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STUDIES OF TRANSURANICS IN AN ARCTIC MARINE ENVIRONMENT

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Since the accidental loss of four nuclear weapons by a B-52 at Thule Airbase, Greenland in 1968, the marine environment at Thule has showed enhanced levels of Pu and Am. Most of the activity is confined to the benthic environment within a distance of 50 km from the crash site of the B-52. Samplings of sediments, benthos, seaplants, fish, and water have been carried out in 1968,1970, 1974, 1979, and in 1984. The study presented herein intends to answer the following questions: What is the mean residence time of these transuranics in the benthic communities? Do Pu and Am behave differently in the environment?

Introduction

On 21 January 1968, a B-52 airplane from the U.S. Strategic Air Command crashed on the ice in Bylot Sound 11 km west of Thule Air Base, Greenland (Fig. 1). The environment was contaminated by plutonium and americium from the nuclear weapons carried by the plane. Most of the activity was removed from the surface of the sea-ice. However, approximately half a kilogram of plutonium went to the sea bottom at the point of impact. The environmental behavior of this contamination has since been studied by expeditions to Thule in 1968, 1970, 1974, 1979, and in 1984[1,2,3]. This report will summarize some of the results obtained hitherto. The dose commitment to man from the Thule accident will depend upon the environmental rather than the radiological mean residence time of plutonium. The radiological mean residence time of ^{239}Pu is approximately 35000 years. However, plutonium disappear faster than that from the biosphere. From studying the decay of the Pu-inventories

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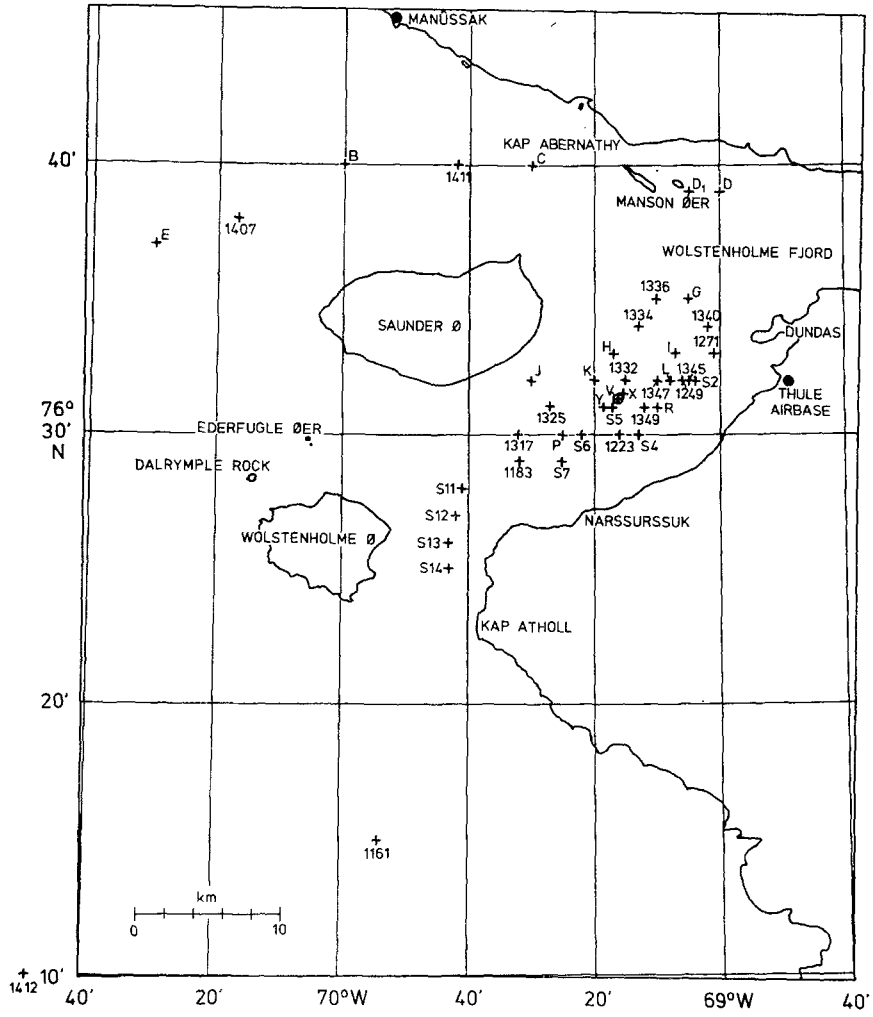

Fig. 1. Sampling locations at Thule in August 1984

in the benthic communities at Thule, we may get an estimate of the environmental mean residence time of plutonium, and thus be able to estimate a dose commitment.

Methods and materials

The methods of sampling and radiochemical procedures have been described in earlier publications [1,2,3].

The 1984 expedition, which was a cooperation between Risø National Laboratory, Denmark, Bedford Institute of Oceanography, Canada, International Laboratory of Marine Radioactivity, Monaco, and the University of Lund, Sweden, took place on board the Canadian ship CSS Baffin. As opposed to earlier expeditions, this one concentrated on sediment samples collected in the deep trench in Bylot Sound. The hypothesis was that earlier samplings might have underestimated the inventories, because a relatively large amount of the activity might have gone to the trench.

Results and discussion

Sediments

The fallout background of $^{239,240}Pu$ in the Thule sediments was determined from the measurement of ^{137}Cs in the sediment samples collected in 1979 and 1984 ([3] and Table 1). These samples followed the regression:

$$\text{Bq } ^{137}\text{Cs m}^{-2} = 580\ e^{-0.053x}, \qquad (1)$$

where x was the distance in km from the point of impact. If we define a 3 cm thick sediment-layer containing less than 10 Bq ^{239,240} Pu m^{-2} as free of accidental Pu, we may consider such layers as representative for fallout Pu only. The material contained 11 such samples from Bylot Sound. The $^{239,240}Pu/^{137}Cs$ mean ratio in these samples was 0.26 ± 0.10 (1 SD). This ratio is compatible with that estimated in 1979, when we found 0.36 ± 0.17 in 5 samples [3]. Applying the ratio from 1984, the regression from the Pu-fallout background becomes:

$$\text{Bq } ^{239,240}\text{Pu m}^{-2} = 150\ e^{-0.053x} \qquad (2)$$

Table 1
^{137}Cs in 3 cm thick sediment layers collected at Thule in August 1984 (Bq m^{-2})

Station	Distance in km	0–3	3–6	6–9	9–12	12–15	15–18	18–21	$\sum$
V	0	108	177	215	223	164			887
S5	0.58	130	247	152	58	25	<18		~630
Y	0.73	62	226	132	62	27	<25		~534
X	0.98	71	127	198	177	83			655
1332	1.66	60	132	152	188	174			706
K	1.72	24	66	114					203
"	"	51	72	30	19				172
1223	2.48	73	75	100	152	222			622
S4	3.07	106	350	153	37	19			666
S6	3.12	111	198	145	41	<22	9		~527
H	3.15	90	185	166	130	119	48		738
R	3.24	149	290	360	154	36			989
L	3.45	80	130	95	20				324
P	4.07	136	277	206	82	21			722
1325	4.18	63	88	128	68				347
1347	4.26	117	264	226	96	<19	< 8		~731
1249	5.09	120	239	121	43	30	<15		~567
1334	5.35	113	211	244	161	79	20	22	849
S7	5.38	112	214	74	<24	<17			~442
I	5.48	103	121	44	<25				~293
1345	5.51	82	225	285	220	96	20	35	963
J	5.59	105	115	56	<19				~295
1317	6.75	29	91	44	27				191
1336	7.56	92	228	167	111	73	56		727
1183	7.62	177	217	60	<26	<12			~490
1271	7.75	112	298	381	274	83			1148
1340	8.31	146	92	25	12				275
G	8.69	139	176	52	<19	<24			~410
S11	11.90	46	127	128					301
S12	13.30	22	129	182	59				392
S13	14.81	46	131	86					263
D	16.11	49	36	16	27	32	17		178
S14	16.12	61	70	33					164
C	17.00	130	130	68	<18				~346
1411	19.27	18							18
B	24.39	76	38						114
1407	28.48	13							13
1412	55.93	52	49	39	21	20	<16		~197

The total deposit of $^{239,240}Pu$ could be expressed by the regressions:

$$\text{in 1984 : Bq } ^{239,240}\text{Pu m}^{-2} = 7300\ e^{-0.13x} \quad (3)$$

$$\text{in 1979 : "-} = 6500\ e^{-0.13x} \quad (4)$$

$$\text{and in 1974 : "-} = 17000\ e^{-0.28x} \quad (5)$$

Table 2
^{239,240}Pu in 3 cm thick sediment layers collected at Thule in August 1984 (Bq m^{-2})

Station	Distance in km	0-3	3-6	6-9	9-12	12-15	15-18	18-21	$\sum$
V	0	5600	25300	12700	12300	45600			102000
S5	0.58	2900	3600	1670	400	900	160		9600
Y	0.73	430	1590	530	72	14	3.6		2600
X	0.98	2100	6400	66000	13600	2200			90000
1332	1.66	1260	13100	8100	3300	35000			61000
K	1.72	230	280	550					1060
"	"	510	1340	1830	79				3800
1223	2.48	480	325	850	660	2270			4600
S4	3.07	390	4800	160	23	6			5400
S6	3.12	640	1310	285	94	90	98		2500
H	3.15	1080	4800	1840	1240	1840	1240		12000
R	3.24	570	89700	380	130	7			91000
L	3.45	880	1070	280	67				2300
P	4.07	2100	2300	360	112	28			4900
1325	4.18	380	1160	900					2400
1347	4.26	390	1330	300	29	4	13		2100
1249	5.09	1000	9200	290	600	52	4		11100
1334	5.35	2400	3700	1850	4300	1260	30	37	13600
S7	5.38	470	1570	330	500	24			2900
I	5.48	550	280	71	12				910
1345	5.51	370	880	940	660	82	33		3000
J	5.59	300	800	124					1220
1317	6.75	105	310	144					560
1336	7.56	900	2100	720	480	2100	210		6500
1183	7.62	670		100	19				-
1271	7.75	460	890	700	340	45			2400
1340	8.31	530	88	25	6				650
G	8.69	450	540	510	25	4			1530
S11	11.90	23	163	183					370
S12	13.30	80	340	1540					1960
S13	14.81	180	149	82					410
D	16.11	62	5	5	13	12	4		101
S14	16.12	142	75	38					260
C	17.00	470	97	32	3				600
1411	19.27	6							
B	24.39	13	18						
1407	28.48	6							
1412	55.93	10	6	3	2.5	1.8			23

The inventory I of accident Pu remaining in the sea area is calculated from the expression:

$$I = 2\pi \cdot 10^{-6}\left[\int_0^{7.5} x(F(x)-F_0(x))\,dx + 0.4\int_{7.5}^{a} x(F(x)-F_0(x))\,dx\right] \text{ TBq}$$

where F(x) is eqv (3), (4) or (5), $F_0(x)$ is eqv (2), and a is

Table 3
^{239,240}Pu dry matter concentrations in sediments, Macoma and brittle stars collected at Thule in August 1984 (Bq kg^{-1})

Station	Distance in km	Sediment 0-3	Sediment 3-6	Sediment 6-9	Sediment 9-12	Macoma soft tiss	Macoma shell	Brittle star
V	0	470	1280	540	450			
S5	0.58	280	201	60	20			
Y	0.73	67	88	24	3.6			
X	0.98	182	370	2800	530			
1332	1.66	129	640	420	163			
K	1.72	39/42	12/54	25/74	3.6	12.3	36.0	1.34
1223	2.48	46	16	34	26			
1360	2.66					10.4	5.2	
S4	3.07	46	250	8.5	1.2			
S6	3.12	64	83	13.6	3.9			
H	3.15	140	290	96	65	9.9	13.3	1.23
R	3.24	49	5400	20	6.6			
L	3.45	56	48	12.6	3.6			
P	4.07	152	117	18	5			
1325	4.18	24	54	30				
1347	4.26	47	84	18.2	1.55			
1249	5.09	112	567	15.7	31	3.4	5.0	
1334	5.35	280	250	106	240			
S7	5.38	37	66	17	25			
I	5.48	55	13.2	2.8	0.47			
1345	5.51	60	61	56	40			
J	5.59	12.9	25	3.4				
1317	6.75	32	15	4.3		3.0	2.4	0.60
1336	7.56	116	114	40	25			
1183	7.62	36		3.2	0.73			
1271	7.75	58	51	38	20	2.4	0.83	
1340	8.31	43	5.5	1.43	0.70	3.2	2.3	0.58
G	8.69	42	33	27	1.19	5.5	1.82	
S11	11.90	5.6	5.7	6.8				
S12	13.30	18.8	13.2	43		2.3	1.23	0.29
S13	14.81	20	4.3	4.0		2.0	1.75	0.36
D	16.11	3.8	0.25	0.22	0.40	0.79		
S14	16.12	12.8	2.5	1.62				
C	17.00	36	4.9	1.71	0.17	2.8	2.7	1.50
1411	19.27					0.64	0.84	
B	24.39					0.27	0.39	1.06
1407	28.48							
E	32.98					0.29	0.30	0.54
1412	55.93	1.24	0.50	0.21	0.17			

the distance where the background (2) intercepts the total activity curve ((3), (4), or (5)).
(a = 21 km in 1974 and 50 km in 1979 and in 1984)
It is assumed that the curves describe the activity distri-

bution in all directions. Furthermore, we assumed that there was only sea within 7.5 km from the point of impact, and assumed that land constitutes 60% of the surface area beyond 7.5 km.

The calculated inventories of $^{239,240}Pu$ were 0.98 TBq in 1974, 1.21 TBq in 1979, and 1.38 TBq in 1984. These estimates were not significantly different. We prefer the most recent value, because it includes more samples than any of the other ones, although our earlier sampling grids have not underestimated the Pu inventory significantly. We have, thus, not seen any tendency toward particularly high Pu sedimentation at the deep stations (S11-S14 are located at 250-300 metres of depth, deeper than other stations, and these stations are all relatively low in Pu-sedimentation). The distance from the point of impact is more important than the depth of the station.

The mean $^{241}Am/^{239,240}Pu$ ratio in the sediments collected in 1984 was 0.13. An anova (analysis of variance) showed no significant difference in the ratio among locations, and between sampling years (1979 and 1984). However, the variation as a function of sampling depth was probably significant ($P \sim 95\%$). The ratio increased with sampling depth. In the top sediment layers the ratio was, thus, approximately 0.10, while in the deeper layers it was 0.15. This may be influenced by the fact that global fallout becomes relatively more impoortant in the deeper layers, and as the $^{241}Am/^{239,240}Pu$ is higher in global fallout than in Thule debris we may expect an increase with depth.

The mean $^{238}Pu/^{239,240}Pu$ in sediments was 0.018 in 1984, which is not significantly different from the ratio of 0.017 observed in 1979. An anova showed no significant difference in the ratio for the various locations and sediment layers.

From the above ratios and the inventory estimate of $^{239,240}Pu$ the inventories of ^{241}Am and ^{238}Pu at Thule were estimated at 0.18 TBq and 0.025 TBq, respectively.

Biota

Macoma calcarea is the dominating species among the benthos at Thule. The biomass of Macoma is 0.10 kg dry matter per m^2. The $^{239,240}Pu$ activity in Macoma in 1984 followed the regression:

$$Bq\ m^{-2} = 1.05\ e^{-0.14x}. \qquad (6)$$

Integrating this equation as shown above for sediments, we estimate the inventory in the Macoma community from the accident to 0.19 GBq $^{239,240}Pu$, with an estimated uncertainty of 0.04 GBq. Fig. 2 shows the estimated inventories in Macoma from previous years. The decay of the inventories is given by:

$$I_T = 1.68\ T^{-0.85}, \qquad (7)$$

where T is the time in years since the accident (1968). After approximately 30,000 years the decay rate of the power function (7) will be equal to that of the decay rate of ^{239}Pu (λ = $2.84 \times 10^{-5}\ a^{-1}$). The time integral of the Pu concentration in the Macoma community is calculated by integrating (7) over 30,000 years, and assuming that the remaining activity at that time follows the decay of ^{239}Pu to infinity. Hence, the time integral becomes 37 GBq $^{239,240}Pu$ a. This is caused by an activity release to the sediments of 1.38 TBq, and the transfer factor to the Macoma community thus becomes

$$\frac{0.037}{1.38} = 0.027 \text{ Bq per Bq } a^{-1}$$

The area contaminated with accident Pu is:

$$\pi[(50^2-7.5^2)\cdot 0.4+7.5^2]\ km^2 = 3.25 \times 10^9\ m^2$$

The dry weight of Macoma is 0.02 kg soft tissue and ~ 0.08 kg shell per m^2. The dry weight concentrations are the same in soft tissue and in shells. Hence, the soft tissue contains 20% of the Pu inventory, or 0.2x37=7.4 GBq a. The fresh weight of

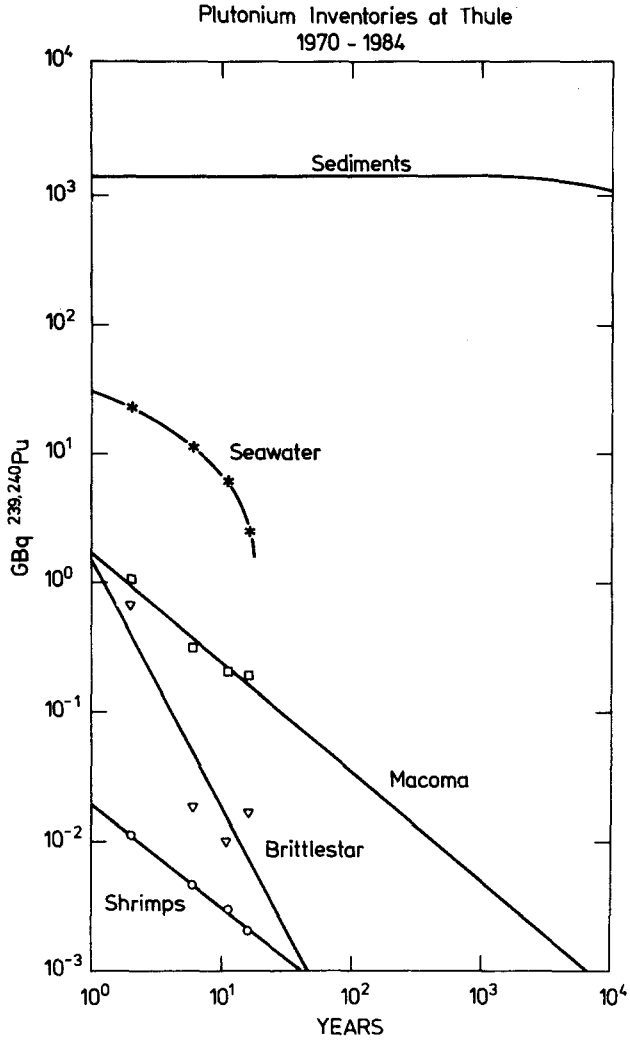

Fig. 2. ^{239,240}Pu inventories in environmental samples collected at Thule 1970–1984. Within the contaminated area ($3.25 \cdot 10^9$ m^2) the fresh weight biomass of shrimps was $0.11 \cdot 10^9$ kg, of brittle star: $0.062 \cdot 10^9$ kg, and of Macoma: flesh: $0.32 \cdot 10^9$ kg, and shell: $0.26 \cdot 10^9$ kg. The seawater mass was $3 \cdot 10^{14}$ kg and the mass (dry weight) of the 0–15 cm sediment layer was $3 \cdot 10^{11}$ kg. The abscissa is the time in years since the accident in 1968.

the Macoma soft tissue is 0.1 kg m^{-2}. Hence, the time integral 23 Bq kg^{-1} a. If we assume that a member of the critical group consumes 20 kg of soft tissue of Macoma per year, the committed individual intake is 20x23 Bq ^{239,240}Pu = 460 Bq. This corre-

sponds to a dose commitment of 0.57 mSv, or 57% of one year's natural background radiation, assuming that the ALI value from Pu in diet is 5 times lower than that given by ICRP for occupational exposure.[4/]

The brittle star is another typical member of the benthic community at Thule. The dry matter biomass of the brittlestar community is 9.3 g m^{-2} and the fresh weight is 19 g m^{-2}. Fig. 2 shows the calculated Pu inventories in brittle stars since 1970.

The decay of inventories in brittle stars is given by:

$$I_T = 1.46\ T^{-1.9} \qquad (8)$$

From (8) we may calculate the infinite time integral of Pu in the brittle star community as we did for Macoma to be 0.003 GBq a or 0.05 Bq kg^{-1} a (fresh weight). The transfer factor is then $2x10^{-6}$ Bq per Bq a^{-1}, or 0.04 Bq kg^{-1} per TBq a^{-1}.

Shrimps have also been measured throughout the years, and Fig. 2 shows how the inventories have decayed:

$$I_T = 0.019\ T^{-0.8}. \qquad (9)$$

The transfer factor to the shrimp community was 0.65 GBqa/1.38 TBq = 0.5 GBq per TBq a^{-1}. The biomass of shrimps at Thule is 0.035 kg m^{-2}, and the total mass of shrimps at the contaminated area ($3.25x10^9$ m^2) is $0.11x10^9$ kg. The dose commitment to a member of the critical group consuming 20 kg of shrimps per year would be $20x4.2x0.05x5/2x10^5$ Sv=0.1 mSv, or 10% at one year's background exposure. The dose from ^{210}Po in the shrimps would amount to 0.2 mSv [5/] in 50 years.

If we compare the $^{239,240}Pu$ concentrations in the 0-3 cm sediment layer with that in Macoma and brittle stars an

anova shows that the relative levels (dry matter concentrations) were:

100 : Sediment
10 : Macoma flesh
8 : Macoma shell
3 : Brittle star

There was no interaction between species and locations. Hence, the concentration in benthos depends upon the sediment concentrations rather than the water concentrations, which show no local variation.

The $^{241}Am/^{239,240}Pu$ concentration in Macoma and brittle star was 0.33; i.e., 2.5 times higher than in sediments and the ratio between benthos and sediments did not differ with the distance from the point of impact. Hence there was no influence of global fallout. This shows that the organisms prefer Am to Pu.

Seawater

Fig. 2 shows the plutonium inventories in unfiltered surface seawater collected at Thule throughout the years. The Pu concentrations in surface water are not different from those found at other locations in the Northern North Atlantic in these years. We must thus conclude that the Thule accident has not resulted in enhanced surface water levels at Thule. The plutonium in the seawater arises from nuclear weapon fallout.

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