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# Radioactivity Exploration from the Arctic to the Antarctic.

## Part 2. Ymer-80 Expedition

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### Abstract

Levels of natural radioactivity such as <sup>222</sup>Rn (radon) and its long-lived daughters <sup>210</sup>Pb and <sup>210</sup>Po were measured. The radon gas was trapped on cooled charcoal filters and the long-lived daughter products sampled on fibre filters on a daily basis. In addition, short-lived progenies were followed continuously on the filters in order to achieve a time resolution of about one hour. The average <sup>222</sup>Rn concentration in air measured during the Ymer-80 expedition in samples north of latitude 78.8 °N, was  $33 \pm 4$  (one standard error) mBq.m<sup>-3</sup> during Leg 1 (July-Aug.), and  $105.3 \pm 8.3$  mBq.m<sup>-3</sup> during leg 2 (Aug-Sept.). During a two-week period of persistent polar winds, the mean radon concentration decreased to  $19 \pm 5$  mBq.m<sup>-3</sup>.

During July, August and September, the monthly average concentrations of <sup>210</sup>Pb in air at positions north of 75 °N latitude were  $31 \pm 15$ ,  $89 \pm 61$  and  $105 \pm 57$  μBq.m<sup>-3</sup> respectively with a the grand average for all 3 months of  $75 \pm 28$  μBq.m<sup>-3</sup>.

An extensive radiochemistry program was also established to measure <sup>134+137</sup>Cs and the trans-uranium elements <sup>238+239+240</sup>Pu and <sup>241</sup>Am in water, sediment and biota.

The concentration of <sup>137</sup>Cs in surface seawater along the Norwegian coast was quite constant about  $157 \pm 7$  Bq.m<sup>-3</sup>. At a latitude about 72 °N it start to decrease exponentially at a rate of 0.43 deg.<sup>-1</sup> to about  $20 \pm 10$  Bq.m<sup>-3</sup> above 78 °N.

The results of <sup>137</sup>Cs in sediments indicate that the total integrated area-content of <sup>137</sup>Cs in a 1000 m water-column and sediment is about 12 kBq.m<sup>2</sup>.

In biota, the highest activity concentration of <sup>137</sup>Cs about 1000 Bq/kg<sub>dwt</sub> was found in lichens. The activity concentration of <sup>137</sup>Cs in polar bears was about 10 Bq/kg<sub>dwt</sub>, in seals about 1 Bq/kg<sub>dwt</sub>, and in birds about 1-7 Bq/kg<sub>dwt</sub>. The activity-concentration of <sup>137</sup>Cs in *Fucus* and *Laminaria* was about 1-2 Bq/kg<sub>dwt</sub>, and the algae/sea-water activity-concentration ratio was about 75.

The distribution of the trans-uranium element <sup>239+240</sup>Pu in sea water decrease from 14 to 10 mBq.m<sup>-3</sup> up to 73°N but increase again to 17 mBq.m<sup>-3</sup> at high latitudes while <sup>137</sup>Cs decrease.

The average <sup>241</sup>Am/<sup>239+240</sup>Pu activity ratio was found to be  $0.13 \pm 0.04$  (2 S.E), of 31 samples with the range 0.04 - 0.32 in surface water,

Deep-water samples have been sampled at latitudes around  $80.4 \pm 1.4$  °N and along longitudes 2° W – 45.5 °E. The results of the salinity and activity concentration of <sup>137</sup>Cs and <sup>239+240</sup>Pu with depth of water are indicate an exponential decrease with depth of both <sup>137</sup>Cs and <sup>239+240</sup>Pu.

In conclusion, about 25% of <sup>137</sup>Cs present in the Arctic water and sediments originates from fall-out specific to the area. Another 25% originates from mixing with Atlantic-water from latitudes with higher fall-out. The remaining 50% might originate from European reprocessing facilities.

The levels of <sup>241</sup>Am in the Svalbard area originate from *in situ* build-up due to the decay of <sup>241</sup>Pu.

## A. Introduction

The Arctic expedition “Ymer-80” was conducted during the period June 23-October 6 1980, to commemorate the discovery of the Northeast Passage by Adolf Erik Nordenskiöld in 1878-1880.



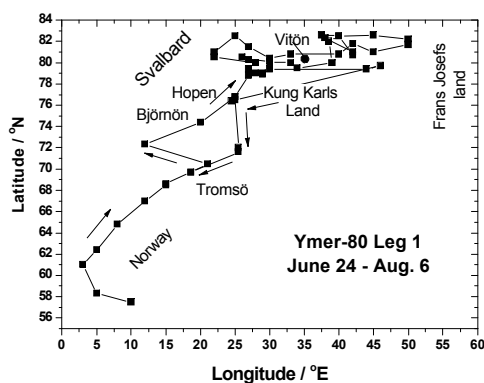
**Figure 2-1**  
The logo of Ymer-80 expedition

The expedition was conducted with Swedish state icebreaker HMS Ymer, which served both as transport and research platform. In the expedition participated 119 scientists and technicians from Sweden and from eight other countries. The first part of the expedition, “*Leg I*” (June 24-August 6, 1980), was focused primarily on oceanographic and biological studies. Glaciological and geomorphological research had previously been conducted on Nordaustlandet and a number of smaller islands around Svalbard, and some researchers were landed there to continue such studies.

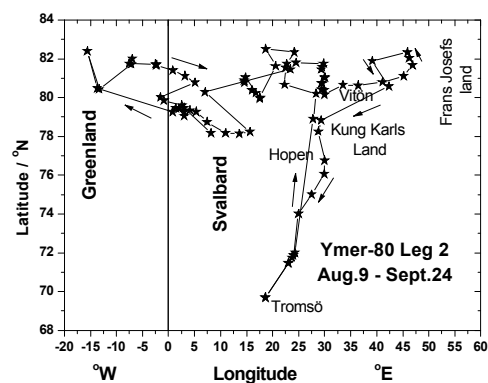


**Figure 2-2**  
The Swedish ice-breaker HMS Ymer

The expedition's second phase “*Leg 2*” (August 9 to September 24, 1980) concentrated on marine geology and geophysics in order to increase our knowledge of the Arctic seabed along the ship's long route from Tromsö to Spitsbergen, further to the north-eastern Greenland and then back to and around the western and northern Svalbard and around Frans Josef Land on the way back to Tromsö (see **Fig. 2-3**). Some of these studies involve seabed samples and seismic work and had a bearing on the contemporary discussion of the Antarctic seabed condition (Elg et al., 1981, Liljequist, 1993, Schytt, 1983, Sundman, 1982).



**Figure 2-3a**  
The route of the Ymer-80 expedition Leg 1.



**Figure 2-3b**  
The route of the Ymer-80 expedition Leg 2.

By initiative of Professor Bengt Forkman at the Nuclear Physics department, a group of scientists from Lund University became engaged in the Ymer-80 expedition studying the radioecology and radiation environment in the Arctic. The aim of this program was to investigate present levels and sources of natural and artificial radioactivity in the Arctic marine environment, and to study the pathways and distribution of the radionuclides in different compartments. Apart from caesium and plutonium isotopes, we also investigated natural actinides, such as uranium and thorium and their daughters in water (Holm et al., 1983).

An extensive air chemistry program was also established in which levels of  $^{222}\text{Rn}$  (radon) and its long-lived daughters  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  were measured. The radon gas was trapped on cooled charcoal filters and the long-lived daughter products sampled on fibre filters on a daily basis. In addition, short-lived progenies were followed continuously on the filters in order to achieve a time resolution of about one hour. In the Arctic summer air north of latitude  $75^\circ\text{N}$  the average activity concentrations of  $^{222}\text{Rn}$  was  $75 \pm 21 \text{ Bq}\cdot\text{m}^{-3}$  and of  $^{210}\text{Pb}$   $75 \pm 28 \mu\text{Bq}\cdot\text{m}^{-3}$ . During a two-week period of persistent polar winds, the mean radon concentration decreased to  $19 \pm 5 \text{ mBq}\cdot\text{m}^{-3}$ . The concentrations of  $^{222}\text{Rn}$  radon in Arctic air depends on exhalation from the sea, contribution by winds from the land masses may, however, contribute significantly to the measured radon concentration. It is shown that steady-state equilibrium models, applied to an air mass over the sea, overestimate the aerosol residence-time calculated from activity ratios. Time-dependent calculations indicate a mean aerosol residence time of 4 to 7 d in Arctic air. Good agreement is observed between radon levels and the time since the air mass left larger land areas. Both the in the  $^{222}\text{Rn}$  and the long-lived daughter measurements are insensitive to contamination from ship and local settlements (Samuelsson et al., 1986).

## **B. $^{222}\text{Rn}$ and $^{210}\text{Pb}$ in the Arctic Air**

Since only a few measurements of  $^{222}\text{Rn}$  and  $^{210}\text{Pb}$  in Arctic air have been performed made it was decide that measurements of these radionuclides would be a part of the extensive air chemistry programme of the Ymer-80 expedition. In the summer of 1978 at Barrow, Alaska, the  $^{210}\text{Po}$  air concentration of  $100 \mu\text{Bq}\cdot\text{m}^{-3}$  has been reported (Rahn and McCaffrey, 1979). During 1961-62 the corresponding value at Thule, Greenland was reported to be about  $200 \mu\text{Bq}\cdot\text{m}^{-3}$  (Patterson jr and Lockhart jr, 1964). In 1979 on the islands of Amchitka, Alaska ( $51^\circ\text{N}$ ,  $55^\circ\text{E}$ ), that correspond to a maritime Arctic location, the air concentration of  $^{210}\text{Po}$  was only  $2 \mu\text{Bq}\cdot\text{m}^{-3}$  (Nevissi and Schell jr, 1980). Low values of  $^{210}\text{Po}$  in the range of  $2\text{-}73 \mu\text{Bq}\cdot\text{m}^{-3}$  has been reported from measurements on Franz Josef Land (Jaworowski, 1969). During 1953-1959 the  $^{222}\text{Rn}$  concentrations has been measured in Alaska. In summer at Kodiak Island ( $57^\circ45' \text{N}$ ,  $152^\circ29' \text{W}$ ) and Wales ( $65^\circ37' \text{N}$ ,  $168^\circ03' \text{W}$ ) the mean values were about  $200 \text{ mBq}\cdot\text{m}^{-3}$ . and  $400 \text{ mBq}\cdot\text{m}^{-3}$ . respectively (Lockhart jr, 1962)).

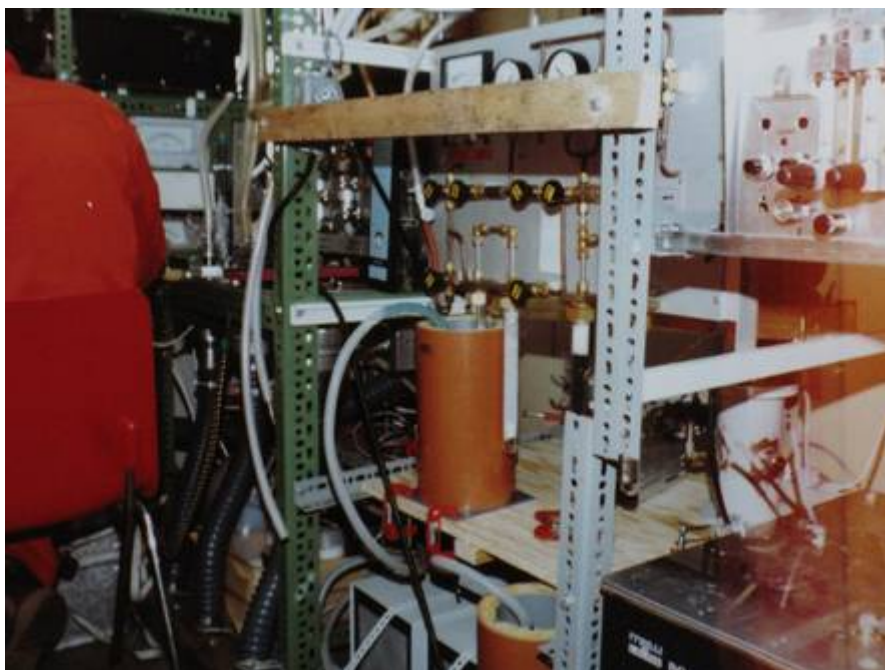
No measurements of  $^{222}\text{Rn}$  in air has previously been carried out in the Arctic maritime regions. Thus, the Ymer-80 expedition offered an unique possibility to measure  $^{222}\text{Rn}$  in the air along its



route in the Arctic.

### **B1. Measurements of $^{222}\text{Rn}$ in surface air**

In order to measure the low radon concentrations anticipated, the air was flowing through a cooled ( $-14^{\circ}\text{C}$ ) charcoal trap (Picatif G210, Pica, France) which effectively adsorb the radon gas. A commercial radon concentrator (RCTS-2, Johnston Lab., Cockeysville, Maryland, USA) with minor modifications was be used. By heating the charcoal trap to about  $380\text{-}400^{\circ}\text{C}$ , and flushing with helium gas, the radon was transferred to vials with ZnS on the walls (LAC-II, Johnston Lab., USA). The alpha particles emitted from the decay of  $^{222}\text{Rn}$  caused emission of scintillating pulses from the ZnS. These pulses were recorded during approximately 12 h, by placing the vial on a PM-tube connected to pulse-counting electronics.



**Figure 2-4**  
The cooled charcoal trap system for collecting  $^{222}\text{Rn}$  in the air mounted on board Ymer (Picatif G210, Pica, France)

### **B2. Measurements of $^{222}\text{Rn}$ decay products in the air**

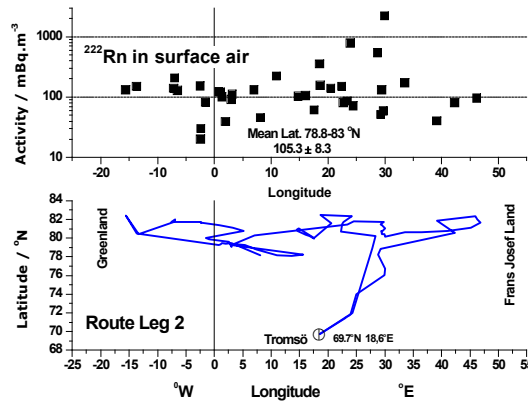
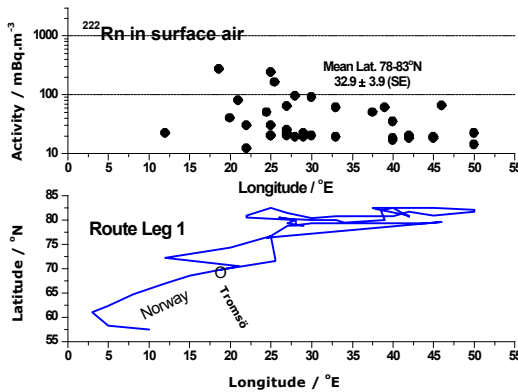
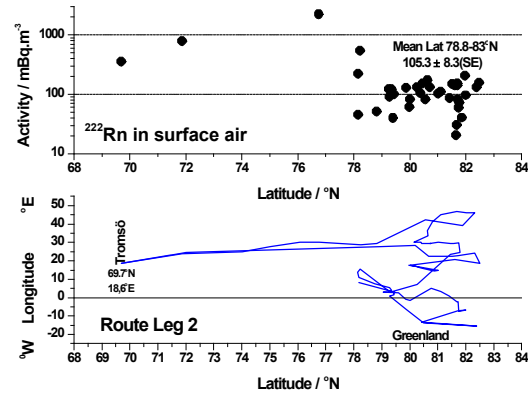
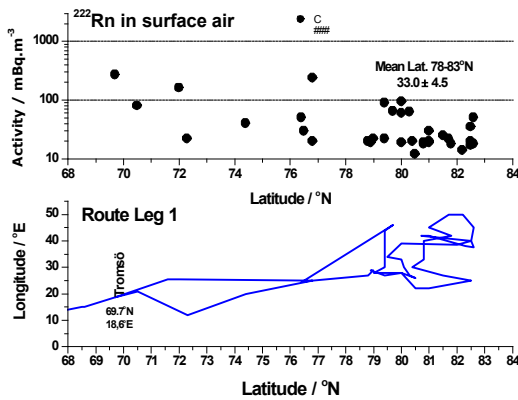
Radon-222 diffuses partly from the earth's crust to the atmosphere where its concentration decreases monotonically by height.  $^{222}\text{Rn}$  decays with a half-life of 3.82 days to the following short lived radon daughters:  $^{222}\text{Rn}$  (3.82 days)  $>$   $^{218}\text{Po}$  (RaA 3.10 min)  $>$   $^{214}\text{Pb}$  (RaB 26.8 min)  $>$   $^{214}\text{Bi}$  (RaC 19.9 min)  $>$   $^{214}\text{Po}$  (RaC' 164.3 ms)  $>$   $^{210}\text{Pb}$  (RaD 22.20 a)  $>$   $^{210}\text{Bi}$  (RaE 5.01 d)  $>$   $^{210}\text{Po}$  (RaF 138.4 d)  $>$   $^{206}\text{Pb}$  (stable). In the atmosphere, the decay products from  $^{222}\text{Rn}$  attach to airborne particles and deposit as dry and wet deposition onto the earth's surface. The decay products

following  $^{214}\text{Po}$  are the long-lived  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$ ,  $^{210}\text{Po}$ , and finally stable Lead-206.

For sampling of the long-lived radon daughters, we used an alpha-in-air monitor with a ruggedized surface-barrier detector (Alpha-3, Eberline, Santa Fe, New Mexico, USA). The air was continuously sampled at a rate of about  $2.3 \text{ m}^3\text{h}^{-1}$  onto membrane filters (SM  $5 \mu\text{m}$ , Sartorius, W. Germany). The filters were changed every 24 h and stored for later analysis of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  at our laboratory in Lund.

### B.3 Results of $^{222}\text{Rn}$ surface air concentration

The  $^{222}\text{Rn}$  concentrations of surface air during the Ymer-80 expedition is shown in **Figure 2-5a and b** (Samuelsson et al., 1986).



**Figure 2-5a**

Radon concentrations in surface air along the route of Leg 1 July-Aug. (Samuelsson et al., 1986).

**Figure 2-5b**

Radon concentrations in surface air along the route of Leg 2 Aug. - Sept. (Samuelsson et al., 1986).

During an extended period in July, the air mass reaching the ship always came from the North Polar area. The radon concentrations were remarkably constant, with a mean value of  $21 \pm 0.24$



(one standard error)  $\text{mBq.m}^{-3}$ .

The average radon concentration for the whole duration of the 'Ymer' expedition (all samples north of latitude  $78.8^\circ$ ) is  $33 \pm 4$  (one standard error)  $\text{mBq.m}^{-3}$  during Leg 1 (July-Aug.), and  $105.3 \pm 8.3$   $\text{mBq.m}^{-3}$  during leg 2 (Aug-Sept.).

#### **B4. Results of Radon daughters in the air**

During July, August and September, the monthly average concentrations of  $^{210}\text{Pb}$  in air at positions north of  $75^\circ\text{N}$  latitude were  $31 \pm 15$ ,  $89 \pm 61$  and  $105 \pm 57$   $\mu\text{Bq.m}^{-3}$  respectively with a the grand average for all 3 months of  $75 \pm 28$   $\mu\text{Bq.m}^{-3}$  (Samuelsson et al., 1986). The values are in agreement with the annual mean value for the Arctic region previous estimated to about  $140$   $\mu\text{Bq.m}^{-3}$  (Jaworowski, 1969).

### **C. Cesium-137 measurements**

#### **C1. $^{137}\text{Cs}$ in surface water**

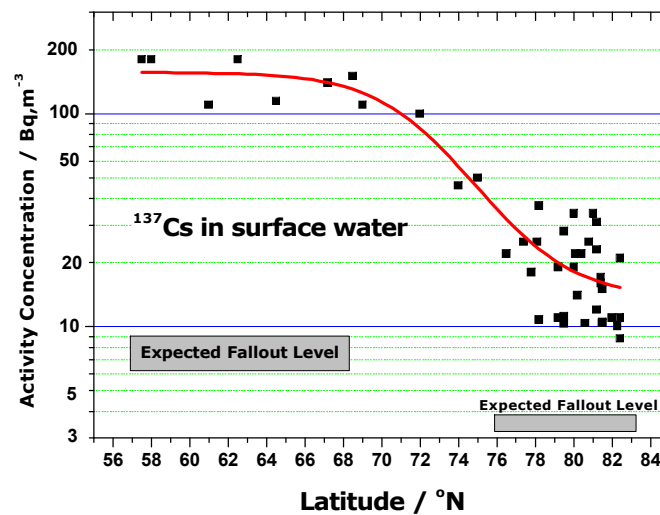
Seawater samples of 100-200 litre were pumped from an inlet at 7 m depth, and collected in special precipitation vessels located at the front deck of the ship (**Fig 2-6**). Caesium was separated by adsorption on to microcrystalline ammonium-molybdo-phosphate after  $^{134}\text{Cs}$  had been added to the sample as a radiochemical yield determinant. Larger volumes of sea-water (1 000-2 000 l) from 7 m depth were filtered through cartridge-filters (Millipore CWSS 012C3) in order to determine the fraction of the investigated radionuclides associated with particulate matter.

The activity concentration of  $^{137}\text{Cs}$  in water samples collected between  $57^\circ\text{N}$  to  $82.8^\circ\text{E}$  is displayed in **Figure 2-7** as a function of latitude. Along the Norwegian coast the concentration of  $^{137}\text{Cs}$  in surface seawater was quite constant about  $157 \pm 7$   $\text{Bq.m}^{-3}$ . At a latitude about  $72^\circ\text{N}$  it start do decrease exponentially at a rate of  $0.43^\circ\text{N}^{-1}$  to about  $20 \pm 10$   $\text{Bq.m}^{-3}$  above  $78^\circ\text{N}$ . It is obvious that the concentration of  $^{137}\text{Cs}$  found along the Norwegian coast is much higher than expected from nuclear weapon fallout. This is explained by the fact that  $^{137}\text{Cs}$  released from European nuclear fuel reprocessing facilities is spread by the Gulf Stream along the Norwegian coast. Other investigations on the distribution of  $^{137}\text{Cs}$  activity concentration in the North Sea and adjacent areas also confirm our results (Kautsky, 1980). The  $^{137}\text{Cs}$  activity concentration in area water also decrease rapidly with increasing distance from the coast (Kautsky, 1980, Aarkrog et al., 1982). Along the Norwegian coast from  $57.8^\circ\text{N}$ ,  $8.1^\circ\text{E}$  to  $69.0^\circ\text{N}$ ,  $14.4^\circ\text{E}$ , the  $^{137}\text{Cs}$  activity concentration in the surface sea-water decreases by less than a factor of two.



**Figure 2-6**

Sea-water samples of 100-200 l were pumped from an inlet of the ship at 7 m depth and collected in two special precipitation 200 l vessels placed, one for precipitation  $^{137}\text{Cs}$  and another for trans-uranium elements. Bertil Person is holding a cartridge-filter (Millipore CWSS 012C3) to be placed in the holder on the wall.



**Figure 2-7**

The lower diagram shows the activity concentration of  $^{137}\text{Cs}$  in water samples collected between  $57^\circ\text{N}$  to  $82.8^\circ\text{E}$  as a function of latitude. The upper diagram shows the sampling sites.

The  $^{137}\text{Cs}$  activity concentration varies with the latitude as a Boltzmann sigmoid equation displayed in **Figure 2-7**.

$$A = \frac{A_1 A_2}{A_1 + A_2 \exp(-k_{\text{Lat}}(\text{Lat}_m - \text{Lat}))} + A_2 \quad [\text{Bq.m}^{-3}]$$

where

- A is the activity concentration of  $^{137}\text{Cs}$  in sea water  $\text{Bq.m}^{-3}$  at various latitudes
- A1 is the minimum activity concentration at high latitudes
- A2 is the minimum activity at low latitudes
- Lat<sub>m</sub> is the latitude at the median activity value  $(A_1+A_2)/2$

The values of the parameters of the fitted curve in **Figure 2-7** are given below:

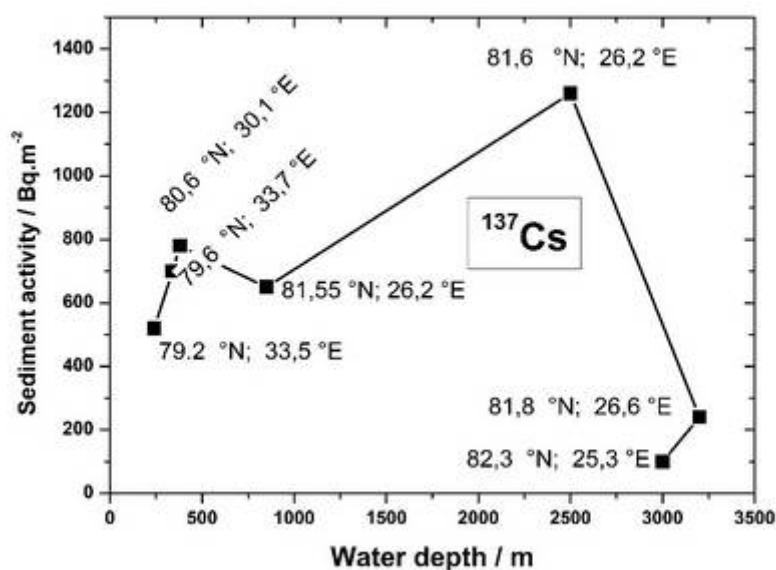
Parameter	Value	SD
A1	156.7	7.3
A2	13.7	4.6
Lat <sub>m</sub>	72.0	0.7
k <sub>Lat</sub>	0.427	0.096
A2/A1	0.087	0.030

The dilution factor of the Atlantic water flowing into the Arctic Ocean estimated by the ratio of A1/A2 is 11.5.

The activity ratio of  $^{134}\text{Cs} / ^{137}\text{Cs}$  was measured at locations between latitudes of 59 -77 °N. Since  $^{134}\text{Cs}$  is an activation product, it should not be present in fallout from nuclear weapons tests. This ratio found was in the order of 0.03 - 0.04, which indicate that it originates from the nuclear fuel processing plant at Sellafield in UK (formerly Windscale). (Kershaw and Baxter, 1995).

## C2. $^{137}\text{Cs}$ in sediments

Caesium isotopes in sediment have so far only been measured in samples from some of our sampling sites. The results obtained are given in **Figure 2-8** are estimated from the few deep water, that vary with the widely spread sites of the sample stations. The results given in **Figure 2-8** indicate that the total integrated area-content of  $^{137}\text{Cs}$  in a 1000 m water-column and sediment is about 12  $\text{kBq.m}^{-2}$ . This value exceeds the integrated  $^{137}\text{Cs}$  area-content of  $2.2 \pm 0.3 \text{ kBq.m}^{-2}$  on Svalbard (78.2°, 16.0°E) which originates from air borne fallout.



**Figure 2-8**

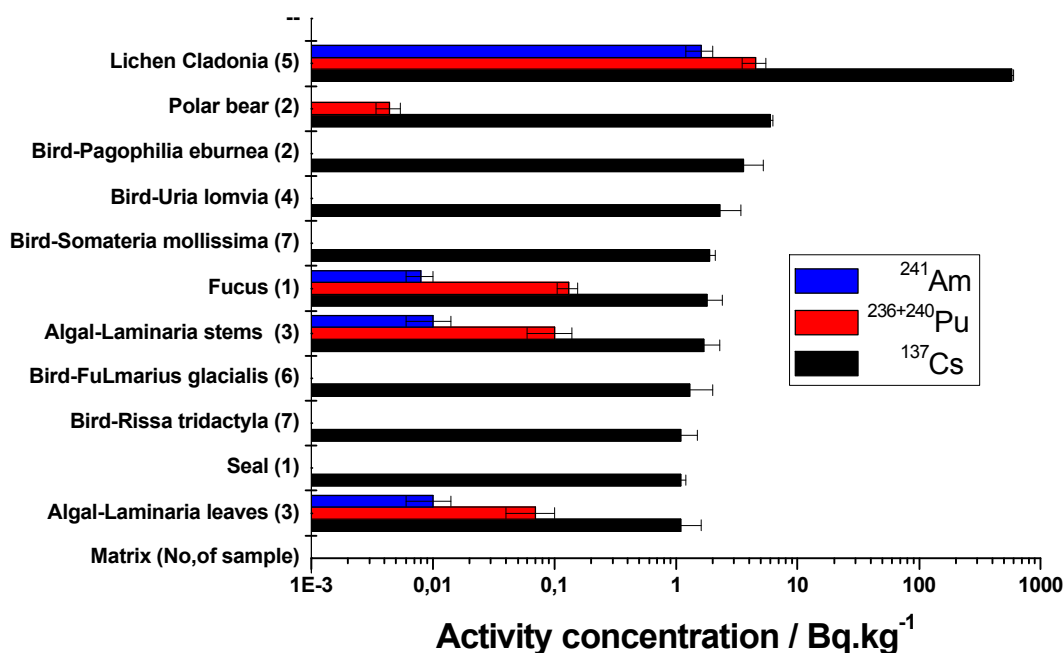
The integrated area content in sediments of  $^{137}\text{Cs}$  versus depth of water at corresponding sampling site (Lat.;Long.)

The amount of  $^{137}\text{Cs}$  associated with particulate matter was as little as of the order of  $4 \cdot 10^{-5}$  along the Norwegian coast and  $(0.1-2) \cdot 10^{-2}$  in the Barents and Greenland Seas. It might be expected that the higher value along the Norwegian coast is due to  $^{137}\text{Cs}$  associated with particles in the drainage from land. The contribution from this run-off, is small compared to the amount in soluble form originating from reprocessing plants and carried by currents of water.

### C3. $^{137}\text{Cs}$ in biota

The results for biological samples are presented in **Figure 2-9**. The highest activity concentration of  $^{137}\text{Cs}$  about  $1000 \text{ Bq/kg}_{\text{dwt}}$  was found in lichens. The activity concentration of  $^{137}\text{Cs}$  in polar bears was about  $10 \text{ Bq/kg}_{\text{dwt}}$ , in seals about  $1 \text{ Bq/kg}_{\text{dwt}}$ , and in birds about  $1-7 \text{ Bq/kg}_{\text{dwt}}$ . The activity-concentrations of  $^{137}\text{Cs}$  in the flesh of polar-bears, seals and birds agree with those found at various places in Greenland (Aarkrog, 1979).

The activity-concentration of  $^{137}\text{Cs}$  in *Fucus* and *Laminaria* was about  $1-2 \text{ Bq/kg}_{\text{dwt}}$ , and the algae/sea-water activity-concentration ratio was about 75. Along the Norwegian coast about the same algae/seawater activity concentration ratio was found in *Fucus vesiculosus* (Christensen, 1982).



**Figure 2-9**

Activity concentration (Bq per kg of dry mass, Bq/kg<sub>dwt</sub>) of <sup>137</sup>Cs, <sup>238+240</sup>Pu and <sup>241</sup>Am in various biological samples collected during August-September 1980. The birds were collected at Kinnvika. Seal and Polar bears collected in Barents sea. Algae and Lichen collected on Svalbard.

## D. Plutonium-239+240 and Americium-241

### D1. Trans-Uranium elements in Sea water

The distribution of the trans-uranium elements <sup>239+240</sup>Pu and <sup>241</sup>Am in the surface water of the Norwegian, Barents and Greenland Seas is quite different from that of <sup>137</sup>Cs. The distribution of the transuranium elements displayed in **Figure 2-10** decrease from 14 to 10 mBq.m<sup>-3</sup> up to 73°N but increase again to 17 mBq.m<sup>-3</sup> at high latitudes while <sup>137</sup>Cs decrease. 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 of the Irish Sea (Lovett and Nelson, 1978).

The activity-concentrations of plutonium-isotopes are higher than would be expected from global fall-out in the Barents and Greenland Seas, which indicate local source. The levels found here are equal to those found in Atlantic seawater 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 (Fukai *et al.*, 1981).

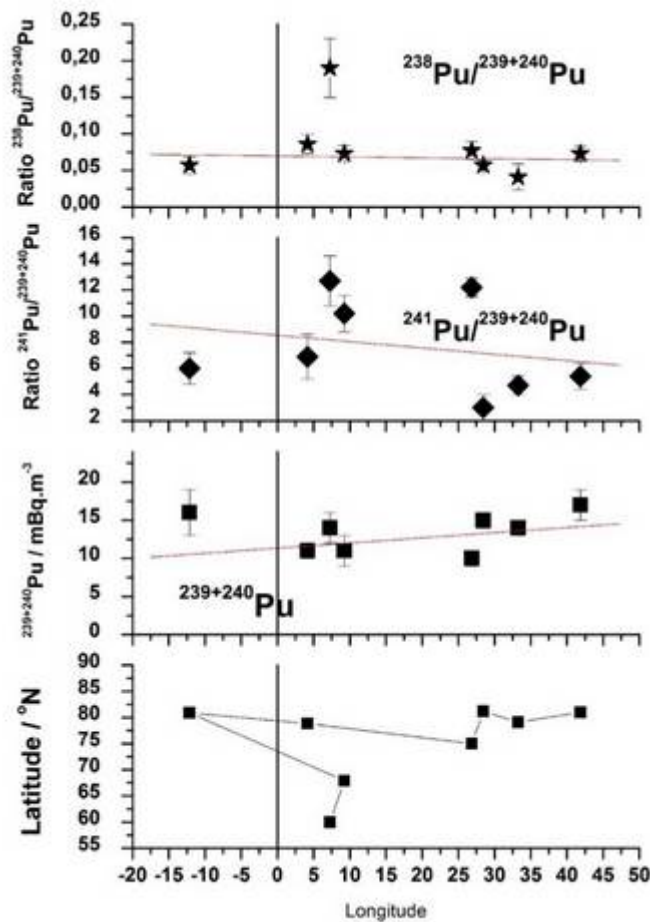
The average  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratio was found to be  $0.13 \pm 0.04$  (2 S.E) of 31 samples with the range 0.04-0.32) in surface water, which is lower than the value  $0.37 \pm 0.05$  found for integrated fall-out on Svalbard. The corresponding activity-ratio in the residue on the cartridge filter sample was  $0.32 \pm 0.09$  (2 S.E.) of 40 samples with the range 0.06-1.3). We estimated that 32% of americium in seawater 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.

The results of the activity ratios of  $^{238}\text{Pu}$  and  $^{241}\text{Pu}$  relative to  $^{239+240}\text{Pu}$  has been pooled for different areas displayed in **Table 2-3** where sampling took place during the Ymer-80 expedition.

**Table 2-3** Average positions of the different sampling areas during the Ymer-90 expedition

Area	Latitude +°N	Longitude +°E -°W	Latitude +°N	Longitude +°E -°W
A	79,5	37,7	82,5	46
B	78,8	29,2	79,4	37,3
C	80,1	22,9	82,3	34
D	71,8	23,7	78,2	30
E	78,2	0,2	79,5	8,1
F	79,5	-15,4	82,3	-8,9
G	64,6	4,3	71,2	14,2
H	57,1	3	62,9	11,5

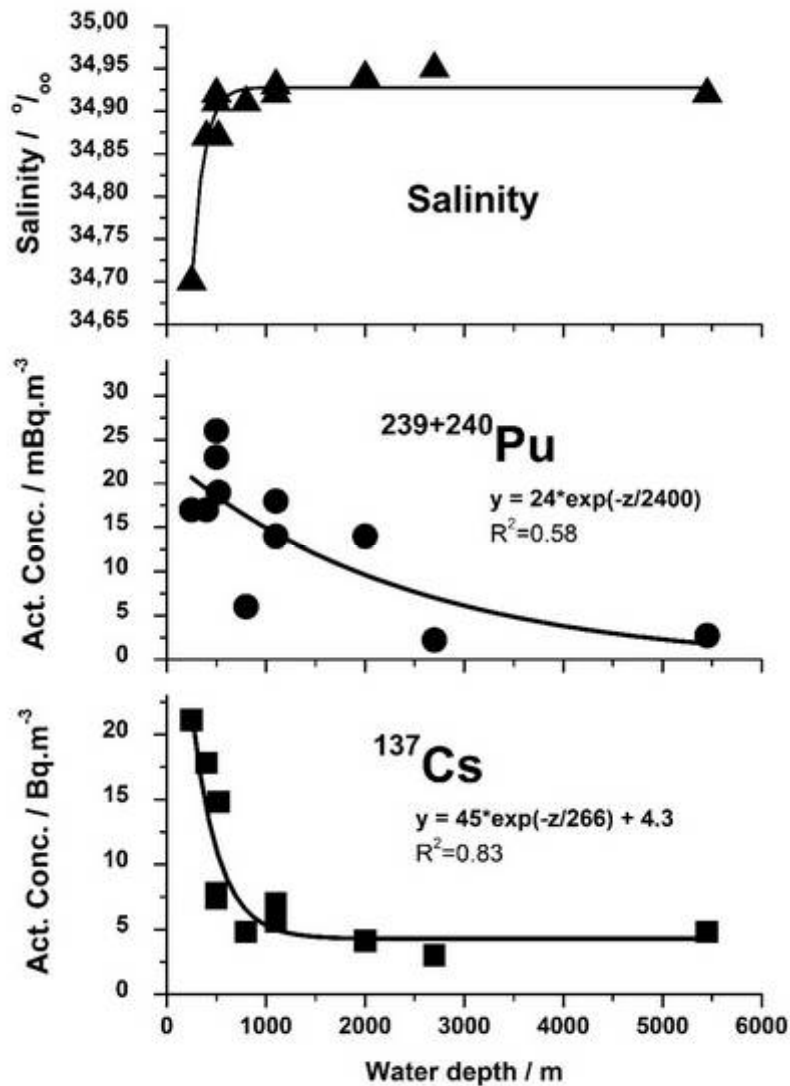




**Figure 2-10**  
Longitudinal distribution of plutonium isotopes water

Deepwater samples were collected with a 100 l Niskin bottle at latitudes around  $80.4 \pm 1,4$  °N and along longitudes  $2^{\circ}$  W –  $45.5$  °E. The results of the salinity and activity concentration of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  with depth of water are shown in **Figure 2-11**. The results indicate an exponential decrease with depth of both  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$ . For  $^{239+240}\text{Pu}$ , however, a sub surface maximum is indicated in agreement with previous observations (Fukai et al., 1979, Livingston and Bowen, 1976).

The activity-ratio of  $^{241}\text{Am}/^{239+240}\text{Pu}$  indicate an increase with depth and the  $^{241}\text{Am}$ -activity concentration in surface water shows no significant correlation with the latitude. This indicates that  $^{241}\text{Am}$  released from reprocessing plants is not transported over long distances. Americium from fallout is stronger associated, with particulate matter than plutonium, and is therefore deposited locally (Holm et al., 1980).



**Figure 2-11**

The variation of salinity and activity concentration of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  with depth of water at latitude  $80.4 \pm 1.4$  °N and longitude  $2^\circ\text{W}$ ;  $45.5$  °E

## D2. Trans-Uranium elements in *Fucus* and *Laminaria*, lichen and moss

The highest activity concentration of  $^{238+240}\text{Pu}$  and  $^{241}\text{Am}$  6 and 2  $\text{Bq}/\text{kg}_{\text{dwt}}$  respectively was found in lichens. The activity concentration of  $^{238+240}\text{Pu}$  in polar bears was about  $4 \text{ mBq}/\text{kg}_{\text{dwt}}$ . The activity-concentration of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  in the algae *Fucus* and *Laminaria* was about 100-10  $\text{Bq}/\text{kg}_{\text{dwt}}$  respectively. Plutonium- and americium-concentration ratios to water were determined in *Fucus* and *Laminaria*, as seen from **Table 2-4**.

Observed activity concentration ratios to water for *Fucus* based on dry weight were in the order of 8 000 and 4 000 for plutonium and americium respectively. In *Laminaria*, the ratios of activity concentrations were estimated to be 4 000, both for plutonium and americium. These values are of the same order of magnitude as those found along the Norwegian coast, but lower than those found in the Southern Baltic Sea (Nilsson et al., 1981, Christensen, 1982).

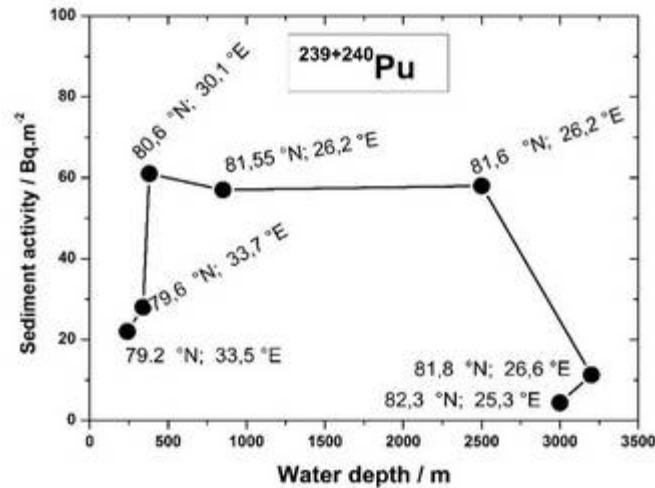
**Table 2-4**

Plutonium-234+240 activity concentration and ratios of  $^{241}\text{Pu}$  and  $^{238}\text{Pu}$  in sediments and biota samples collected during the Ymer80-expedition (Holm et al., 1986)

Place of collection (Water depth) m	Sediment depth cm	Number of Samples	$^{239+240}\text{Pu}$ Activity conc. mBq/kg $\pm$ SD		$^{241}\text{Pu}$ $/^{239+240}\text{Pu}$ Ratio $\pm$ SD		$^{238}\text{Pu}$ $/^{239+240}\text{Pu}$ Ratio $\pm$ SD	
79.2-82.3°N (240-3000)	0-4	10	430	100	4,8	0,8	0,069	0,007
25.3-33.7°E (240-3000)	4-8	7	100	30	3,5	1,1	0,52	0,018
Place of collection	Species	Number of Samples	$^{239+240}\text{Pu}$ Activity conc. mBq/kg $\pm$ SD		$^{241}\text{Pu}$ $/^{239+240}\text{Pu}$ Ratio $\pm$ SD		$^{238}\text{Pu}$ $/^{239+240}\text{Pu}$ Ratio $\pm$ SD	
Svalbard	<i>Larminaria</i> + <i>Fucus</i>	10	90	23	5.8	0.9	0.04	0.005
NE Greenland	<i>Laminaria</i>		1260	630	4.7	0.7	0.05	0.005
Svalbard Isfjord	Lichen ( <i>Cladonia</i> )	6	4200	1100	3.7	0.8	0.061	0.006
Svalbard Isfjord	Lichen ( <i>Cladonia</i> )	2	6600	1300	3.7	0.7	0.044	0.005
NE Greenland	Lichen	1	6900	700	4.2	0.6	0.05	0.006
Svalbard Storöja	Moss	1	11700	1200	4.7	0.9	0.04	0.005
Svalbard Isfjord	Soil (-10 cm)	2	300	30	4.3	0.9	0.042	0.01
Average Ratios (2SE)					4.1	0.4	0.047	0.008

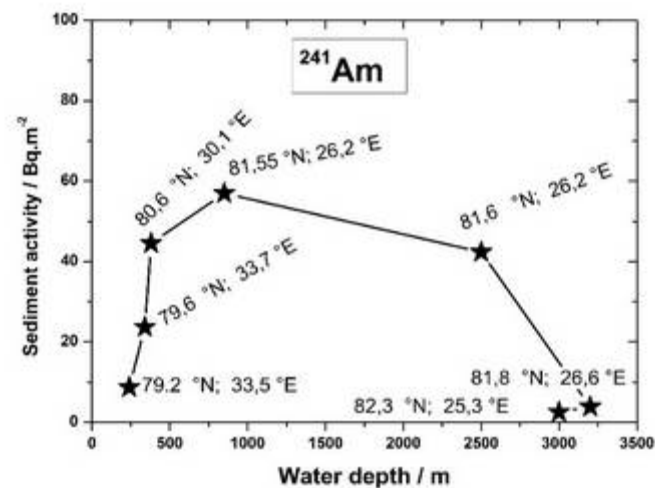
### D3. Trans-Uranium elements in Sediments

As seen in **Figures 2-12 and 2-13** the integrated area contents in sediments for plutonium and americium respectively are shown. In sediments the contents were often greater than was the values about  $26 \pm 3 \text{ Bq}\cdot\text{m}^{-2}$  found in integrated fall-out on Svalbard from carpets of lichen and soil. This is unlikely to be due to drainage from land. It may, however, 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.



**Figure 2-12**

The integrated area content in sediments of <sup>239+240</sup>Pu versus depth of water at corresponding sampling site (Lat.;Long.)



**Figure 2-13**

The integrated area content in sediments of <sup>241</sup>Am versus depth of water at corresponding sampling site (Lat.;Long.).

An estimation of the activity ratio between americium 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.  $0.37 \pm 0.05$ . 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  $^{241}\text{Pu}$ . The isotopic composition of plutonium such as  $^{238}\text{Pu}$  and  $^{241}\text{Pu}$  in relation to  $^{239+240}\text{Pu}$  will indicate if other sources than fall-out are significant.

A mean activity ratio of  $^{238}\text{Pu}/^{239+240}\text{Pu}$   $0.047 \pm 0.008$  may be found in samples from Greenland and Svalbard which only contaminated by  $^{238}\text{Pu}$  from fallout only. The mean of all water samples analysed for  $^{238}\text{Pu}$  and sediment, give an activity ratio of  $0.060 \pm 0.010$  (2 S.E.,  $n = 15$ ) from which it can be calculated that between 30 to 50 %, (depending on which of the figures are used for the activity ratio in fallout),  $^{238}\text{Pu}$  in Barents and Greenland Seas originate from European reprocessing facilities. The Activity concentration of  $^{238}\text{Pu}$  originating from these facilities is between 0.20 and 0.38  $\text{mBq}\cdot\text{m}^{-3}$ , and the corresponding value for  $^{241}\text{Pu}$  is in the range of 16 -39  $\text{mBq}\cdot\text{m}^{-3}$ . The activity contribution in these waters of  $^{239+240}\text{Pu}$  from European reprocessing facilities can thus be estimated to about 0.6-1.4  $\text{mBq}\cdot\text{m}^{-3}$ , which is equivalent to between 5 and 10% of the  $^{239+240}\text{Pu}$  in the Barents and Greenland Seas. The Plutonium release from Sellafield in the oxidation state of Pu(IV) that sediment fast, while Plutonium from nuclear weapons fallout is in oxidation state Pu(V) that is more soluble as carbonate in seawater. Thus the main part of Pu-isotopes in the arctic Ocean originate from nuclear weapons fallout and only a minor part from Sellafield (Holm et al., 1986).

## E. CONCLUSIONS

The average  $^{222}\text{Rn}$  concentration in air measured during the Ymer-80 expedition in samples north of latitude  $78.8^\circ\text{N}$ , was  $33 \pm 4$  (one standard error)  $\text{mBq}\cdot\text{m}^{-3}$  during Leg 1 (July-Aug.), and  $105.3 \pm 8.3$   $\text{mBq}\cdot\text{m}^{-3}$  during leg 2 (Aug-Sept.). During a two-week period of persistent polar winds, the mean radon concentration decreased to  $19 \pm 5$   $\text{mBq}\cdot\text{m}^{-3}$ . During July, August and September, the monthly average concentrations of  $^{210}\text{Pb}$  in air at positions north of  $75^\circ\text{N}$  latitude were  $31 \pm 15$ ,  $89 \pm 61$  and  $105 \pm 57$   $\mu\text{Bq}\cdot\text{m}^{-3}$  respectively with a the grand average for all 3 months of  $75 \pm 28$   $\mu\text{Bq}\cdot\text{m}^{-3}$ .

The concentration of  $^{137}\text{Cs}$  in surface seawater along the Norwegian coast was quite constant about  $157 \pm 7$   $\text{Bq}\cdot\text{m}^{-3}$ . At a latitude about  $72^\circ\text{N}$  it start to decrease exponentially at a rate of  $0.43$   $\text{deg}^{-1}$  to about  $20 \pm 10$   $\text{Bq}\cdot\text{m}^{-3}$  above  $78^\circ\text{N}$ . The results of  $^{137}\text{Cs}$  in sediments indicate that the total integrated area-content of  $^{137}\text{Cs}$  in a 1000 m water-column and sediment is about 12  $\text{kBq}\cdot\text{m}^{-2}$ . About 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 by inflow of Atlantic water from latitudes with higher fall-out. The remaining 50% is assumed to originate from inflow of Atlantic water transporting the release from European nuclear-fuel reprocessing facilities. The highest activity concentration of  $^{137}\text{Cs}$  about 1000  $\text{Bq}/\text{kg}_{\text{dwt}}$  was found in lichens collected on Svalbard. The activity concentration of  $^{137}\text{Cs}$  in flesh of polar bears was about 10  $\text{Bq}/\text{kg}_{\text{dwt}}$ , in seals about 1  $\text{Bq}/\text{kg}_{\text{dwt}}$ , and in birds about 1-7  $\text{Bq}/\text{kg}_{\text{dwt}}$ . The activity-concentration of  $^{137}\text{Cs}$  in

the algae *Fucus* and *Laminaria* was about 1-2 Bq/kg<sub>dwt</sub>, and the algae/sea-water activity-concentration ratio was about 75

The distribution of the trans-uranium element  $^{239+240}\text{Pu}$  in seawater decrease from 14 to 10 mBq.m<sup>-3</sup> up to 73°N but increase again to 17 mBq.m<sup>-3</sup> at high latitudes while  $^{137}\text{Cs}$  decrease. Plutonium and Americium isotopes released by the European reprocessing facilities probably settle mainly in the local sediments in the Irish Sea and are not as Caesium, transported up to the Barents and Greenland Seas (Nelson and Lovett, 1978). Fall-out plutonium, however, which is mainly in a soluble form, is transported from areas with high 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. The average  $^{241}\text{Am}/^{239+240}\text{Pu}$  activity ratio was found to be  $0.13 \pm 0.04$  (2 S.E), of 31 samples with the range 0.04-0.32 in surface water,

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