Radiocesium
Transuranium elements
Arctic Sea
Norwegian coast
Svalbard

Radiocesium
Éléments transuraniens
Arctique
Côte norvégienne
Svalbard

Radio-cesium and transuranium elements in the Greenland and Barents Seas

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ABSTRACT

The results for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am analyses of sea water, sediment and biota from the Swedish Arctic expedition, Ymer-80, in the Barents and Greenland Seas are presented in this paper. It is shown that ^{137}Cs released from European reprocessing facilities is effectively transferred by the Gulf Stream up to these latitudes. The transuranium elements $^{239+240}Pu$ and ^{241}Am present in the investigated areas originate mainly from local fall-out, but a substantial mixing with Atlantic sea water containing these nuclides takes place. The levels of ^{137}Cs found in biota like algae, seals, polar bears and birds are quite moderate and do not differ from those found in other areas, such as Greenland.

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RÉSUMÉ

Radiocésium et éléments transuraniens dans les mers de Barents et du Groënland

Les résultats d'analyses de ^{137}Cs, $^{239+240}Pu$ et ^{241}Am dans des échantillons d'eau de mer, de sédiments et d'origine biologique prélevés au cours de l'expédition suédoise dans l'Arctique Ymer-80, menée dans les mers de Barents et du Groënland, sont présentés. Les observations montrent que le ^{137}Cs rejeté par les installations européennes de retraitement de combustibles est effectivement transporté par le Gulf Stream jusqu'à ces latitudes.
Les éléments transuraniens $^{239+240}Pu$ et ^{241}Am présents dans les zones étudiées proviennent principalement des retombées atmosphériques locales, mais l'on observe un mélange substantiel avec les eaux de l'Atlantique contenant ces éléments.
Les niveaux de ^{137}Cs trouvés dans des échantillons biologiques tels que les algues, phoques, ours polaires et oiseaux sont très modérés, et ne diffèrent pas de ceux observés dans d'autres zones comme le Groënland.

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INTRODUCTION

A Swedish Arctic expedition was conducted during the period June 23-October 6 1980, to commemorate the discovery of the North-East passage by Nordenskjöld in 1879-1880. The vessel used for the expedition was the Swedish icebreaker, Ymer. The use of such a heavy icebreaker made areas permanently covered with ice accessible. The principal cruising route is shown in Figure 1.

The radioecological program was only a small part of the very extensive scientific programme undertaken on board, the aim of which was to investigate present levels and sources of man-made radioactivity in the Arctic marine environment, and also to study the pathways and distribution of these radionuclides in different compartments. Apart from cesium and plutonium isotopes, we also investigated natural actinides, such as uranium and thorium in water, as well as radon and radon daughters in the air.

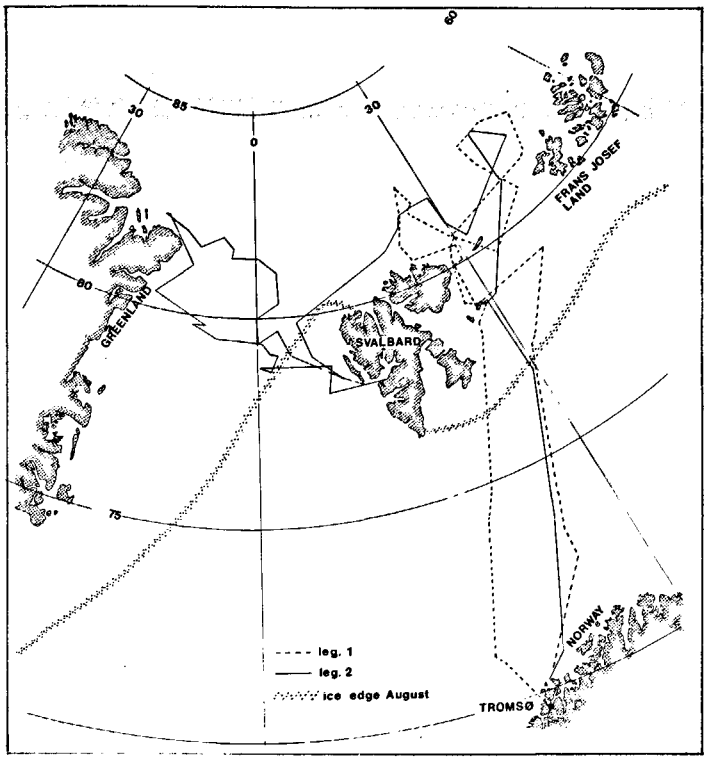

Figure 1
Cruise route for the radioecological programme of Ymer-80.

SAMPLING AND MEASUREMENTS

Sea-water samples of 100-200 l were pumped from an inlet at 7 m depth and placed in special precipitation containers.

Actinides were precipitated by adding ferrichloride and precipitated with sodium-hydroxide solution. ^{242}Pu and ^{243}Am were used and added to the water before precipitation as radiochemical yield determinants.

Cesium was separated by adsorbtion on to microchrystalline ammonium-molybdo-phosphate after ^{134}Cs had been added to the sample as a radiochemical yield determinant.

Larger volumes of sea-water similarly pumped from 7 m depth (1 000-2 000 l) were filtered through cartridge-filters (Millipore CWSS 012C3) in order to determine the fraction of the investigated radionuclides associated with particulate matter. Deepwater samples were collected with a 100 l Niskin bottle.

Algae, such as *Fucus* and *Laminaria,* were collected at sites on the shores of islands.

Terrestrial samples, such as lichen (*Cladonia*) and underlying soil, were collected on the islands of Svalbard in order to determine the integrated fall-out level.

All fauna collected by the expedition were classified and examined at the Swedish Natural Historical Museum in Stockholm. Samples of polar-bears, seals and various birds were obtained from the biology team after the expedition had been completed.

Cesium-137 was measured with gamma spectrometry of the dried precipitates and solid samples. Transuranium elements were measured with alpha-spectrometry after chemical separation by using ion-exchange and liquid extraction according to methods previously described (Talvitie, 1971; Holm *et al.*, 1979).

RESULTS AND DISCUSSION

Cesium-137 measurements

Water samples were collected between 57°N to 82.5°E. Sampling sites are shown in Figure 2. The results are also expressed in the same figure as ^{137}Cs concentration as a function of latitude. It is obvious that enhanced levels of ^{137}Cs, compared to fall-out levels, are found along the Norwegian coast. This is explained by the fact that ^{137}Cs released from European nuclear fuel reprocessing facilities is spread by the Gulf Stream along the Norwegian coast. Other investigations on the distribution of ^{137}Cs in the North Sea and adjacent areas also confirm our results (Kautsky, 1980). The results reported by Kautsky *et al.* and other investigations by us (Aarkrog *et al.*, 1982) indicate a rapid decrease of ^{137}Cs concentrations in area water, with increasing distance from the coast.

Along the Norwegian coast from 57.8°N, 8.1°E to 69.0°N, 14.4°E, the ^{137}Cs-concentration in the surface sea-water decreases by less than a factor of two. The concentration of ^{137}Cs in surface seawater collected during August 8-September 30 1980, north of 68.97°N, 14.4°E, were related to the distance in kilometres from this point.

The ^{137}Cs concentration $C(x)$ varies with the distance x km for locations east of Svalbard according to the following equation:

$$C(x) = 141.\exp(-0.00122.x) \quad (1)$$

(half-distance 568 km; 484-690 km = 95% confidence limit).

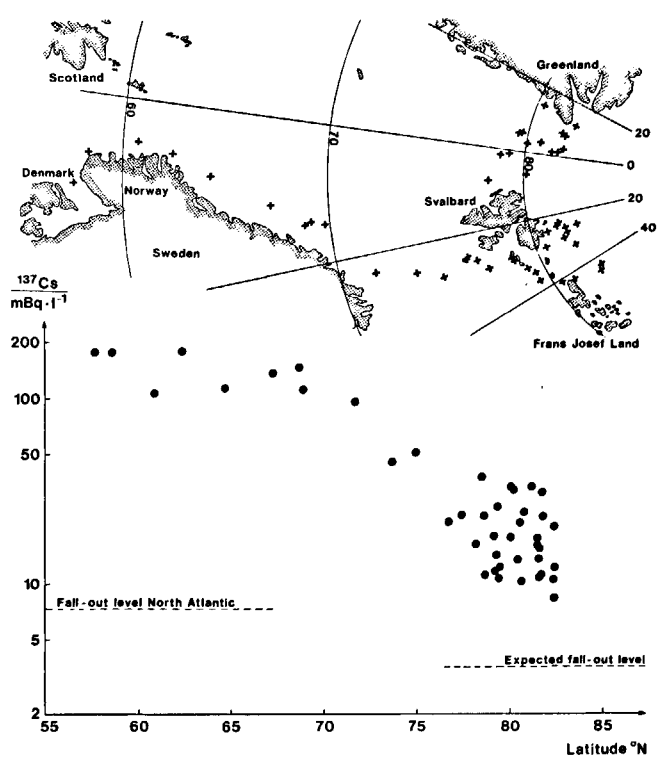

Figure 2
Stations for large volume surface sea water sampling (+) and ^{137}Cs activity concentration in water as a function of latitude (●).

For locations west of Svalbard we found the following equation:

$$C(x) = 148.\exp(-0.00164.x) \quad (2)$$

(half-distance 423 km; 362-508 km = 95% confidence limit).

The ^{137}Cs concentration in sea-water thus probably decreases more rapidly west of Svalbard than east of Svalbard. In other words, more ^{137}Cs released from European reprocessing facilities went to the east than to the west. But the difference between the ^{137}Cs concentrations in surface sea-water from the two areas was not very pronounced.

Seas is of older origin than the higher activity-ratio of $^{134}Cs/^{137}Cs$ would indicate. These ratios have not been constant in time in the effluent from Sellafield, and it is therefore difficult to derive any value for the transport time of cesium from the North Sea to the Arctic Sea.

Cesium isotopes in sediment have so far been measured in samples from some of our sampling sites. The results obtained are given in Table 1, together with the results for transuranium elements. The total area content is difficult to estimate from the few deep water samples collected. Sampling depths vary and the sites of the sample stations are widely spread. The results given in Table 2 indicate, however, that the total integrated area-content in a 1 000 m water-column and sediment is about 12 kBq.m^{-2}. This value exceeds the integrated area-content on land, which was measured to be about 2.2+/−0.3 kBq.m^{-2} on Svalbard (78.2°, 16.0°E).

Table 1
^{137}Cs, $^{239+240}Pu$, ^{241}Am in sediments.

Position	Water depth (m)	Sediment depth (cm)	^{137}Cs (Bq.kg^{-1})	$^{239+240}Pu$ (mBq.kg^{-1})	$^{241}Am/^{239+240}Pu$ (activity ratio)
82.3°N 25.3°E	3 000	0-4	1.1	33	0.44
		4-8	*ND*	8.5	0.71
			(Σ) 99	Σ 4.2	
81.8°N 26.6°E	3 200	0-4	2.6	133	0.68
		4-8	*0.7*	*21*	—
			(Σ) 240	(Σ) 11.3	
81.6°N 26.2°E	2 500	0-4	16.7	700	0.82
		4-8	*1.5*	*130*	0.64
			(Σ) 1 260	(Σ) 58	
81.5°N 26.2°E	850	0-4	7.8	600	1.3
		4-8	*1.9*	*210*	0.7
			(Σ) 650	(Σ) 57	
79.6°N 33.7°E	340	0-5	18.1	390	0.69
		5-9	*2.6*	*60*	1.0
			(Σ) 700	(Σ) 28	
80.6°N 30.1°E	380	0-4	10.0	1 010	0.77
		4-8	*4.1*	*210*	0.69
			(Σ) 780	(Σ) 61	
79.2°N 33.5°E	240	0-4	4.4	260	0.79
		4-8	*2.6*	*46*	—
			(Σ) 520	(Σ) 22	

(Σ) = integrated area content (0-8 cm) Bq.m^{-2}.

A regression was also calculated for the samples collected during the first part of the cruise (i.e. June 27-August 4), and the corresponding equation east of Svalbard was then:

$$C(x) = 77.\exp(-0.00138.x) \quad (3)$$

(half-distance 502 km; 379-740 km = 95% confidence limit).

Activity-concentration thus showed a similar decrease with distance as in August-September (cf. Equation 1), but concentration levels were about 1.8 times higher during the period August-September than in June-August (95% confidence limit, 1.5-2.5). This might be explained by the maximal melting of ice during the latter period.

The activity ratio of $^{134}Cs/^{137}Cs$ was measured at locations between latitudes of 59-77°N. This ratio was found to be in the order of 0.03-0.04, which is about the same value as is reported for the effluence from the Sellafield (formerly Windscale) reprocessing plant. This does not necessarily mean that there is any connection between the cesium isotopes at these locations. The cesium which is present in the Barents and Greenland

The amount of ^{137}Cs associated with particulate matter was as little as of the order of 4.10^{-5} along the Norwegian coast and $(0.1\text{-}2).10^{-2}$ in the Barents and Greenland Seas. It might be expected that the higher value along the Norwegian coast is due to ^{137}Cs associated with particles in the drainage from land. The contribution from this run-off is, however, small compared to the amount of ^{137}Cs in soluble form originating from reprocessing plants and carried by currents of water.

The results for biological samples are presented in Table 2. An algae/sea-water activity-concentration ratio for ^{137}Cs of about 75 (on dry weight basis) was found for *Fucus* and *Laminaria*. About the same algae/sea-water activity ratios were found along the Norwegian coast (Christensen, 1982). The activity-concentrations of ^{137}Cs in the flesh of polar-bears, seals and birds agree with those found at places in Greenland (Aarkrog, 1979).

Table 2
Cesium, plutonium and americium activity concentrations (dry weight) in biological samples.

Matrix (No. of sample)	Place of collection	Date of collection	^{137}Cs ($Bq.kg^{-1}$)	$^{239+240}Pu$ ($mBq.kg^{-1}$)	^{241}Am ($mBq.kg^{-1}$)
Fucus (1)	Svalbard	10/9/1980	1.8±0.6	130±25	8±2
Laminaria leaves (3)	Svalbard	8/7-10/9/1980	1.1±0.5	70±30	10±4
Laminaria stems (3)	Svalbard	8/7-10/9/1980	1.7±0.6	100±40	10±4
Lichen *Cladonia* (5)	Svalbard	10/9/1980	580±20	4 500±1 000	1 600±400
Seal (1)	Barents sea	15/9/1980	1.1±0.1	NA	NA
Polar bear (2)	Barents sea	10/9/1980	6.0±0.3	4.4±1.0	NA
Birds:					
Somateria mollissima (7)	Kinnvika Svalbard	18/8/1980	1.9±0.2	NA	NA
Rissa tridactyla (7)	80°N, 23°E-82°N, 46°E	12/7-27/7/1980	1.1±0.4	NA	NA
Pagophilia eburnea (2)	Kinnvika	18/8/1980	3.6±1.6	NA	NA
Uria lomvia (4)	Kinnvika and 81°N, 42°E	5/8-20/8/1980	2.3±1.1	NA	NA
Fulmarius glacialis (6)	Kinnvika	18/8/1980	1.3±0.7	NA	NA

(n) = number of samples (for birds individuals). NA = not analysed.

Plutonium-239+240 and Americium-241

The distribution of the transuranium elements in the surface water of the Norwegian, Barents and Greenland Seas is quite different from that of ^{137}Cs. As seen from the results shown in Figure 3, the distribution of the transuranium elements is very homogenous, with an average activity-concentration of (13 +/− 2)$\mu Bq.l^{-1}$ in 48 samples (2S.E.). In contrast with cesium, there is no significant latitude dependence for the transuranium elements. These results indicate that plutonium-isotopes released from European reprocessing plants are not transported by the ocean currents to the Arctic areas. This supports the results reported by Lovett and Nelson (1978), which suggest that this released plutonium is not in a soluble form, and is thus deposited locally into the sediments.

On the other hand, the activity-concentrations of plutonium-isotopes are higher than would be expected from global fall-out in the Barents and Greenland Seas. The levels found here are equal to those found in Atlantic sea-water further south, which is contaminated with fall-out. This might be explained by the fact that fall-out plutonium is predominantly present in soluble form (Fukai *et al.*, 1981) and that the mixing between Atlantic and Arctic Ocean waters is very effective.

Only a very limited number of deep-water samples have been analysed, and the results obtained so far are shown in Table 3, together with the results for cesium and americium. It was not possible to detect the generally observed sub-surface maximum (Fukai *et al.*, 1979; Livingston, Bowen, 1976) in our investigation, possibly due to the small number of samples, but the results show an increase with depth of the activity-ratio of $^{241}Am/^{239+240}Pu$.

The ^{241}Am-activity concentration in surface water shows, as in the case of plutonium, no significant correlation with the latitude. This indicates that americium released from reprocessing plants is not transported over long distances. Americium from fall-out is, to a higher degree than plutonium, associated with particulate matter, and is therefore deposited locally (Holm *et al.*, 1980).

The average $^{241}Am/^{239+240}Pu$ activity ratio was found to be 0.13 +/−0.04 (2 S.E. of 31 samples with the range 0.04-0.32) in surface water, which is lower than the value 0.37 +/− 0.05 found for integrated fall-out on Svalbard. The corresponding activity-ratio in the residue on the cartridge filter sample was 0.32 +/− 0.09 (2 S.E. of 40 samples with the range 0.06-1.3). We estimated that 32% of americium in sea water was compared to about 22% for plutonium. These values are higher than, for example, those in the Mediterranean Sea, which amount to 13% for americium and 5% for plutonium (Holm *et al.*, 1980).

The fraction of the elements associated with particulate matter must be related to the content and type of particulate matter in the water. During the summer,

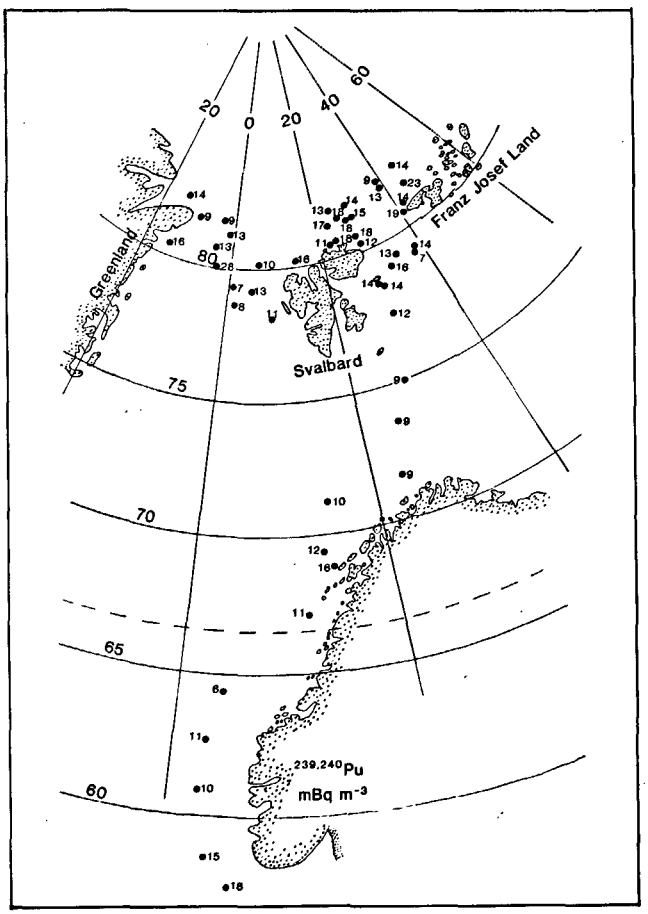

Figure 3
$^{239+240}Pu$ activity concentrations in surface sea water ($\mu Bq \times l^{-1}$).

there is a high content of organic particulate matter present in the water of the Barents and Greenland Seas.

The influence of the type of particulate matter for different elements and sedimentation processes is not yet fully understood. The distribution-factor, K_D, is defined as the ratio between radioactivity-concentration per kilogram dry suspended matter and the activity concentration per litre in sea-water. For plutonium, the value of the distribution-factor K_D is approximately $(2\text{-}4).10^5$, depending on the oxidation state of plutonium; for americium, it is approximately $(2\text{-}4).10^6$ (Lovett, Nelson, 1978). A higher load of suspended matter in the water will cause a larger fraction of the elements to be associated with particulate matter. Using the values of distribution-factors (K_D) mentioned above, we estimated the content of particulate matter to be between 0.3 and 2 g of dry matter/m^3 of water.

It is interesting to compare the values of K_D for suspended particulate matter and water mentioned above with corresponding distribution factors for surface sediment and water. By using the values given in Tables 1 and 3, we have estimated the distribution factor for surface sediment (0-4 cm) and surface water to be $(0.2\text{-}0.5).10^5$ for plutonium and $(1\text{-}2).10^6$ for americium.

Even in sediments alone (Table 1) the integrated area-contents for plutonium and americium were often greater than $26 +/- 3$ Bq.m^{-2}, which was the value found in integrated fall-out on Svalbard from carpets of lichen and soil. This is unlikely to be due to drainage from land, but may be explained by fall-out plutonium transported to this area from latitudes with higher fall-out, the sediment acting as a sink for passing contaminated water.

The ratios between the activity concentration of americium and plutonium in the sediments were often higher than would be expected from integrated fall-out. This is in agreement with the more rapid settling and higher association to particulate matter for americium than for plutonium.

An estimation of the activity ratio between americium and plutonium in water plus sediment due to integrated fall-out shows that, due to the higher ratio in sediment and the lower ratio in water, the value of integrated the activity-ratio is about the same as on land on Svalbard, i.e. $0.37 +/- 0.05$.

The higher association of americium with particulate matter, and its rapid settling, indicate that americium in the Barents and Greenland Seas mainly originates from *in situ* build-up from ^{241}Pu. The isotopic composition of plutonium, such as ^{238}Pu and ^{241}Pu in relation to $^{239+240}Pu$, will indicate if other sources than fall-out are significant. This will be subject of further investigations.

Plutonium- and americium-concentration ratios to water were determined in *Fucus* and *Laminaria,* as seen from Table 2. Observed activity concentration ratios to water for *Fucus* based on dry weight were in the order of 8000 and 4000 for plutonium and americium respectively. In *Laminaria,* the ratios of activity concentrations were estimated to be 4000, both for plutonium and americium. These values are of the same order of magnitude as those found along the Norwegian coast (Christensen, 1982), but lower than those found in the Southern Baltic Sea (Nilsson *et al.,* 1981).

Table 3
^{137}Cs, $^{239+240}Pu$ concentrations and $^{241}Am/^{239+240}Pu$ activity ratios in deep water samples.

Depth (m)	Position	Salinity (‰)	^{137}Cs (mBq.1^{-1})	$^{239+240}Pu$ (µBq.1^{-1})	$^{241}Am/^{239+240}Pu$ (activity ratio)
250	80.1°N 30.0°E	34.70	21.1	17	0.13
400	81.4°N 40.0°E	34.87	17.8	17	—
500	78.8°N 0.1°E	34.91	7.8	23	0.24
500	79.6°N 4.7°E	34.92	7.4	26	—
520	82.4°N 45.5°E	34.87	14.8	19	—
800	81.7°N 9.1°E	34.91	4.8	6.0	—
1100	78.8°N 7.5°E	34.92	7.0	16	—
1100	82.4°N 45.1°E	34.93	5.6	14	0.28
2000	81.7°N 8.9°E	34.94	4.1	15	—
2700	81.7°N 29.8°E	34.95	3.0	2.2	—
5450	79.1°N 2.2°E	34.92	4.8	2.7	0.65

CONCLUSIONS

Some 25% of the cesium-137 present in water and sediments in the area studied is estimated to originate from fall-out specific to the area. Another 25% originates from mixing with Atlantic-water from latitudes with higher fall-out, and the remaining 50% is assumed to originate from European reprocessing facilities.

Plutonium and americium isotopes released by the European reprocessing facilities probably settle mainly in the local sediments (Nelson, Lovett, 1978) and are not, like cesium, transported up to the Barents and Greenland Seas. Fall-out plutonium, which is dominant in a soluble form, is transported from areas with higher fall-out levels and increases the activity concentration in water by a factor of two.

The levels of americium-241 in the Svalbard area are increased by *in situ* build-up due to the decay of plutonium-241.

Acknowledgements

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