COMPARATIVE STUDIES OF PLUTONIUM INVENTORIES IN SOILS AND MARINE SEDIMENTS

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1. INTRODUCTION

The rapid growth in the worldwide use of nuclear energy and concern for the safety of man and his environment have led in recent years to increasing interest in the long-lived transuranium nuclides. Plutonium is of special importance in this context because, compared to other transuranics, it is already created in substantial amounts in present reactors, and in breeder reactors it will occur in large quantities. Plutonium is already present in the environment. The global inventory from worldwide fallout has thus been estimated at (325 ± 36) kCi $^{239,240}Pu$ (1); and Windscale has discharged approx. 10 kCi into the Irish Sea (2). Minor amounts of $^{239,240}Pu$ have been released to local environments as a result of the nuclear-weapon incidents at Palomares in Spain and at Thule in Greenland.

The present study estimates the plutonium inventories at Thule from the B-52 accident in 1968 and in Danish soils and sediments from nuclear weapons testing. A further aim of the study was to compare the vertical distributions of $^{239,240}Pu$ and ^{137}Cs in soils and sediments.

2. MATERIAL AND METHODS

The Thule samples consisted of 3 cm thick core samples collected by a 135 mm diameter coring tube (Model HAPS) out to a distance of 16 km from the point of impact where the B-52 plane crashed (3). The HAPS was also used to collect sediment samples from inner Danish waters. The soil samples were collected down to a depth of 50 cm from locations distributed throughout Denmark (4). All common types of Danish soil were represented. Uncultivated as well as cultivated soils were collected from each location. The uncultivated samples were analyzed in depth sections of 0-10 cm, 10-20 cm, 20-30 cm and 30-50 cm, and the cultivated in 0-20 cm (the ploughing layer), 20-30 cm and 30-50 cm sections.

The radiochemical plutonium analysis followed the classical procedure (5) adapted to 10 g. aliquots of dried material. Cesium-137 was measured by Ge(Li)-γ-spectroscopy.

3. RESULTS AND DISCUSSION

The vertical distribution in soils as well as in sediments of $^{239,240}Pu$ and ^{137}Cs was with good approximation described by exponential regression lines. Similar observations have been made for the contamination of sediments in the Irish Sea (2) and of a single sample of sandy loam soil from the USA (6). The exponential distribution facilitated the estimates of the total integrated activity amounts in the soil and sediment columns as:

$$A\int_0^\infty e^{-kx}dx = \frac{A}{k} \quad (x: \text{ sample depth})$$

3.1. Estimate of the plutonium inventory at Thule

The horizontal distribution of the Pu-activity at Thule followed an exponential expression:

$mCi\ ^{239,240}Pu\ km^{-2} = 460\ e^{-0.28R}$ (R: distance in km from point of impact)

Taking land area into consideration, the inventory in the marine sediments at Thule was estimated at 25-30 Ci $^{239,240}Pu$. Table 1 shows that the half-depth of the activity was constant (~ 2 cm) out to a distance of 10 km from the point of impact, whereafter it increased and approached the half-depth in fallout-contaminated sediments (~ 5 cm). As the contamination at Thule is the result of a single release of plutonium and not, as is the case for fallout Pu, of several releases distributed over decades, it was expected that the half-depths would differ as observed.

Distance in km from point of impact	Regression equation $mCi\ ^{239,240}Pu\ km^{-2}\ cm^{-1}$	Number of locations	Integrated mean deposition (d → ∞cm) $mCi\ ^{239,240}Pu\ km^{-2}$
16 - 17	$0.5\ e^{-0.2\ d}$	2	2.5
9 - 11	$9\ e^{-0.3\ d}$	2	30
2 - 4	$12\ e^{-0.3\ d}$	6	40
0 - 1	$210\ e^{-0.3\ d}$	3	700

TABLE 1. The vertical distribution of $^{239,240}Pu$ in marine sediments collected in 1974 at Thule, Greenland (d is the depth of the sample in cm). Each location was represented on average by 5 sample sections, the maximum sampling depth being 15 cm.

3.2. Plutonium and ^{137}Cs in Danish sediments and soils

Danish marine sediments contained the same amount of Pu as the uncultivated soils (Table 2). From the measurements of ^{90}Sr deposition in Denmark (4), and from the observed ratio between $^{239,240}Pu$ and ^{90}Sr in nuclear weapons debris (1), the accumulated mean level in Denmark was estimated at 1.3 mCi $^{239,240}Pu\ km^{-2}$, which is compatible with the actual observations. Cultivated soils apparently contained less $^{239,240}Pu$ than this level. Some plutonium may thus have been removed from the arable land during the cultivation of the soil. This was unexpected as ^{90}Sr and ^{137}Cs were only little depleted in cultivated soils (4). Further studies are, however, necessary before any firm conclusions may be drawn. It appeared that the penetration of plutonium was greater than that of ^{137}Cs in sediments as well as in soils. This agrees with the observations of other authors (2), (6). The half-depths of Pu and ^{137}Cs were apparently less in sediments than in soil samples. This may be an artifact, because the HAPS corer used for the sediments only works in soft materials, where the penetration depth of the radionuclides may be less as a consequence of a higher ion exchange capacity than that, e.g., of sandy soils.

Sample type	Nuclide	Regression equation mCi km^{-2} cm^{-1}	Number og locations	Integrated mean deposition ($d \to \infty$) mCi km^{-2}
Marine Sediments from inner Danish waters	$^{239,240}Pu$	$0.2\ e^{-0.15\ d}$	8	1.3
	^{137}Cs	$2.4\ e^{-0.18\ d}$		13
Uncultivated Danish Soils	$^{239,240}Pu$	$0.13\ e^{-0.10\ d}$	8	1.3
	^{137}Cs	$11\ e^{-0.12\ d}$		92
Cultivated Danish Soils	$^{239,240}Pu$	$0.07\ e^{-0.09\ d}$	10	0.8
	^{137}Cs	$10\ e^{-0.11\ d}$		91

TABLE 2. The vertical distribution of $^{239,240}Pu$ and ^{137}Cs in marine sediments and soils collected in 1975 in Denmark (4). The sediment samples consisted on average of 4 depth sections, the maximum sampling depth being 21 cm. The samples of uncultivated and cultivated soils consisted of 4 and 3 depth sections respectively, the total sampling depth being 50 cm (d is the depth of the sample in cm).

3.3. An anomalous vertical activity distribution

One of the soil samples collected from uncultivated soils in Denmark showed an unexpected distribution of the activity (Table 3). In contrast to the other samples, the radionuclide levels in this sample did not decrease with increasing sampling depth. More than half of the activity in the soil column was in fact found from 30-50 cm, and some activity may have penetrated even deeper than 50 cm. It was remarkable that the Pu/Cs and Pu/Sr ratios did not vary with sampling depth. Thus, even under anomalous circumstances, plutonium

Unit	0-10 cm	10-20 cm	20-30 cm	30-50 cm	0-50 cm
mCi $^{239,240}Pu$ km^{-2}	0.81	0.75	0.90	2.64	5.1
mCi ^{137}Cs km^{-2}	30	32	44	118	223
mCi ^{90}Sr km^{-2}	5.7	6.4	8.5	22	43
$^{239,240}Pu/^{137}Cs$	0.027	0.023	0.020	0.022	0.023
$^{239,240}Pu/^{90}Sr$	0.14	0.12	0.11	0.12	0.12

TABLE 3. The vertical distribution of $^{239,240}Pu$, ^{137}Cs and ^{90}Sr in an anomalous sample of Danish uncultivated soil collected in Jutland in 1975 (4).

may be closely related to the ^{137}Cs as well as to the ^{90}Sr activity levels. The sample was taken from a meadow in a stream valley with run-off from neighbouring hillsides. This may explain why the $^{239,240}Pu$ and ^{137}Cs levels were 2-3 times higher than expected. Had the entire soil column acted as other Danish soils, we would have seen a decreasing Pu/Sr ratio with increasing sampling depth. The relatively high ^{137}Cs and $^{239,240}Pu$ concentrations at this location may be due to a selective mobilization of these nuclides, perhaps influenced by the high organic matter content in the environment. Further studies of this location may clarify the situation.

4. CONCLUSION

The Pu levels in marine sediments and soil decreased exponentially with increasing sampling depth. The half-depth of the Pu deposit in sediments from a single release was approx. 2 cm. Plutonium from fallout showed a half-depth of approx. 5 cm in marine sediments and of 7 cm in soils. Sediments contained the same integrated Pu levels as untouched soils and the levels agreed with the total deposit of plutonium, i.e. 1.3 mCi km^{-2}. The Pu/Cs ratio was nearly constant down through the sample column, perhaps with a slight tendency to an increase with sample depth. In fallout-contaminated sediments the mean Pu/Cs ratio was approx. 0.09, in uncultivated soils 0.015, and in cultivated soils 0.01.

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