$Pu ratio was significantly higher in bivalves than in sediments. Robertson (pers. comm.) has performed a more comprehensive study of $^{238}$Pu in the Thule sediments and demonstrated a variation of the $^{238}$Pu/$^{239,240}$Pu ratio with depth which may be a result of biological activity in the sediments.

*Table 11* $^{238}$Pu/$^{239,240}$Pu *in sediments, bivalves and worms collected near the point of impact at Thule in 1974*

| Location (Fig. 1) | Sample type | $^{238}$Pu/$^{239,240}$Pu × 100 |
|---|---|---|
| Q | PK-sediment (0–3 mm) | 1.6 |
| X | PK-sediment (0–4.3 cm) | 1.7 |
| Y | PK-sediment (0–1.8 cm) | 1.9 |
| V | PK-sediment (0–2.4 cm) | 1.6 |
| X | *Macoma calcarea* (soft parts) | 2.7 |
| Y | *Macoma calcarea* (soft parts) | 2.4 |
| V | *Macoma calcarea* (soft parts) | 1.7 |
| V | *Macoma calcarea* (soft parts) | 1.8 |
| Y | *Clinocardium ciliatum* (soft parts) | 1.7 |
| Y | *Chlamys islandica* (soft parts) | 2.3 |
| V | *Hiatella striata* (soft parts) | 2.2 |
| V | *Hiatella striata* (soft parts) | 1.8 |
| V | Mixed sample of bivalves (soft parts) | 1.8 |
| X | Worms | 2.1 |
| Y | Worms | 1.6 |
| V | Worms | 1.6 |
| V | Worms | 1.9 |

*Table 12 Summary of* $^{239,240}$Pu *data from the sample collections in 1968, 1970 and 1974*

| Sample | Unit | | 1968 I 0–1 km | 1968 II >1 km | 1970 I 0–1 km | 1970 II >1 km | 1974 I 0–1 km | 1974 II >1 km |
|---|---|---|---|---|---|---|---|---|
| Seawater (surface 0–100 m) | fCi $l^{-1}$ | max | 12 | 67 | 3 | — | — | — |
| | | min | 4 | 2 | 2 | — | — | — |
| | | geometric mean | 6 (4) | 6 (8) | 2 (2) | 1 (1) | 1 (1) | — |
| "PK" Sediments (0–1 cm) (ash) | pCi $g^{-1}$ | max | 130 | 16 | 86 | 4.7 | 50 | 78 |
| | | min | 7 | 0.1 | 6 | 0.4 | 8 | 0.3 |
| | | geometric mean | 23 (9) | 1.0 (7) | 13 (9) | 1.2 (10) | 17 (3) | 5.0 (9) |
| Sea plants (wet weight) | pCi $kg^{-1}$ | max | — | 74 | — | 6 | — | 8 |
| | | min | — | 6 | — | 1 | — | 0.4 |
| | | geometric mean | — | 19 (7) | — | 2.1 (7) | — | 2.4 (3) |
| Worms (ash weight) | pCi $g^{-1}$ | max | — | — | 46 | 1.1 | 10.5 | 6.9 |
| | | min | — | — | 0.3 | 0.1 | 2.1 | 0.06 |
| | | geometric mean | 230 (1) | — | 3.4 (6) | 0.48 (11) | 5.7 (4) | 0.54 (15) |
| Bivalves (soft parts) (fresh weight) | pCi $kg^{-1}$ | max | 76,000 | 5,400 | 73,000 | 13,000 | 1900 | 300 |
| | | min | 320 | 5 | 50 | 1 | 75 | 2 |
| | | geometric mean | 4,600 (10) | 83 (33) | 390 (15) | 23 (79) | 240 (12) | 15 (30) |
| Brittlestars Seastars (fresh weight) | pCi $kg^{-1}$ | max. | 1,120 | — | 4,400 | 1,700 | 250 | 64 |
| | | min | 190 | — | 10 | 10 | 62 | 1 |
| | | geometric mean | 380 (4) | — | 140 (7) | 44 (7) | 81 (4) | 9 (14) |
| Shrimps (fresh weight) | pCi $kg^{-1}$ | max | — | 12,000 | 76 | 170 | 160 | 64 |
| | | min | — | 22 | 16 | 1 | 33 | 1 |
| | | geometric mean | 41 (1) | 1,130 (4) | 35 (2) | 16 (10) | 72 (2) | 9 (8) |
| Fish (fresh weight) | pCi $kg^{-1}$ | max | — | 470 | — | 400 | — | 10 |
| | | min | — | 1 | — | 0.2 | — | <0.1 |
| | | geometric mean | — | 40 (10) | — | 4 (11) | — | 1 (4) |
| Birds (Fresh weight entrails) | pCi $kg^{-1}$ | max. | — | 7 | — | 130 | — | — |
| | | min | — | 0.2 | — | <0.1 | — | — |
| | | geometric mean | — | 2.2 (5) | — | 0.9 (9) | — | 0.3 (1) |
| Seal and walrus (entrails, etc.) (fresh weight) | pCi $kg^{-1}$ | max. | — | 4.4 | — | 3.9 | — | 5.5 |
| | | min | — | 0.1 | — | <0.1 | — | <0.1 |
| | | geometric mean | — | 1.0 (10) | — | 0.5 (12) | — | 0.5 (15) |

(figures in brackets indicate the number of samples)
I Zone I, less than 1 km from the point of impact
II Zone II, outside zone I

## CONCLUSIONS

The plutonium contamination at Thule resulting from the B-52 accident in 1968 was mainly confined to the sediments which contained ~25–30 Ci. Beyond 40 km from the point of impact the environmental plutonium levels were not significantly above the fallout background.

The vertical distribution of the plutonium activity in the sediments decreased exponentially with a half-depth of ~1.7 cm. The total deposit in the sediments decreased exponentially with distance from the point of impact. The half-distance was estimated to be ~3 km. Marine worms and molluscs also showed exponentially decreasing plutonium concentrations with increasing distance, but the half-distance was 5–6 km, longer than for the sediments, probably a result of biological transport of the plutonium. The plutonium levels of other bottom animals, such as brittlestars and shrimps, also decreased with distance from the point of impact but less markedly than the worms and molluscs.

When compared with the initial sampling in 1968, the Pu concentrations in 1970 and 1974 were generally lower by an order of magnitude; the decrease from 1970 to 1974 was less marked. Molluscs, which constituted the largest number of samples, contained in 1974 ~60% of the plutonium concentration found in 1970, but for the other samples the decrease was not evident. The plutonium levels in sediment and worms, for example, did not change significantly from 1970 to 1974. We may, therefore expect a rather slow reduction in the plutonium levels of the marine environment at Thule from now on.

Neither in 1970 nor in 1974 did we find evidence of any significant increase of the plutonium concentration in the higher animals such as fish, seabirds and marine mammals at Thule. The plutonium originating from the accident has to date been confined to the bottom fauna, and man has not been at risk.

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## REFERENCES

Aa71a Aarkrog A., 1971, "Radioecological Investigations of Plutonium in an Arctic Marine Environment," *Health Phys.* **20,** 31.

Aa71b Aarkrog A., 1971. "A Re-examination of Plutonium at Thule, Greenland, in 1970," *Proc. Symp. on Radioecology* p. 1213. Rome, Italy.

Aa70 Aarkrog A. 1970, "Radioecological Investigations," Project Crested Ice., Risö Report No. 213, p. 74.

Bo75 Bowen V. T., Livingston H. D. and Burke J. C., 1975 "Distributions of Transuranium Nuclides in Sediment and Biota of the North Atlantic Ocean," *Proc. Symp. on Transuranium Nuclides in the Environment* p. 107, San Francisco, U.S.A.

Ed75 Edgington D. N. and Robbins J A., 1975, "The Behaviour of Plutonium and other Long-lived Radionuclides in Lake Michigan. II. Patterns of Deposition in the Sediments, " *Proc. Symp. on Impacts of Nuclear Releases into the Aquatic Environment,* p. 245, Otaniemi, Finland.

Fo75 Fowler S., Heyrand M. and Beasley T. M., 1975, "Experimental Studies on Plutonium Kinetics in Marine Biota," *Proc Symp. on Impacts of Nuclear Releases into the Aquatic Environment,* p. 157, Otaniemi, Finland.

Han75 Hanson W. C., 1975, "Ecological Considerations of the Behaviour of Plutonium in the Environment," *Health Phys.* **28,** 529.

Har73 Hardy E. P., Krey P. W. and Volchok H. L., 1973, "Global Inventory and Distribution of Fallout Plutonium," *Nature, Lond.* **241,** 444.

He75 Hetherington J. A., Jefferies D. F. and Lovett M. B., 1975, "Some Investigations into the Behaviour of Plutonium in the Marine Environment," *Proc. Symp. on Impacts of Nuclear Releases into the Aquatic Environment,*" p. 193, Otaniemi, Finland.

Ka73 Kanneworff E. and Nicolaisen W., 1973, "The HAPS a Frame Supported Bottom Corer," *Ophelia* **10,** 119

Mc75 McLendon H. R., Stewart D. M., Boni A. L., Corey J. C., McLeod K. W. and Pinder J. E., 1975, "Relationships among Plutonium Contents of Soil. Vegetation and Animals collected on an adjacent to an Integrated Nuclear Complex in the humid Southeastern U.S.A.," *Proc IAEA Symp. on Transuranium Nuclides in the Environment* p. 347, San Francisco, U.S.A.

No72 Noshkin V. E., 1972, "Ecological Aspects of Plutonium Dissemination in Aquatic Environments," *Health Phys.* **22,** 537.

No75 Noshkin V. E., Wong K. M., Marsh K., Eagle R. and Holladay G., 1975, "Plutonium Radionuclides in the Groundwaters at Enewetak Atoll," *Proc. IAEA Symp. on Transuranium Nuclides in the Environment,* p. 517, San Francisco, U.S.A.

Ta71 Talvitie N. A., 1971, Radiochemical Determination of Plutonium in Environmental and Biological Samples by Ion Exchange," *Analyt. Chem.* **43,** 1827.

Vi50 Vibe C., 1950, "The Marine Mammals and the Marine Fauna in the Thule District (Northwest Greenland) With Observations on Ice Conditions in 1939–41," *Meddr. Grønland* **150,** 116.

Wa75 Wahlgren M. A. and Marshall J. S., 1975, "The Behaviour of Plutonium and other Long-Lived Radionuclides in Lake Michigan. I Biological Transport, Seasonal Cycling and Residence Times in the Water Column," *Proc. Symp on Impacts of Nuclear Releases into the Aquanc Environment,* p. 227, Otaniemi, Finland.