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Radioactivity Exploration from the Arctic to the Antarctic

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Radioactivity Exploration from the Arctic to the Antarctic

Bertil RR Persson, Elis Holm, and Kjell-Åke Carlsson



Lund 2015

*This book is dedicated to
professor emeritus Bengt Forkman
(Nuclear Physics, Lund University)
who in 1979 took the initiative to arrange an environmental radioactivity
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Radioactivity exploration from the Arctic to the Antarctic.

Chapter 1. Introduction

Bertil R.R. Persson, Elis Holm, and Kjell-Åke Carlsson

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This book is dedicated to professor emeritus Bengt Forkman (Nuclear Physics, Lund University) who in 1979 took the initiative to arrange an environmental radioactivity research program for the Ymer-80 expedition. He engaged his old friend Bertil Persson whom he during the 1960th inspired to university studies in Lund who just been promoted to professor of Radioecology at the Swedish University of Agricultural Sciences in Uppsala. Bertil Persson was tutor for Elis Holm at Lund University, who for his thesis was engaged in radiochemical analysis of plutonium isotopes in the environment. Elis Holm became deeply involved in management of Ymer-80 and all the following expeditions as well. We found a superior talent for the logistics in Kjell-Åke Carlsson (who was mechanical engineer at the department of radiation physics at Lund University). Without him, we would not have been able to solve all the thousands of practical issues and contacts with authorities and sponsors. He also contributed with the diaries extensive photographic and video documentation of all the expeditions. We have together compiled this book although several others were partly engaged in the various expeditions to whom we are deeply thankful for their contributions. They will appear as contributors in the separate parts of this book:

1. Ymer-80 (Bengt Forkman, Boel Forkman, Lars Ahlgren (diseased))
2. Swedarp 1988-1989 (Per Roos, Birgitta Roos)
3. Arctic Ocean 1991
4. Tundra Expedition
5. Arctic Ocean 1996 (Dan Josefsson, Mats Ericsson)

Our first 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. The vessel used for the expedition was the Swedish icebreaker, M/S Ymer. The use of such a heavy icebreaker made areas permanently covered with ice accessible to extensive scientific activity by various research teams. The aim of our radio-ecological research was to investigate present levels and sources of both natural and man-made radioactivity in the Arctic air and marine environment, and to study the pathways and distribution of these radionuclides in different compartments. Apart from caesium and plutonium isotopes, we also investigated natural radioactive elements, such as uranium and thorium in water, as well as radon and radon daughters in the air (Holm et al., 1983, Samuelsson et al., 1986). In the Arctic water samples were collected between 57°N to 82.8 °E, and enhanced levels of ¹³⁷Cs was found along the Norwegian coast caused by ¹³⁷Cs released from European nuclear fuel

reprocessing facilities spread by the Gulf Stream along the Norwegian coast. We also found hot spot of plutonium in the Arctic Ocean. During the expedition, far out in the ice. Far out in the Arctic ice Bertil Persson received a call on short wave radio from the Chancellor of Lund University, Håkan Westling, who asked if he would accept promotion as professor in Medical radiation Physics and Head of Radiation Physics at Lund University Hospital. The answer was “Yes”, and so the exploration of environmental radioactivity proceeded.

The second expedition “Swedarp” took place during Nov 1988 to Feb 1989. The research platform was the ship, M/S Stena Arctica, with air-sampling device installed on board. We started from Gothenburg (67.4°N; 12°E) with the first destination Montevideo (34.8°S; 56.2°W). From Montevideo, we continued to the Swedish permanent base “Svea” at the North shelf of Antarctica. After unloading supply and equipment for the continental research group, the ship continued to the Argentinean base “Marambio”. The expedition members were allowed to visit “Paulet Island” with the remains of the stone-hut, built by the Swedish captain Carl Anton Larsen and his crew during 1903-04, after that their vessel "Antarctic" was shattered by the ice masses in the Weddell Sea and sank.

The third expedition in 1991 was to the Arctic Ocean with the Swedish icebreaker M/S Oden. The research program was focused on oceanography and geology in the western parts of the Eurasian Basin, the north west Markov Basin and parts of the Barents sea (Josefsson, 1998 , Roos et al., 1998). By using pumps of the ship, samples of surface-water, were collected and processed in 200 l vessels in our laboratory accommodated in a container on board.

The fourth expedition was the joint Swedish-Russian “Tundra Ecology-94” expedition during 1994 with the Russian ice-breaking research vessel R/V Akademik Fedorov a platform, along a coastline of 3500 km-from the Kola Peninsula 10°E to Kolyuchinskaya Bay 173°E. Air sampling was performed during the route along the Norwegian and North Siberian coastlines and water samples were collected from the vessels cooling water system. Continuous sampling of caesium took place with a separate pump and a pipe hanging from the rail.

Finally, in 1996 we returned to the Arctic Ocean with the Swedish icebreaker M/S Oden. This expedition focused on studying the distribution of radionuclides in different water masses of the central Arctic Ocean. The expedition crossed the Barents Sea, entered the Nansen Basin at the St. Anna Trough, and continued north across the Amundsen Basin. The main part of the expedition was concentrated on the north Lomonosov Ridge and the return route passed the North Pole and went south along 10 °E towards Svalbard. Water samples from the surface and subsurface layers, as well as bottom sediments, were collected for analysis of fission products and transuranic elements in seawater and sediment.

Acknowledgement

The expeditions were organized by the Swedish Polar Research Secretariat, which is a government agency that promotes and coordinates Swedish polar research to the Arctic and Antarctic regions. We greatly acknowledge the support from the organizers and the leaders of the expeditions. Financial support by the Natural Research Council (NFR), Magnus Bergvalls Foundation, Carl Jönssons understödsfond and all others is greatly acknowledged.

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We also wish to express our gratitude to all those who supported our expeditions with goods and equipment. Among those were: AB TetraPak, Alfa Laval AB, SAB-NIFE AB, Esselte Office AB, Nordic Baltzer AB, Arla Economic Association, Christian Berner AB, Scanpump AB, Kiviks Musteri AB, Venilationsutveckling AB, Gambro AB, Vattenteknik AB, Millipore AB, Hitachi Sales Scandinavia AB, Nikkon Sweden.

We also wish to express our gratitude to Professor Svante Björk and coworkers from the department of Quaternary Geology at Lund University who provided us with the sediment cores. A special thanks to Mrs. Gertie Johansson, Birgit Amilon, and Carin Lingårdh for their good care of samples, skillful radiochemical separations and radioactivity measurements.

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Chapter 2. Ymer-80 Expedition

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With contributions by:

Lars Ahlgren(†), Boel Forkman

Abstract

Levels of natural radioactivity such as ^{222}Rn (radon) and its long-lived daughters ^{210}Pb and ^{210}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 ^{222}Rn concentration in air measured during the Ymer-80 expedition in samples north of latitude 78.8°N , was 33 ± 4 (one standard error) $\text{mBq}\cdot\text{m}^{-3}$ during Leg 1 (July-Aug.), and 105.3 ± 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 ± 5 $\text{mBq}\cdot\text{m}^{-3}$.

During July, August and September, the monthly average concentrations of ^{210}Pb in air at positions north of 75°N latitude were 31 ± 15 , 89 ± 61 and 105 ± 57 $\mu\text{Bq}\cdot\text{m}^{-3}$ respectively with a the grand average for all 3 months of 75 ± 28 $\mu\text{Bq}\cdot\text{m}^{-3}$.

An extensive radiochemistry program was also established to measure $^{134+137}\text{Cs}$ and the trans-uranium elements $^{238+239+240}\text{Pu}$ and ^{241}Am in water, sediment and biota.

The concentration of ^{137}Cs in surface seawater along the Norwegian coast was quite constant about 157 ± 7 $\text{Bq}\cdot\text{m}^{-3}$. At a latitude about 72°N it starts to decrease exponentially at a rate of 0.43 deg^{-1} to about 20 ± 10 $\text{Bq}\cdot\text{m}^{-3}$ above 78°N .

The results of ^{137}Cs in sediments indicate that the total integrated area-content of ^{137}Cs in a 1000 m water-column and sediment is about 12 $\text{kBq}\cdot\text{m}^{-2}$.

In biota, the highest activity concentration of ^{137}Cs about 1000 $\text{Bq}/\text{kg}_{\text{dwt}}$ was found in lichens. The activity concentration of ^{137}Cs in 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}Cs in *Fucus* and *Laminaria* was about $1-2$ $\text{Bq}/\text{kg}_{\text{dwt}}$, and the algae/sea-water activity-concentration ratio was about 75 .

The distribution of the trans-uranium element $^{239+240}\text{Pu}$ in sea water decrease from 14 to 10 $\text{mBq}\cdot\text{m}^{-3}$ up to 73°N but increase again to 17 $\text{mBq}\cdot\text{m}^{-3}$ at high latitudes while ^{137}Cs decrease.

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The average $^{241}\text{Am}/^{239+240}\text{Pu}$ 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,

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

In conclusion, about 25% of ^{137}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 ^{241}Am in the Svalbard area originate from *in situ* build-up due to the decay of ^{241}Pu .

2.1 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 1*” (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.

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

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route from Tromsø 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 Tromso (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 2-2

The Swedish ice-breaker HMS Ymer

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).

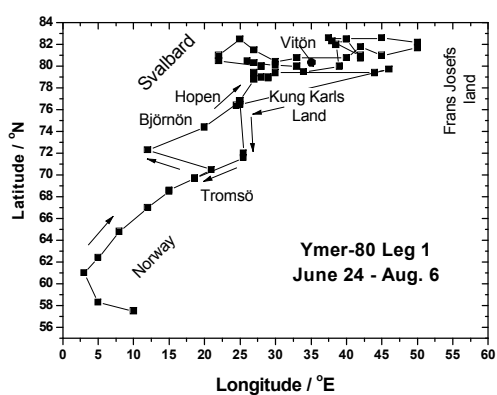


Figure 2-3a

The route of the Ymer-80 expedition Leg 1.

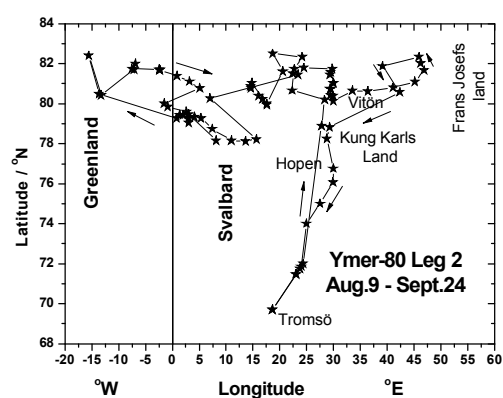


Figure 2-3b

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}Rn (radon) and its long-lived daughters ^{210}Pb and ^{210}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°N the average activity concentrations of ^{222}Rn was $75 \pm 21 \text{ Bq.m}^{-3}$ and of ^{210}Pb $75 \pm 28 \mu\text{Bq.m}^{-3}$. During a two-week period of persistent polar winds, the mean radon concentration decreased to $19 \pm 5 \text{ mBq.m}^{-3}$. The concentrations of ^{222}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}Rn and the long-lived daughter measurements are insensitive to contamination from ship and local settlements (Samuelsson et al., 1986).

2.2 ^{222}Rn and ^{210}Pb in the Arctic Air

Since only a few measurements of ^{222}Rn and ^{210}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}Po air concentration of $100 \mu\text{Bq.m}^{-3}$ has been reported (Rahn and McCaffrey, 1979). During 1961-62 the corresponding value at Thule, Greenland was reported to be about $200 \mu\text{Bq m}^{-3}$ (Patterson jr and Lockhart jr, 1964). In 1979 on the islands of Amchitka, Alaska (51°N , 55°E), that correspond to a maritime Arctic location, the air concentration of ^{210}Po was only $2 \mu\text{Bq.m}^{-3}$ (Nevissi and Schell jr, 1980). Low values of ^{210}Po in the range of $2\text{-}73 \mu\text{Bq.m}^{-3}$ has been reported from measurements on Franz Josef Land (Jaworowski, 1969). During 1953-1959 the ^{222}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 mBq.m^{-3} . and 400 mBq.m^{-3} . respectively (Lockhart jr, 1962)).

No measurements of ^{222}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}Rn in the air along its route in the Arctic.

2.2.1 Measurements of ^{222}Rn in surface air

In order to measure the low radon concentrations anticipated, the air was flowing through a cooled (-14°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 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}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.

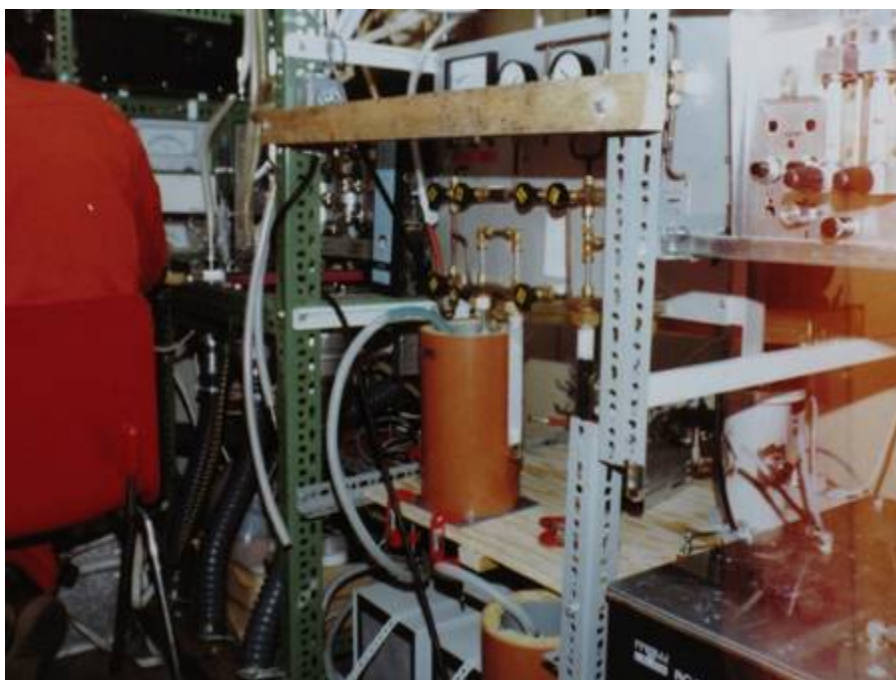


Figure 2-4

The cooled charcoal trap system for collecting ^{222}Rn in the air mounted on board Ymer (Picatif G210, Pica, France)

2.2.2. Measurements of ^{222}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}Rn decays with a half-life of 3.82 days to the following short-lived radon daughters: ^{222}Rn (3.82 days) \rightarrow ^{218}Po (RaA 3.10 min) \rightarrow ^{214}Pb (RaB 26.8 min) \rightarrow ^{214}Bi (RaC 19.9 min) \rightarrow ^{214}Po (RaC' 164.3 ms) \rightarrow ^{210}Pb (RaD 22.20 a) \rightarrow ^{210}Bi (RaE 5.01 d) \rightarrow ^{210}Po (RaF 138.4 d) \rightarrow ^{206}Pb (stable). In the atmosphere, the decay products from ^{222}Rn attach to airborne particles and deposit as dry and wet deposition onto the earth's surface. The decay products following ^{214}Po are the long-lived ^{210}Pb , ^{210}Bi , ^{210}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 $5\text{ }\mu\text{m}$, Sartorius, W. Germany). The filters were changed every 24 h and stored for later analysis of ^{210}Po and ^{210}Pb at our laboratory in Lund.

2.2.3 Results of ^{222}Rn surface air concentration

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

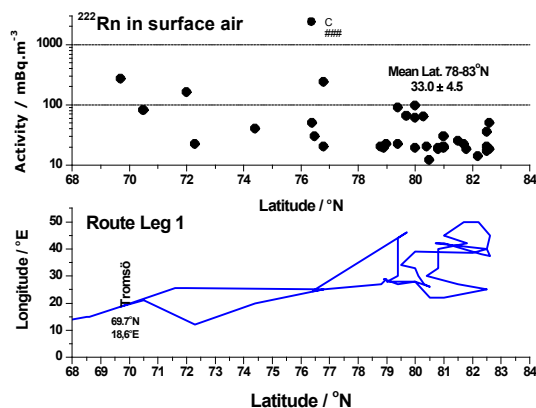


Figure 2-5a

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

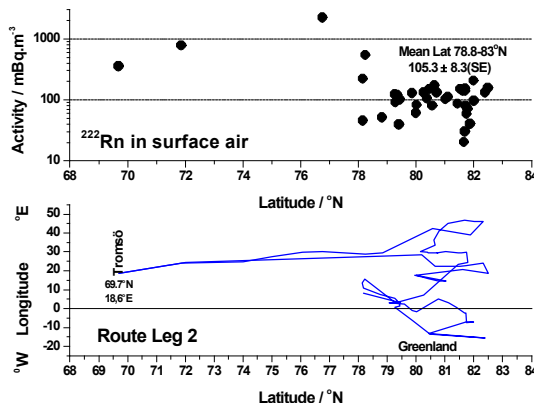


Figure 2-5b

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 ± 0.24 (one standard error) mBq.m^{-3} .

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

2.2.4 Results of Radon daughters in the air

During July, August and September, the monthly average concentrations of ^{210}Pb in air at positions north of 75°N latitude were 31 ± 15 , 89 ± 61 and 105 ± 57 $\mu\text{Bq.m}^{-3}$ respectively with a the grand average for all 3 months of 75 ± 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).

2.3 Cesium-137 measurements

2.3.1 ^{137}Cs in surface water

Seawater samples of 100-200 litre pumped from an inlet at 7 m depth, were collected in special precipitation vessels located at the front deck of the ship (**Fig 2-6**). After ^{134}Cs had been added to the sample as a radiochemical yield determinant, Caesium was separated by adsorption on to microcrystalline ammonium-molybdo-phosphate. 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}Cs in water samples collected between 57°N to 82.8°E is displayed in **Figure 2-7** as a function of latitude. Along the Norwegian coast the concentration of ^{137}Cs in surface seawater was quite constant about $157 \pm 7 \text{ Bq.m}^{-3}$. At a latitude about 72°N it starts to decrease exponentially at a rate of 0.43°N^{-1} to about $20 \pm 10 \text{ Bq.m}^{-3}$ above 78°N . It is obvious that the concentration of ^{137}Cs found along the Norwegian coast is much higher than expected from nuclear weapon fallout. 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 activity concentration in the North Sea and adjacent areas also confirm our results (Kautsky, 1980). The ^{137}Cs activity concentration in area water also decreases rapidly with increasing distance from the coast (Kautsky, 1980, Aarkrog et al., 1982). Along the Norwegian coast from 57.8°N , 8.1°E to 69.0°N , 14.4°E , the ^{137}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, one for precipitation ^{137}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.

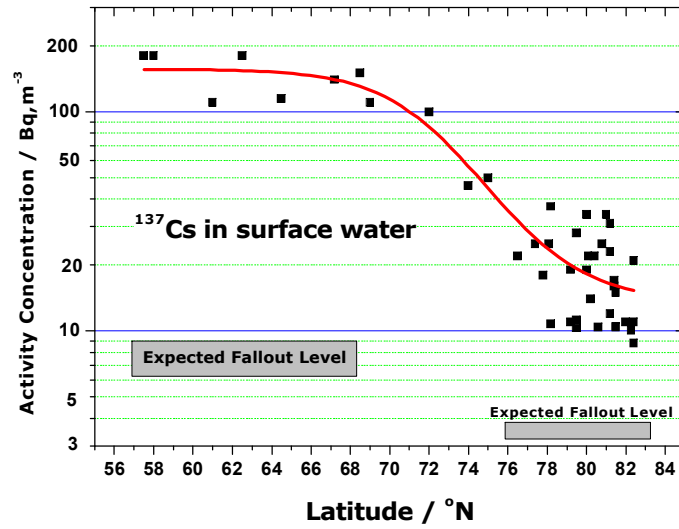


Figure 2-7

The lower diagram shows the activity concentration of ¹³⁷Cs in water samples collected between 57°N to 82.8 °E as a function of latitude. The upper diagram shows the sampling sites.

The ¹³⁷Cs activity concentration varies with the latitude as a Boltzmann sigmoid equation displayed in **Figure 2-7**.

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

where

A is the activity concentration of ¹³⁷Cs in sea water Bq.m⁻³ at various latitudes

A1 is the minimum activity concentration at high latitudes

A2 is the minimum activity at low latitudes

Lat_m is the latitude at the median activity value (A1+A2)/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 _m	72.0	0.7
k _{Lat}	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}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).

2.3.2 ^{137}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}Cs in a 1000 m water-column and sediment is about 12 kBq.m⁻². This value exceeds the integrated ^{137}Cs area-content of 2.2 ± 0.3 kBq.m⁻² on Svalbard (78.2°, 16.0°E) which originates from air borne fallout.

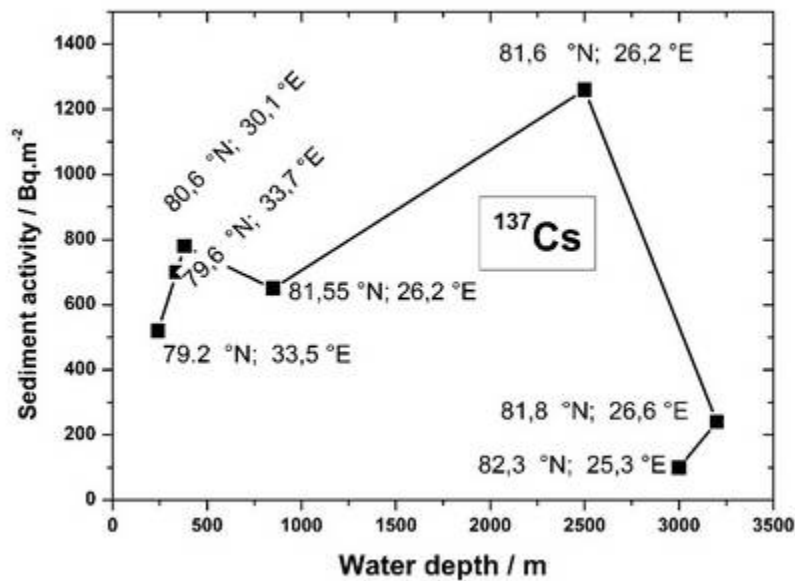


Figure 2-8

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

The amount of ^{137}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}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.

2.3.3 ^{137}Cs in biota

The results for biological samples are presented in **Figure 2-9**. The highest activity concentration of ^{137}Cs about 1000 Bq/kg_{dwt} was found in lichens. The activity concentration of ^{137}Cs in polar bears was about 10 Bq/kg_{dwt}, in seals about 1 Bq/kg_{dwt}, and in birds about 1-7 Bq/kg_{dwt}. The activity-concentrations of ^{137}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}Cs in *Fucus* and *Laminaria* was about 1-2 Bq/kg_{dwt}, and the algae/seawater 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).

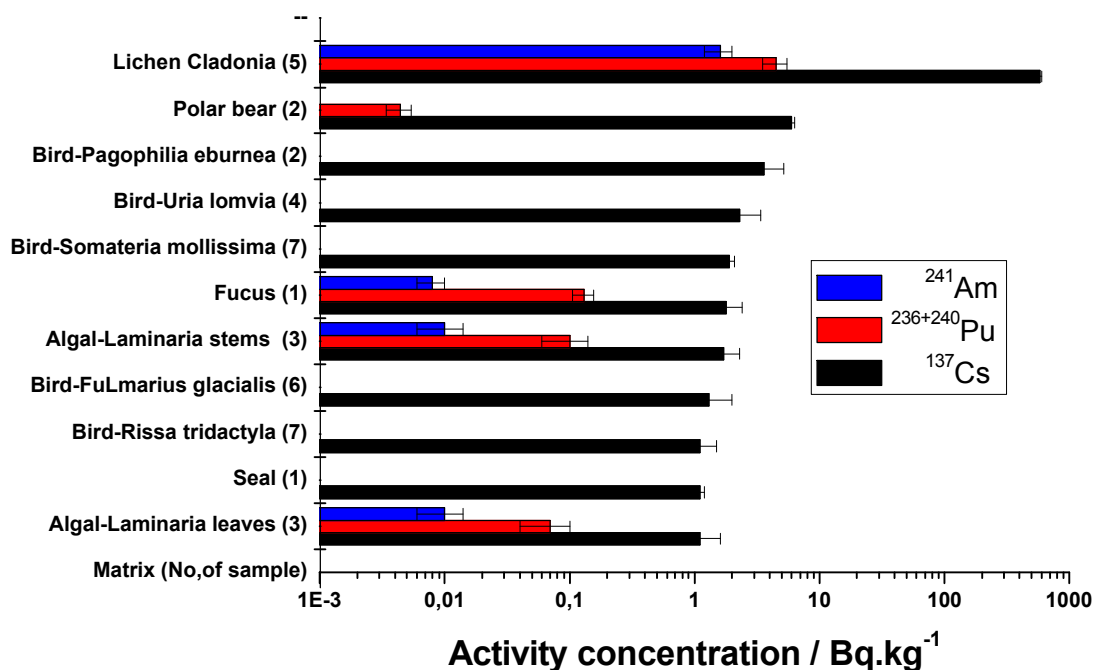


Figure 2-9

Activity concentration (Bq per kg of dry mass, Bq/kg_{dwt}) of ^{137}Cs , $^{238+240}\text{Pu}$ and ^{241}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.

2.4 Plutonium-239+240 and Americium-241

2.4.1 Trans-Uranium elements in Sea water

The distribution of the trans-uranium elements $^{239+240}\text{Pu}$ and ^{241}Am in the surface water of the Norwegian, Barents and Greenland Seas is quite different from that of ^{137}Cs . The distribution of the transuranium elements displayed in **Figure 2-10** decrease from 14 to 10 $\text{mBq}\cdot\text{m}^{-3}$ up to 73°N but increase again to 17 $\text{mBq}\cdot\text{m}^{-3}$ at high latitudes while ^{137}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 ± 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 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}Pu and ^{241}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

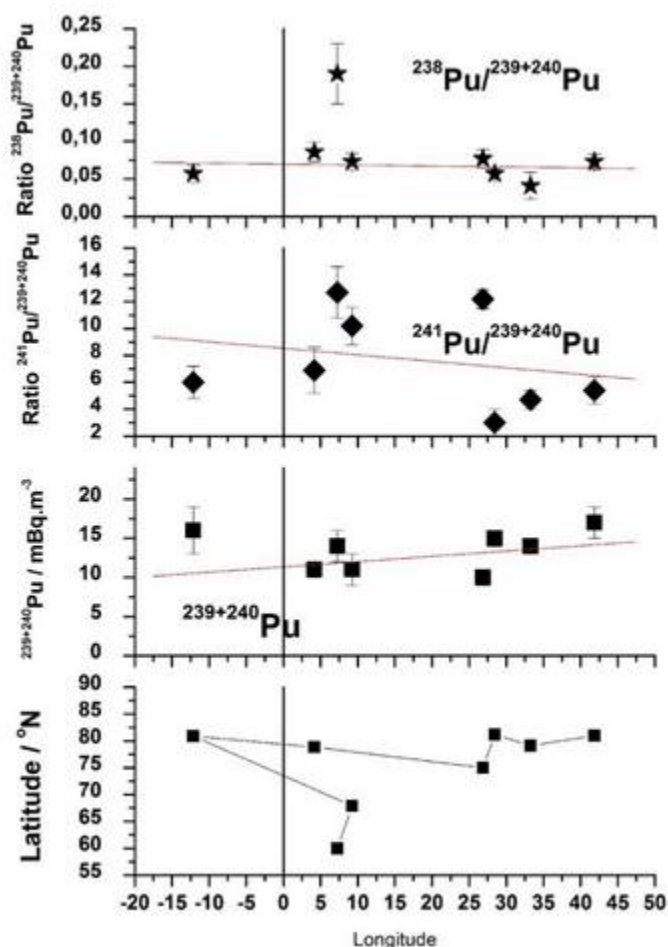


Figure 2-10

Longitudinal distribution of plutonium isotopes water

Deepwater samples were collected with a 100 l Niskin bottle at latitudes around 80.4 ± 1.4 °N and along longitudes 2° W – 45.5 °E. The results of the salinity and activity concentration of ^{137}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}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}Am -activity concentration in surface water shows no significant correlation with the latitude. This indicates that ^{241}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).

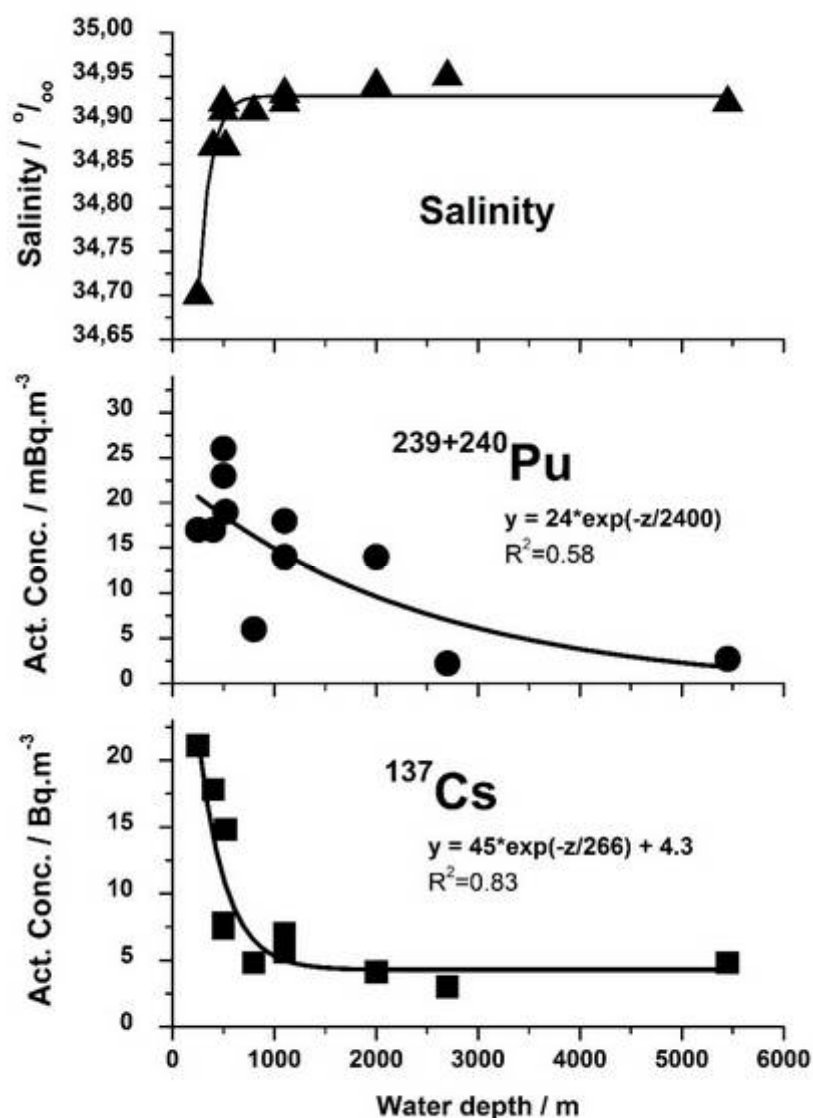


Figure 2-11

The variation of salinity and activity concentration of ¹³⁷Cs and ²³⁹⁺²⁴⁰Pu with depth of water at latitude 80.4±1,4 °N and longitude 2°W; 45.5 °E

2.4.2 Trans-Uranium elements in *Fucus* and *Laminaria*, lichen and moss

The highest activity concentration of ²³⁸⁺²⁴⁰Pu and ²⁴¹Am 6 and 2 Bq/kg_{dwt} respectively was found in lichens. The activity concentration of ²³⁸⁺²⁴⁰Pu in polar bears was about 4 mBq/kg_{dwt}. The activity-concentration of ²³⁹⁺²⁴⁰Pu and ²⁴¹Am in the alga *Fucus* and *Laminaria* was about 100-10 Bq/kg_{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}Pu and ^{238}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	$\pm\text{SD}$	^{241}Pu $/^{239+240}\text{Pu}$ Ratio	$\pm\text{SD}$	^{238}Pu $/^{239+240}\text{Pu}$ Ratio	$\pm\text{SD}$	
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	$\pm\text{SD}$	^{241}Pu $/^{239+240}\text{Pu}$ Ratio	$\pm\text{SD}$	^{238}Pu $/^{239+240}\text{Pu}$ Ratio	$\pm\text{SD}$	
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

2.4.3 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 $26 \pm 3 \text{ Bq.m}^{-2}$ 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.

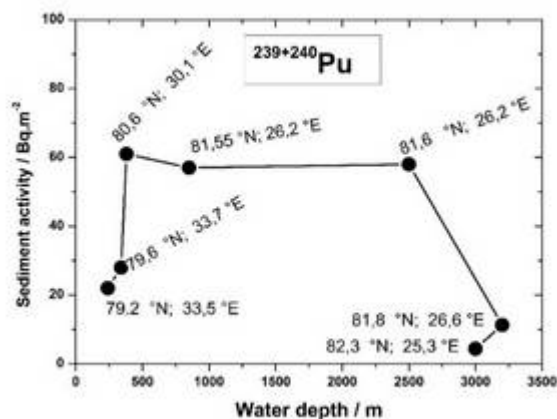


Figure 2-12
The integrated area content in sediments of $^{239+240}\text{Pu}$ versus depth of water at corresponding sampling site (Lat.;Long.)

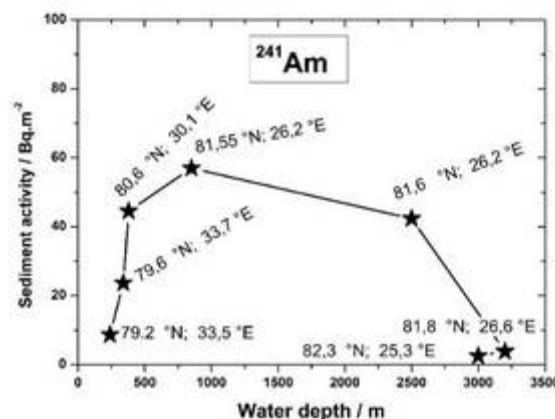


Figure 2-13
The integrated area content in sediments of ^{241}Am versus depth of water at corresponding sampling site (Lat.;Long.)

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 ± 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 $^{239+240}\text{Pu}$ will indicate if other sources than fall-out are significant.

A mean activity ratio of $^{238}\text{Pu}/^{239+240}\text{Pu}$ 0.047 ± 0.008 may was found in samples from Greenland and Svalbard which only contaminated by ^{238}Pu from fallout only. The mean of all water samples analysed for ^{238}Pu and sediment, give an activity ratio of 0.060 ± 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}Pu in Barents and Greenland Seas originate from European reprocessing facilities. The Activity concentration of ^{238}Pu originating from these facilities is between 0.20 and 0.38 mBq.m^{-3} , and the corresponding value for ^{241}Pu is in the range of 16 - 39 mBq.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 mBq.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 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).

2.5 CONCLUSIONS

The average ^{222}Rn concentration in air measured during the Ymer-80 expedition in samples north of latitude 78.8°N , was 33 ± 4 (one standard error) $\text{mBq}\cdot\text{m}^{-3}$ during Leg 1 (July-Aug.), and 105.3 ± 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 ± 5 $\text{mBq}\cdot\text{m}^{-3}$. During July, August and September, the monthly average concentrations of ^{210}Pb in air at positions north of 75°N latitude were 31 ± 15 , 89 ± 61 and 105 ± 57 $\mu\text{Bq}\cdot\text{m}^{-3}$ respectively with a the grand average for all 3 months of 75 ± 28 $\mu\text{Bq}\cdot\text{m}^{-3}$.

The concentration of ^{137}Cs in surface seawater along the Norwegian coast was quite constant about 157 ± 7 $\text{Bq}\cdot\text{m}^{-3}$. At a latitude about 72°N it start to decrease exponentially at a rate of 0.43 deg^{-1} to about 20 ± 10 $\text{Bq}\cdot\text{m}^{-3}$ above 78°N . The results of ^{137}Cs in sediments indicate that the total integrated area-content of ^{137}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}Cs about 1000 $\text{Bq}/\text{kg}_{\text{dwt}}$ was found in lichens collected on Svalbard. The activity concentration of ^{137}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}Cs in the algae *Fucus* and *Laminaria* was about $1-2$ $\text{Bq}/\text{kg}_{\text{dwt}}$, 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 $\text{mBq}\cdot\text{m}^{-3}$ up to 73°N but increase again to 17 $\text{mBq}\cdot\text{m}^{-3}$ at high latitudes while ^{137}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 ± 0.04 (2 S.E), of 31 samples with the range $0.04-0.32$ in surface water,

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Radioactivity exploration from the Arctic to the Antarctic.

Chapter 3. The SWEDARP expedition Oct 1988 – April 1989

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*“Snowhill” painted on board M/S Stena Arctica by the artist Lars Lerin, who followed and documented the SWEDARP expedition.
(copy by permission of Lars Lerin)*

Abstract

The Swedish Antarctic Research Expedition named “*SWEDARP*” was performed during October 1988 through April 1989. The aim of our project was to explore the radioactivity in air and water from Gothenburg to the Antarctic.

Between Gothenburg and the Equator, the average activity concentration of ^7Be in air was 4.3 ± 0.7 $\text{mBq}\cdot\text{m}^{-3}$. The activity concentration of ^7Be in the South Atlantic down to Antarctica varied between 1.3 and 1.7 with an average of 1.5 ± 0.8 $\text{mBq}\cdot\text{m}^{-3}$.

The average activity concentration of ^{210}Pb in air during autumn 1988 on the route Gothenburg-Montevideo was about 290 ± 270 $\mu\text{Bq}\cdot\text{m}^{-3}$, and on the return in spring 1989 it was about 230 ± 140 $\mu\text{Bq}\cdot\text{m}^{-3}$. At the Equator, the average activity concentration of ^{210}Pb in November 1988 was about 630 ± 170 and in April 1989 about 260 ± 210 $\mu\text{Bq}\cdot\text{m}^{-3}$. In the South Atlantic down to Antarctica during February-March 1989, the activity concentration of ^{210}Pb varied between 11 and 58 $\mu\text{Bq}\cdot\text{m}^{-3}$.

The average activity concentration of ^{210}Po in air during the route Gothenburg- Montevideo in 1988 was about 63 ± 58 , and on the return, it was about 60 ± 44 $\mu\text{Bq}\cdot\text{m}^{-3}$. At the Equator, the average activity concentration in Nov 1988 was about 132 ± 45 , and in April 1989 about 70 ± 60 $\mu\text{Bq}\cdot\text{m}^{-3}$. The activity concentration of ^{210}Po in the South Atlantic down to Antarctica during February-March 1989 varied between 6 and 14 $\mu\text{Bq}\cdot\text{m}^{-3}$.

The inventory ($\text{Bq}\cdot\text{m}^{-2}$) of various deposited ratio nuclides, measured in moss, lichen, soil and lake sediments was: 280 ± 110 for ^{210}Pb (excess); 155 ± 95 for ^{137}Cs , 4.6 ± 2.6 for $^{239+240}\text{Pu}$, 1.6 ± 1.0 for ^{241}Am and, 0.95 ± 0.5 for ^{238}Pu .

^{137}Cs activity concentration ($\text{Bq}\cdot\text{m}^{-3}$) in surface waters samples along the route of the SWEDARP expedition was about 3 $\text{Bq}\cdot\text{m}^{-3}$ above 20 °N, but decreased to about 2 $\text{Bq}\cdot\text{m}^{-3}$ between 20 °N and 40 °S where it decreased again to about 1 $\text{Bq}\cdot\text{m}^{-3}$, and at 55 °S there is a further decrease to about 0.3 $\text{Bq}\cdot\text{m}^{-3}$. Radio-Caesium, which originates from atmospheric nuclear weapons tests carried out in the late 1950's and the early 1960's shows a long residence time in surface waters of the North and South Atlantic Oceans with half-time, corrected for physical decay estimated to be more than about 100 years. Concentration factor for ^{137}Cs estimated to 15000 for fish in the Antarctic is much higher than the value of 500 given by The IAEA 1985. The MacroAlgae/Water concentration factors, was found to be about 1800 in the Antarctic compared to 100 in the Arctic.

The $^{239+240}\text{Pu}$ activity concentrations in the surface water was about 8 $\text{mBq}\cdot\text{m}^{-3}$ in the latitude band 5°-25 °N, about 3 $\text{mBq}\cdot\text{m}^{-3}$ in the latitude band 25-5 °N; and about 1,5 $\text{mBq}\cdot\text{m}^{-3}$ in the latitude band 0 °S-60 °S. Plutonium isotopes $^{239+240}\text{Pu}$ from atmospheric nuclear weapons tests has a low half-life of 7-8 years due to the higher affinity to sinking particles for Pu than Cs. In the southern hemisphere,

^{238}Pu was found as the result of the 1964 burn-up of a satellite in the atmosphere over the Mozambique Channel, and was measured in a few water samples from the southern hemisphere. The activity ratio of ^{238}Pu to $^{239+240}\text{Pu}$ was in the range of 0.14-0.22.

The Average activity concentrations ($\text{Bq}/\text{kg}_{\text{dw}}$) of ^{137}Cs are in flesh of seals 2.5, penguins 0.2 and, fish 2.5 in liver of seals 0.6 and penguins 0.7 and in kidneys of seals 0.5. The Average activity concentrations ($\text{Bq}/\text{kg}_{\text{dw}}$) of ^{210}Po are in flesh of seals 8, penguins 4 and, fish 7, in liver of seals 110 and penguins 43 and, fish 5, and in kidneys of seals 91 and of penguins 43. The Average activity concentrations ($\text{Bq}/\text{kg}_{\text{dw}}$) of ^{210}Pb are in flesh of seals 0,3, penguins 0,2, and fish 0.6, in liver of seals 4 and penguins 1 and, fish 1, and in kidneys of seals 1 and of penguins 1. The Average activity concentration of ^{137}Cs in samples of Krill is about 1.7 $\text{Bq}/\text{kg}_{\text{dw}}$ of ^{210}Po and ^{210}Pb are in samples of Amphipods, 87 and 1.4 $\text{Bq}/\text{kg}_{\text{dw}}$ respectively.

3.1 Route of the SWEDARP expedition

The Swedish Antarctic Research Expedition named “*SWEDARP*” was performed during October 1988 through April 1989 organized by the Swedish Polar Research program. The ship M/S Stena Arctica (**Figure 3-1**), was used as research platform, with air sampling device and laboratory container loaded on board in Gothenburg.



Figure 3-1
The first M/S Stena Arctica under Stena Bulk, loading in the harbour at Gothenburg for the SWEDARP expedition.

Photo: Kjell-Åke Carlsson

The route of the SWEDARP expedition is displayed in **Figure 3-2**.

We started in Gothenburg (67.4°N 12°E) with the first destination Montevideo (34.8°S 56.2°W). From Montevideo, we continued to the Swedish permanent base “*Svea*” at the North shelf of Antarctica. After unloading supply and equipment for the continental research group at “*Svea*”, the ship continued to the Argentinean base “*Marambio*” (**Figure 3-3**).

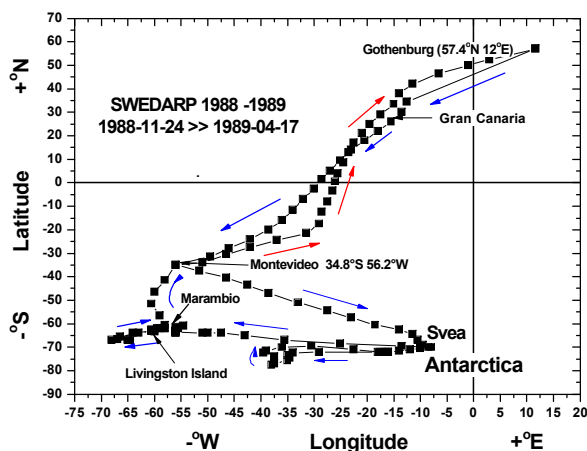


Figure 3-2

Route of the SWEDARP expedition 1988 -1989. The black square dots indicate the locations of air sampling.



Figure 3-3
The expedition approaching the Argentine research station "Esperanza base" at 63°24' S, 56°59' W.
Photo: Kjell-Åke Carlsson

The expedition members were allowed to visit the Esperanza base with the remains of the stone-hut on "Paulet Island" where Captain C.A. Larsen stayed, after his vessel "Antarctic" sank in 1903.



Figure 3- 4
During the Antarctic winter 1903-04, Gunnar Andersson, Duse and Grundén survived in this stone-hut, where prayer and supplication of faith and hope embedded in their souls, until the corvette Uruguay came to rescue them. Hope Bay, Antarctica, Argentina 1905.
Photo: Kjell-Åke Carlsson

3.2 Measurements of ^{210}Pb , ^{210}Po as well as ^7Be in surface air.

During the SWEDARP expedition to the Antarctica, surface air samples of ^{210}Pb , ^{210}Po as well as ^7Be were collected with a so-called "Andersen" air-sampling device installed on board the ship M/S Stena Arctica. Air volumes of about 1 500 m³ were collected at each occasion on membrane filters (size

0.25×0.25 m) at a flow rate of 100 m³h⁻¹. The sampler had previously been involved in an inter-calibration project of air samplers (Vintersved, 1994). During the Arctic Ocean expedition in 1991, the Andersen sampler was compared with a FOA transportable reference air sampler (Microsorban filter, 0.56·0.56 m², 1100 m³h⁻¹). The samplers were placed close together on deck of the cruise vessel. The ⁷Be results for the Anderson sampler were normalized to that of the FOA sampler. FOA nowadays FOI, is a Swedish research institute in the areas of defence and security.

The time between collection and analysis of the filters was maximum 2 months. Filters were whenever possible sent to Sweden by mail from Montevideo or Marambio Base in Antarctica or by personnel leaving the expedition. The filters were measured back home in Lund, for ⁷Be by gamma spectrometry using a high performance Germanium detector (HPGe Canberra). ²¹⁰Po and ²¹⁰Pb were measured by radiochemical procedure after adding ²⁰⁹Po as radiochemical yield determinant and the samples were wet-ashed by using a mixture of concentrated nitric acid and hydrochloric acid. Polonium was spontaneously deposited on nickel discs, and the activity of ²⁰⁹Po and ²¹⁰Po was measured by alpha spectrometry using surface ion implanted silicon detectors. The radiochemical yield was about (70-80 %).

Remaining traces of polonium were removed by anion exchange. The solution was then kept for about 8-12 months, to allow in-growth of ²¹⁰Po from ²¹⁰Pb. New ²⁰⁹Po yield determinant was added again, and ²⁰⁹Po and ²¹⁰Po was deposited on nickel discs and measured by alpha spectrometry.

The activity of ingrown ²¹⁰Po was calculated, with corrections for build-up from ²¹⁰Pb from sampling to analysis. Decay correction for the time elapsed between plating and measurement of ²¹⁰Po was done as well. Finally, after all appropriate corrections for radioactive decay were done, the activity concentrations of ²¹⁰Po and ²¹⁰Pb in air at sampling time were obtained.

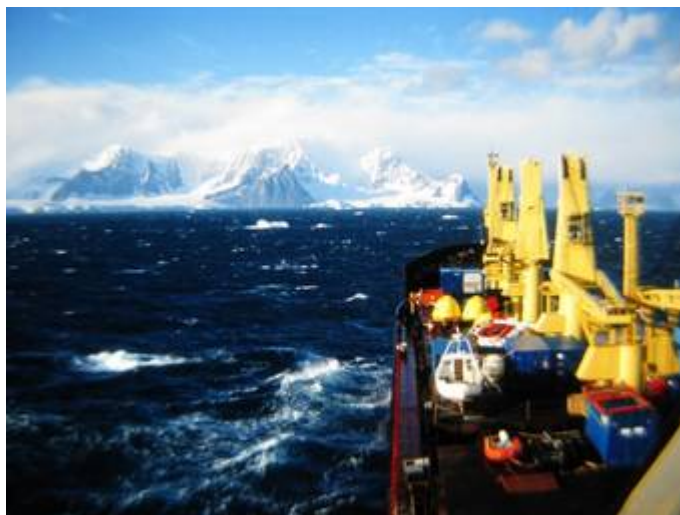


Figure 3-5

Sampling of radioactivity in air and water around Antarctica.

Photo: Kjell-Åke Carlsson

In **Figure 3-6** are given the results from the measurements of ⁷Be activity concentrations as well as of ²¹⁰Pb, and ²¹⁰Po in the surface air, recorded during the routes between Gothenburg 57°43'N 11°59'E - Montevideo 34°50'S 56°11'W and return. In **Table 3-1** are given the average activity

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concentrations of ^7Be ($\text{mBq}\cdot\text{m}^{-3}$), ^{210}Pb , and ^{210}Po ($\mu\text{Bq}\cdot\text{m}^{-3}$) in the surface for various segments of the route Gothenburg $57^\circ43'\text{N}$ $11^\circ59'\text{E}$ - Montevideo $34^\circ50'\text{S}$ $56^\circ11'\text{W}$ – Antarctica and return.

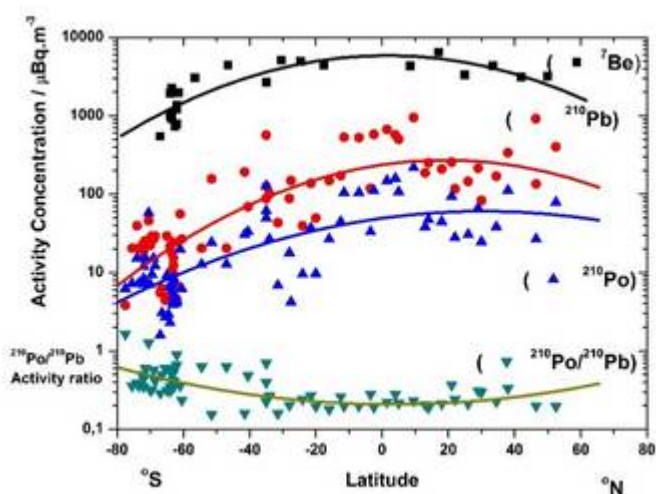


Figure 3-6

Activity concentrations of ^7Be as well as of ^{210}Pb , and ^{210}Po in the surface air as recorded on the routes Gothenburg $57^\circ43'\text{N}$ $11^\circ59'\text{E}$ - Montevideo $34^\circ50'\text{S}$ $56^\circ11'\text{W}$ and return.

The curves are 2^{nd} degree polynomial fittings.

Table 3-1

Average activity concentrations of ^7Be ($\text{mBq}\cdot\text{m}^{-3}$), ^{210}Pb , and ^{210}Po ($\mu\text{Bq}\cdot\text{m}^{-3}$) in the surface air on various segments on the route Gothenburg $57^\circ43'\text{N}$ $11^\circ59'\text{E}$ - Montevideo $34^\circ50'\text{S}$ $56^\circ11'\text{W}$ – Antarctica and return.

Time	Latitude N(+)/S(-)	Longitude W(-)/E(+)	Number of samples	^7Be		^{210}Pb		^{210}Po	
				Ave.	SD	Ave.	SD	Ave.	SD
1988									
1124>1130	34.5 > 14	- 12.5 > - 56	6			163	61	36	9
1201>1205	9.5 > - 11.5	- 25 > - 56	6			626	169	132	45
1124>1212	34 > - 35	- 12.5 > - 56	5			288	268	63	58
1213>1227	- 35 > - 70	- 56 > - 8.5	15			45	45	21	17
1989									
0101>0205	- 72 > - 70	- 16 > - 8	15			22	9	9	3
0211 >0318	- 64 > - 60.5	- 50 > - 58	33	1,3	0,6	15	13	6	4
0319>0324	- 56.5 > - 35	- 59 > - 56	6	3,4	0,9	305	227	61	58
0206>0324	- 70 > - 35	- 13.5 > - 56	39	1,7	1,0	58	126	14	27
0401>0407	12.5 > - 13	- 28.5 > - 23.5	7	4,3	0,3	262	207	69	60
0325>0417	- 35 > N 52	- 56 > 3	24	4,3	1,1	232	140	60	44

In **Table 3-2** are given the results of average activity ratios of $^7\text{Be}/^{210}\text{Pb}$, and $^7\text{Be}/^{210}\text{Po}$ in the surface air on the routes Gothenburg 57°43'N 11°59'E - Montevideo 34°50'S 56°11'W - Antarctica and return.

Table 3-2

Average activity ratios (\pm SD) of $^7\text{Be}/^{210}\text{Pb}$, and $^7\text{Be}/^{210}\text{Po}$ at various route sections recorded during the SWEDARP expedition 1989.

Time	Latitude	Longitude	Number of samples	^7Be		^{210}Pb		^{210}Po	
	N(+)/S(-)	W(-)/E(+)		mBq/m ³	SD	Ave.	SD	Ave.	SD
1988									
1124>1130	34.5 > 14	- 12.5 > - 56	6			163	61	36	9
1201>1205	9.5 > - 11.5	- 25 > - 56	6			626	169	132	45
1124>1212	34 > - 35	- 12.5 > - 56	5			288	268	63	58
1213>1227	- 35 > - 70	- 56 > - 8.5	15			45	45	21	17
1989									
0101>0205	- 72 > - 70	- 16 > - 8	15			22	9	9	3
0211 >0318	- 64 > - 60.5	-50 > - 58	33	1,3	0,6	15	13	6	4
0319>0324	- 56.5 > - 35	- 59 > - 56	6	3,4	0,9	305	227	61	58
0206>0324	- 70 > - 35	- 13.5 > - 56	39	1,7	1,0	58	126	14	27
0401>0407	12.5 > - 13	- 28.5 > - 23.5	7	4,3	0,3	262	207	69	60
0325>0417	- 35 > N 52	- 56 > 3	24	4,3	1,1	232	140	60	44

The activity concentration of ^7Be in the South Atlantic down to Antarctica varied between 1.3 and 1.7 with an average of $1.5 \pm 0.8 \text{ mBq.m}^{-3}$. At the Equator and up to Gothenburg the average activity concentration of ^7Be was $4.3 \pm 0.7 \text{ mBq.m}^{-3}$.

The activity concentration of ^{210}Pb in the South Atlantic down to Antarctica during February-March 1989 varied between 11 and $58 \text{ }\mu\text{Bq.m}^{-3}$. At the Equator, the average activity concentration of ^{210}Pb in November 1988 was about 630 ± 170 and in April 1989 about $260 \pm 210 \text{ }\mu\text{Bq/m}^3$. The average activity concentration of ^{210}Pb during the route Gothenburg- Montevideo in 1988 was about $290 \pm 270 \text{ }\mu\text{Bq.m}^{-3}$ and on the return Montevideo-Gothenburg it was about $230 \pm 140 \text{ }\mu\text{Bq.m}^{-3}$.

The activity concentration of ^{210}Po in the South Atlantic down to Antarctica during February-March 1989 varied between 6 and $14 \text{ }\mu\text{Bq.m}^{-3}$. At the Equator the average activity concentration in Nov 1988 was about 132 ± 45 , and in April 1989 about $70 \pm 60 \text{ }\mu\text{Bq.m}^{-3}$. The average activity concentration of ^{210}Po during the route Gothenburg- Montevideo in 1988 was about 63 ± 58 , and on the return Montevideo-Gothenburg it was about $60 \pm 44 \text{ }\mu\text{Bq.m}^{-3}$.

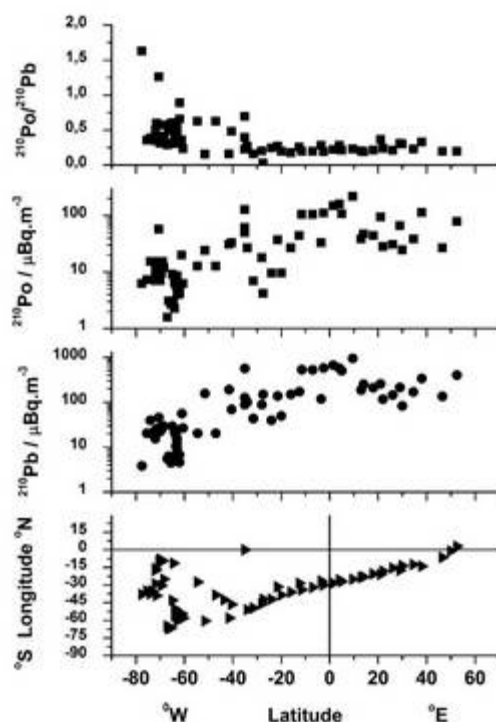


Figure 3-7 Activity concentrations of ^{210}Pb , and ^{210}Po and the $^{210}\text{Po}/^{210}\text{Pb}$ -activity ratio in the surface air as recorded on the routes Gothenburg $57^{\circ}43'\text{N}$ $11^{\circ}59'\text{E}$ - Montevideo $34^{\circ}50'\text{S}$ $56^{\circ}11'\text{W}$ and return.

3.3. Deposition of ^{210}Pb , ^{137}Cs , $^{239+240}\text{Pu}$, ^{238}Pu , and ^{241}Am in the Antarctic Peninsula Area

During the SWEDARP expedition samples of lichens (identified as being close to *Alectoria nigricans*), various types of moss, grass (*Dechampsia Antarctica* and *Colobantimus*) and soil, all with a known area (between $1/16$ and $1/4$ m^2), were collected on the South Shetland Islands (Livingston Island, King George Island, Deception Island and Horseshoe Island).

^{210}Pb , ^{137}Cs , $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am were analyzed in lichen, moss, grass and soil samples, as well as in lake sediments from the South Shetland Islands, the Antarctic, in order to evaluate the flux and deposition of these elements (Roos et al., 1994). Average inventories of the analysed radionuclides in samples collected 1988, are given in Table 3-3.



Figure 3-8

Moss carpets at Livingston Island 62°38' S, 60°30' W.

Photo: Bertil Persson

Table 3-3 Average inventories of the analysed radionuclides in samples collected 1988 in Antarctica

Radionuclide	Inventory Bq.m ⁻²	Samples
²¹⁰ Pb (excess)	280 ± 110	(n=15)
¹³⁷ Cs	155 ± 95	(n =19)
²³⁹⁺²⁴⁰ Pu	4.6 ± 2.6	(n =19)
²⁴¹ Am	1.6 ± 1.0	(n =19)
²³⁸ Pu.	0.95 ± 0.5	(n =19)

From the maximum value of unsupported ²¹⁰Pb the annual deposition of ²¹⁰Pb, is estimated to be 18 ± 5 Bq m⁻². The ratios ²³⁹⁺²⁴⁰Pu and ²⁴¹Am /²³⁹⁺²⁴⁰Pu are 0.21 ± 0.04 and 0.35 ± 0.08 respectively, which agree well with expected values in this area. A significant difference in ¹³⁷Cs /²³⁹⁺²⁴⁰Pu activity ratios was observed between lichens and moss, grass and soil which may be an effect of submerging and melt water. From one of three lakes studied it is possible to perform ²¹⁰Pb dating with reasonably accuracy showing an average annual sedimentation rate in this lake was about 45 g.m⁻².a⁻¹.

As shown in **Table 3-4**, there is a strong correlations were between the deposition of the natural radionuclide ^{210}Pb , and the artificial radionuclides ^{137}Cs , $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am .

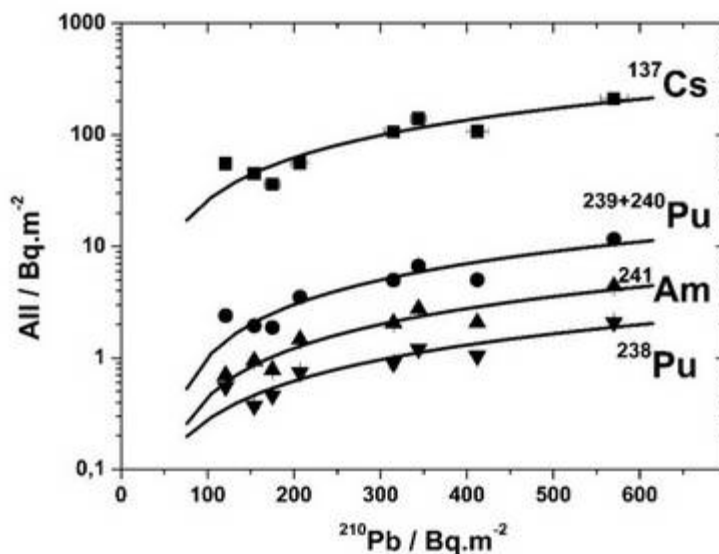


Figure 3-9

The correlations between the deposition of the natural radionuclide ^{210}Pb , and the artificial radio-nuclides ^{137}Cs , $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am .

Due to the wide spread in the values of the deposition level of the artificial radionuclides, the results are displayed in a logline diagram **Figure 3-9**.

Table 3-4

Linear regression coefficients $k=Y/X$ between the deposition X of the natural radionuclide ^{210}Pb , and the deposition Y of artificial radionuclides ^{137}Cs , $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am and corresponding coefficients of variation R .

Radionuclides	k	SD	R
^{137}Cs	0.34	0.02	0.94
$^{239+240}\text{Pu}$	0.017	0.001	0.94
^{241}Am	0.0068	0.0005	0.95
^{238}Pu	0.0032	0.0002	0.94

3.4 Measurements of various Caesium and Plutonium isotopes in water

Radio Caesium (^{134}Cs , ^{137}Cs) and Plutonium (^{238}Pu , $^{239+240}\text{Pu}$) was measured in water samples collected along the route 73 °N to 72 °S. The sampling volume in the Antarctic was often about 1400 l (Holm et al., 1991).

3.4.1 Caesium 137

The result of ^{137}Cs activity concentration ($\text{Bq}\cdot\text{m}^{-3}$) in surface waters samples along the route of the SWEDARP expedition from the North (73°N) and South Atlantic (to 72°S) are displayed in



Figure 3-10

Interior of the water laboratory container on board, where seawater samples of 100-200 l were collected in two special precipitation vessels placed to the right. One for precipitation ^{137}Cs and another for trans-uranium elements. Cartridge-filters (Millipore CWSS 012C3) are placed in the holders on the wall to the left. (Photo: *Kjell-Åke Carlsson*)

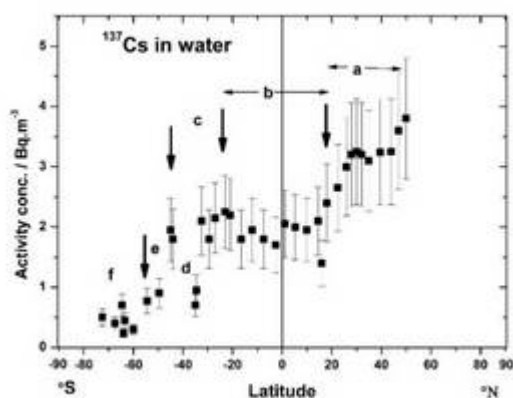


Figure 3-11.

^{137}Cs activity concentration ($\text{Bq}\cdot\text{m}^{-3}$) in surface waters from the North and South Atlantic. All data corrected for physical decay to February 1989.

Following separate regions in **Figure 3-11** can be distinguished in the result of ^{137}Cs activity concentration ($\text{Bq}\cdot\text{m}^{-3}$) in surface waters samples along the route of the SWEDARP expedition from the North (73°N) and South Atlantic (to 72°S):

- above 20°N where the activity concentration are about $3 \text{ Bq}\cdot\text{m}^{-3}$,
- between 20°N and 30°S where the activity concentration decrease from 3 to about $2 \text{ Bq}\cdot\text{m}^{-3}$
- between $30 - 45^\circ\text{S}$ where the activity concentration are fluctuating about $2 \text{ Bq}\cdot\text{m}^{-3}$

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- d. at 40 °S the activity concentrations decrease to about 1 Bq.m⁻³, when entering the South Sea current and
- e. between 55 - 45 °S where the activity concentration are fluctuating between 1 -2 Bq.m⁻³
- f. below the circumpolar Antarctic water at 55 °S (e) there is a decrease to about 0.3 Bq.m⁻³, which are accompanied, by decreases in water temperature.

3.4.2 Caesium 134

In 1986 large amounts of ¹³⁴Cs was released in the Chernobyl accident. The ¹³⁴Cs/¹³⁷Cs-activity ratio in samples of Atlantic surface water In April/May 1986 was about 0.47 and was estimated to be about 0.17 in 1989. This ratio is significantly lower than that in the effluents from European nuclear fuel reprocessing plants, ¹³⁴Cs was only found at few locations north of 35°N, the most southerly latitude we would expect any impact of the release from Chernobyl accident. It is, however, most probably that the ¹³⁴Cs recorded is derived from European reprocessing plants rather than from the Chernobyl accident.

3.4.3 Plutonium 239+240.

The results for ²³⁹⁺²⁴⁰Pu from the SWEDARP expedition are displayed the **Figure 3-12**. The long physical half -life of the plutonium isotopes ²³⁹⁺²⁴⁰Pu makes it unnecessary to correct for physical decay between 1973 and 1989. The plutonium activity concentrations in the surface water are about 8 mBq.m⁻³ in the latitude band 5°-25 °N, about 3 mBq.m⁻³ in the latitude band 25-5 °N, and about 1,5 mBq.m⁻³ in the latitude band 0 °S-60 °S. Slightly higher values were measured around the Antarctic Peninsula. As a comparison we can mention that based on the Swedish Ymer-80 expedition in 1980 the ²³⁹⁺²⁴⁰Pu activity concentrations were about 11-15 mBq.m⁻³ from in the Norwegian Sea and the Barents and Greenland Seas between 55 to 82 °N.

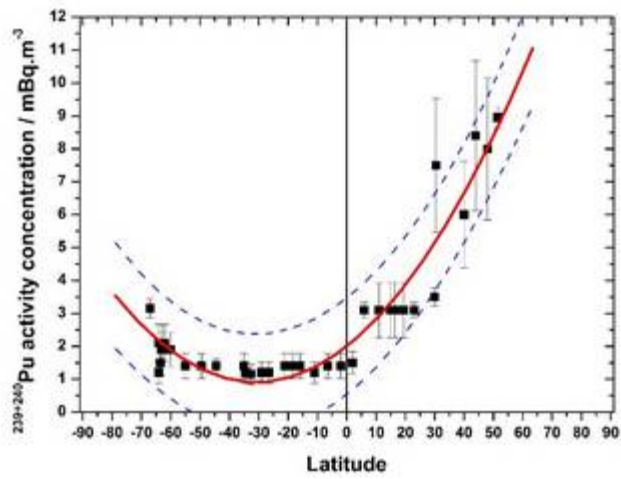


Figure 3-12.

Activity concentration (mBq.m⁻³) of ²³⁹⁺²⁴⁰Pu in surface waters from the North and South Atlantic. Polynomial Regression for Data **R²(COD) = 0.91**: $Y = 2.0 + 0.07 \times X + 0.0011 \times X^2$

3.4.4 Plutonium 238

In the southern hemisphere, ²³⁸Pu originates mainly from a satellite (SNAP-9A containing 1 kg of Pu metal) that in 1964 re-entered the atmosphere and burned up at high altitude over the Mozambique Channel. This event significantly increased both the environmental levels of ²³⁸Pu in addition to the activity ratio to ²³⁹⁺²⁴⁰Pu (which is typically 0.025 in fallout from Nuclear weapons tests), especially in the southern hemisphere.

During the SWEDARP expedition, Plutonium-238 activity was measured in a few water samples from the southern hemisphere- The activity ratio of ²³⁸Pu to ²³⁹⁺²⁴⁰Pu was ranging from 0.14-0.22. In macro algae from the Antarctic Peninsula we found an activity ratio of ²³⁸Pu to ²³⁹⁺²⁴⁰Pu of 0.27±0.03 (n = 6, 1. S.E.), and in terrestrial samples (carpets of mosses and lichens) from the Antarctic Peninsula area an activity ratio of 0.24±0.02 (n =24, 1 S.E.).

3.5 Measurements of various Radionuclides in Biota.

3.5.1 Radionuclides in Marine animals

The activity concentration of ¹³⁷Cs, ²¹⁰Po and ²¹⁰Pb was analyzed in samples of various species of seals, penguins and fish are displayed in Figures 3-13,14,15 for flesh, liver and kidney.

To the right in these Figures are also given data for Amphipods and Krill. The horizontal bars in the figures 3-13,14,15 represent the average activity concentration of the radionuclide in question.

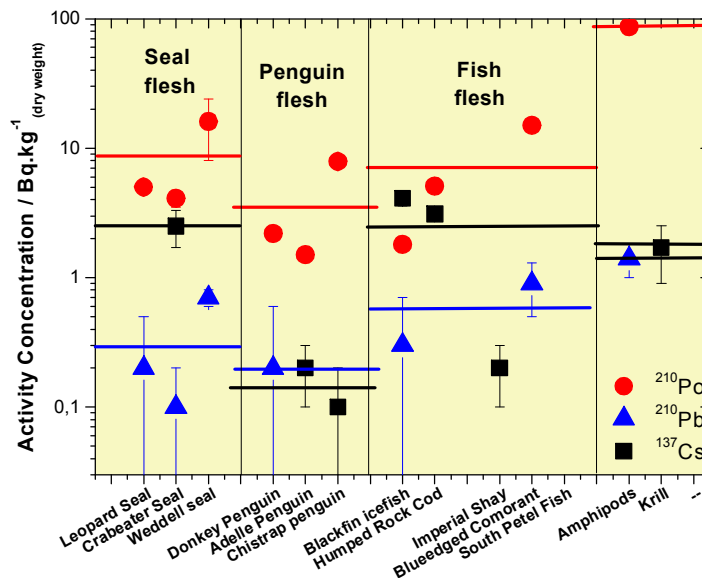


Figure 3-13.

The activity concentration of ¹³⁷Cs, ²¹⁰Po and ²¹⁰Pb in flesh-samples of seals, penguins and fish. To the right data for Amphipods and Krill.

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The Average activity concentration of ^{137}Cs , ^{210}Po and ^{210}Pb was analyzed in samples of various species of seals, penguins and fish are for flesh , liver and kidney as well as Amphipods and Krill are given in **Table 3-5**.

The pattern of average concentration is very similar for seals and fish. For penguins, however, the similar pattern as amphipods do indicate that also these animals might also be an important part of their diet.

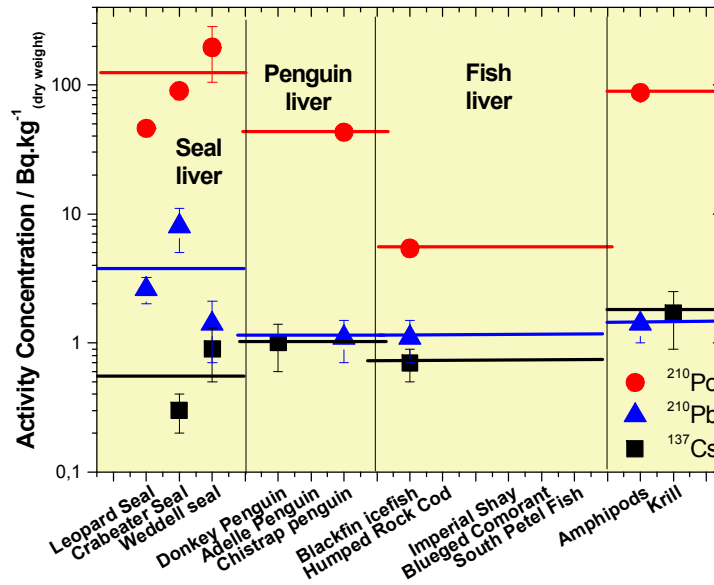


Figure 3-14.

The activity concentration of ^{137}Cs , ^{210}Po and ^{210}Pb in liver-samples of seals, penguins and fish. To the right data for Amphipods and Krill.

The pattern of average concentration in liver do indicate that Amphipods is an important part of fish diet.

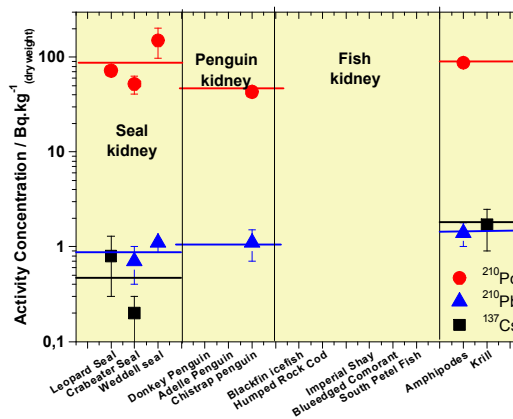


Figure 3-15.

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The activity concentration of ^{137}Cs , ^{210}Po and ^{210}Pb in kidney-samples of seals, penguins and fish. To the right data for Amphipods and Krill.

Table 3-6

The average activity concentration (Bq per kg dry weight) of ^{137}Cs , ^{210}Po and ^{210}Pb in flesh, liver and kidney samples of various species of seals, penguins and fish, as well as in Amphipods and Krill.

	^{137}Cs		^{210}Po		^{210}Pb	
	Bq/kg _{DW}	SE	Bq/kg _{DW}	SE	Bq/kg _{DW}	SE
Flesh						
Seal		2.5 ±0.8		8 ±4		0.3 ±0.2
Penguins		0.2 ±0.1		4 ±2		0.2 ±0.4
Fish		2.5 ±0.1		6.9 ±0.3		0.6 ±0.4
Liver						
Seal		0.6 ±0.3		110 ±44		4 ±2
Penguins		0.7 ±0.2		43 ±2		1.1 ±0.4
Fish				5.4 ±0.3		1.1 ±0.4
Kidney						
Seal		0.5 ±0.3		91 ±30		0.9 ±0.2
Penguins				43 ±2		1.1 ±0.4
Amphipodes						1.4 ±0.4
Krill				87 ±3		
		1.7 ±0.8				

The concentration factor for ^{137}Cs of 15000 estimated for fish in the Antarctic is much higher than the value of 500 given by the IAEA 1985 (IAEA, 1985).

3.5.2 Macro Algae

The activity concentration of ^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Am , ^{137}Cs , ^{210}Po and ^{210}Pb was analyzed in 10 samples of Macro Algae collected during the SEDARP expedition in the Antarctic. The results are displayed in **Figure 3-16**. Generally the activity concentrations in macro algae relatively to the fallout levels are higher in the Antarctic, with a value of 3500 compared to 110 in the Arctic.

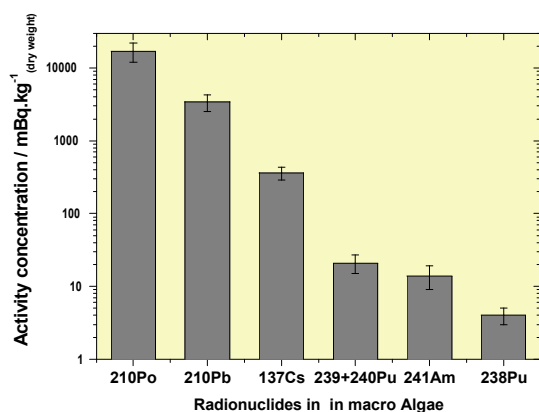


Figure 3-16.

The average activity concentration of ^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Am , ^{137}Cs , ^{210}Po and ^{210}Pb in 10 samples of Macro algae collected during the SWEDARP expedition in the Antarctic

Concentration factors Algae/Water of the radionuclides displayed in **Figure 3-16** was found to be much higher than in the Arctic (Holm et al., 1983). In macro algae we derive a concentration factor for the two regions to 150 and 1800 respectively.

3.6 Conclusions

Radio-caesium, which originates from atmospheric nuclear weapons tests carried out in the late 1950's and the early 1960's shows a long residence time in surface waters of the North and South Atlantic Oceans. The half-time, corrected for physical decay, is estimated to be more than about 100 years. In open sea-water radio-caesium can be regarded as a conservative tracer for evaluating oceanographic processes. In dose commitment estimates for radiological assessment, only physical decay has to be taken into account.

Plutonium isotopes $^{239+240}\text{Pu}$ from atmospheric nuclear weapons tests has a half-life of 7-8 years. In 1989 the average of $^{239+240}\text{Pu}/^{137}\text{Cs}$ - activity ratio in 37 samples of Atlantic surface waters was $(3.3 \pm 0.7) \cdot 10^{-3}$ compared to $12 \cdot 10^{-3}$ in fresh fallout from nuclear weapons tests. The activity ratio shows a minimum at latitudes 30-40 °S. The lower value is due to the higher affinity to sinking particles for Pu. In the southern hemisphere, ^{238}Pu could be found as the result of the 1964 burn-up of a satellite in the atmosphere over the Mozambique Channel.

The Average activity concentrations (Bq/kg_{dw}) of ^{137}Cs are in flesh of seals 2.5, penguins 0.2 and, fish 2.5 in liver of seals 0.6 and penguins 0.7 and in kidneys of seals 0.5. The Average activity concentrations (Bq/kg_{dw}) of ^{210}Po are: in flesh of seals 8, penguins 4 and, fish 7 in liver of seals 110 and penguins 43 and, fish 5, and in kidneys of seals 91 and of penguins 43. The Average activity concentrations (Bq/kg_{dw}) of ^{210}Pb are in flesh of seals 0,3, penguins 0,2, and fish 0,6, in liver of seals 4 and penguins 1 and, fish 1, and in kidneys of seals 1 and of penguins 1. The Average activity concentration of ^{137}Cs in samples of Krill is about 1.7 Bq/kg_{dw} of ^{210}Po and ^{210}Pb are in samples of Amphipods, 87 and 1.4 Bq/kg_{dw} respectively.

The concentration factor for ^{137}Cs of 15000 estimated for fish in the Antarctic is much higher than the value of 500 given by The IAEA 1985. Concentration factors Algae/Water was found to be about 1800 in the Antarctic compared to 100 the Arctic.

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Chapter 4: The Arctic Ocean-91 expedition

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Abstract

The Arctic Ocean expedition in 1991 with the Swedish icebreaker M/S Oden was focused on oceanography and geology. The aim of our project was exploring the activity concentrations in surface air of ^7Be , ^{210}Pb , and ^{210}Po in the surface air, radioactive isotopes of Caesium (^{134}Cs , ^{137}Cs) and plutonium ($^{239+240}\text{Pu}$) in seawater.

During the cruise in the Arctic Ocean during 1991-07-28 to 1991-10-04 the average activity concentrations in surface air of ^7Be was $0.6\pm 0.4 \text{ mBq}\cdot\text{m}^{-3}$, ^{210}Pb $46\pm 34 \text{ }\mu\text{Bq}\cdot\text{m}^{-3}$ and ^{210}Po $37\pm 23 \text{ }\mu\text{Bq}\cdot\text{m}^{-3}$.

The activity concentration of ^{137}Cs in the surface of the Arctic Ocean was in the range of 8-12 $\text{Bq}\cdot\text{m}^{-3}$. When crossing the Nansen basin the activity concentration of ^{137}Cs increased to about 18 $\text{Bq}\cdot\text{m}^{-3}$ at 88 °N 80 °E, and there was an accumulation of ^{137}Cs in an area around at 88 °N and 80-100 °E and locally increased activity at 83 °N 10 °E.

The $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios was about 0.02 due to the contribution mainly from Sellafield and a few percent contribution from Chernobyl. The $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio decreased to about 0.002-0.005 in areas of high ^{137}Cs activity concentration which exclude contribution of ^{134}Cs of nuclear reactor fuel.

The activity concentration of $^{239+240}\text{Pu}$ in the surface of the Arctic Ocean was in the range of 6 - 8 $\text{mBq}\cdot\text{m}^{-3}$. But locally the activity concentration of $^{239+240}\text{Pu}$ was found to be increased to 11 $\text{mBq}\cdot\text{m}^{-3}$ at 86°N 48-53°E, and to 16 $\text{mBq}\cdot\text{m}^{-3}$ at 83°N 10°E.

4.1. Introduction

The Arctic Ocean expedition in 1991 with the Swedish icebreaker M/S Oden (Figure 4-2) was focused on oceanography and geology in the western parts of the Eurasian Basin, the north west Markov Basin and parts of the Barents sea (Josefsson, 1998 , Roos et al., 1998).



Figure 4-1

The expedition logo



Figure 4-2

M/S Oden parked in the Arctic ice.

Photo: Kjell-Åke Carlsson

The route of the expedition is displayed in **Figure 4-3**.

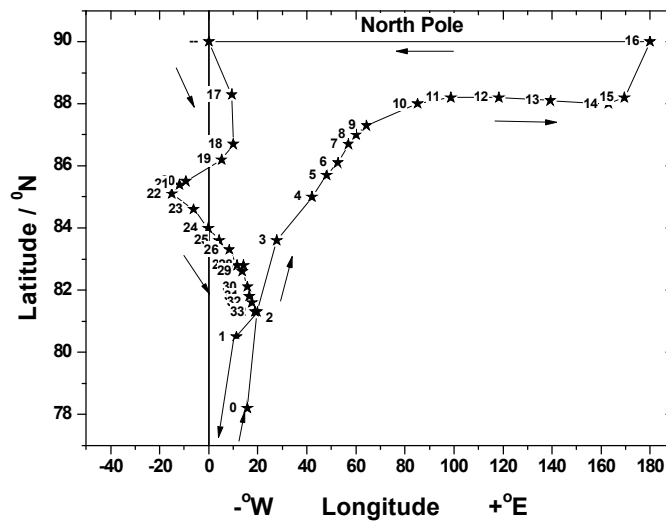


Figure 4-3

Route of the Arctic Ocean -91 expedition with station numbers

In **Table 4-1** is given the present and potential sources of anthropogenic radioactivity of the Arctic Ocean (Aarkrog, 1994). These sources are global fallout from nuclear weapons testing in the

atmosphere, inflow of discharges from nuclear reprocessing in Western Europe, and fallout from the Chernobyl accident. Local fallout from the Novaya Zemlya test site, and discharges from nuclear facilities into the Siberian Rivers, and dumping of nuclear waste into the Barents and Kara Seas are regional sources for contamination of the Arctic Ocean. Because conclusive information is missing, the data given in **Table 4-1** are rough estimates for ^{90}Sr and ^{137}Cs activity discharges to Russian rivers such as Ob, Yenisey, and Lena.

Table 4-1

Summary of the inventories of ^{90}Sr and ^{137}Cs from present and potential sources of anthropogenic radioactivity in the Arctic Ocean (Aarkrog, 1994).

Source	^{90}Sr (PBq = 10^{15} Bq)	^{137}Cs (PBq = 10^{15} Bq)
Global fallout	2.6	4.1
Sellafield discharges	1-2	10-15
USSR river discharges	1-5	1-5
Regional & local fallout		
Run-off of global fallout	1-5	0.5
Chernobyl	0	1.5
TOTAL	6-11	17-30
Source	^{90}Sr (PBq = 10^{15} Bq)	^{137}Cs (PBq = 10^{15} Bq)
Komsomolet submarine, Barents Sea 1989 (Petrov, 1991)	2.9	3.1
Sr-90 powered Lighthouses, Siberian coast (Aarkrog et al., 1994)	10 - 15 per unit	
Dumped submarines at Novaya Zemlya (Yablokov, 1993)	~ 40	~ 45

4.2 ^7Be , ^{210}Pb , and ^{210}Po in surface air

During the Arctic Ocean-91 expedition to the Antarctica, surface air samples of ^{210}Pb , ^{210}Po as well as ^7Be were collected with a so-called “Andersen” air sampling device installed on board the ship M/S Stena Arctica. Air volumes of about 1 500 m³ were collected at each occasion on membrane filters (size 0.25×0.25 m) at a flow rate of 100 m³h⁻¹. The Andersen sampler was compared with a FOA

transportable reference high volume air sampler (Microsorban filter, $0.56 \times 0.56 \text{ m}^2$, $1\ 100 \text{ m}^3 \text{ h}^{-1}$). The samplers were placed close together on deck of the cruise vessel as show in **Figure 4-4**. The ^7Be results for the Anderson sampler were normalized to that of the FOA sampler.

The activity concentrations of ^7Be ($\text{mBq} \cdot \text{m}^{-3}$), ^{210}Pb , and ^{210}Po ($\mu\text{Bq} \cdot \text{m}^{-3}$) measured during 1991 from July 28 to October 4 in the surface air over the Arctic Ocean during 1991 are displayed in **Figure 4-5a-c**.

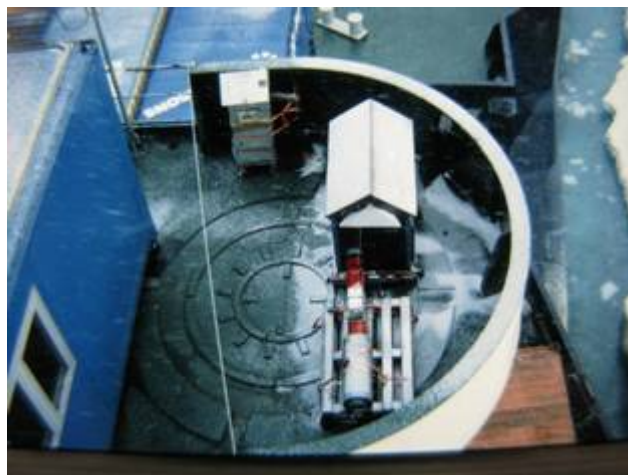


Figure 4-4
The Andersen air- sampler and the FOA transportable reference high volume air sampler placed close together

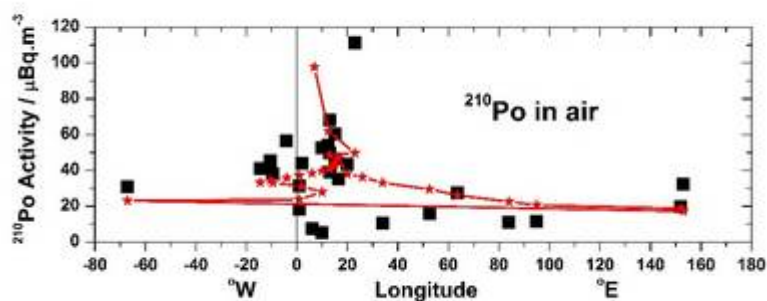


Figure 4-5a
Longitudinal distribution of ^{210}Po activity concentration in air over the Arctic Ocean during 1991.

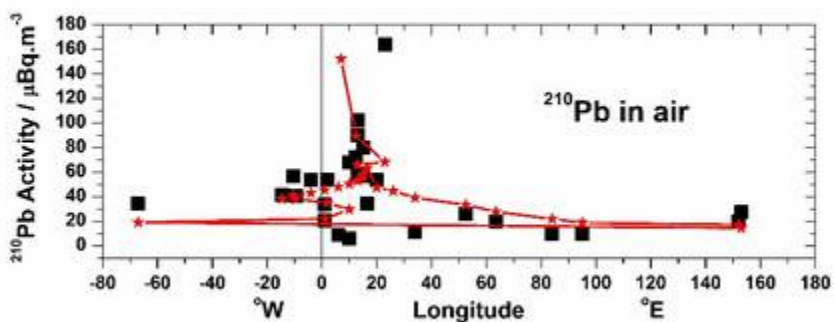


Figure 4-5b

Longitudinal distribution of ^{210}Pb activity concentration in air ^{7}Be , and over the Arctic Ocean during 1991

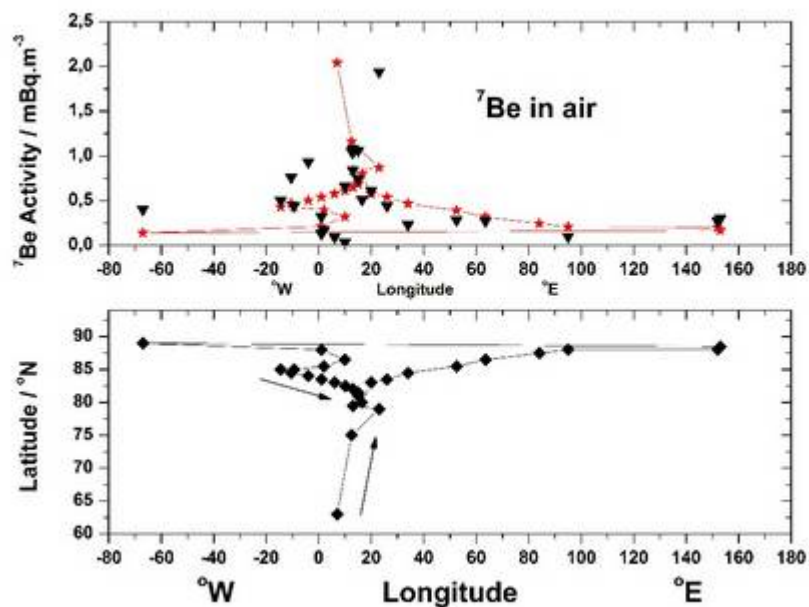


Figure 4-5c

Longitudinal distribution of ^{7}Be activity concentration in air and the route of Arctic Ocean expedition during 1991.

Equations of the PLS model:

$$^{7}\text{Be} [\text{mBq.m}^{-3}] = 6.657 - 0.0733 \times (\text{Latitude } ^{\circ}\text{N}) - 4.66\text{E-}05 \times (\text{Longitude } ^{\circ}\text{E})$$

Goodness of fit statistics $R^2 = 0.395$

$$^{210}\text{Pb} [\mu\text{Bq.m}^{-3}] = 480.919 - 5.215 \times (\text{Latitude } ^{\circ}\text{N}) - 0.033 \times (\text{Longitude } ^{\circ}\text{E})$$

Goodness of fit statistics $R^2 = 0.354$

$$^{210}\text{Po} [\mu\text{Bq.m}^{-3}] = 284.944 - 2.968 \times (\text{Latitude } ^\circ\text{N}) - 0.033 \times (\text{Longitude } ^\circ\text{E})$$

Goodness of fit statistics $R^2 = 0.285$

Table AO91-2

Air concentrations of ^7Be , ^{210}Pb and ^{210}Po in Arctic during 1991-07-28 to 1991-10-04

Isotope	Date	Average	SD	SE
Be-7 A	910728-0906	0.62	0.52	0.14 mBq.m ⁻³
Pb-210 A	910728-0906	49.2	46.2	12.8 μBq.m ⁻³
Po-210 A	910728-0906	36.8	28.5	7.9 μBq.m ⁻³
Be-7 B	910907-1004	0.51	0.33	0.09 mBq.m ⁻³
Pb-210 B	910907-1004	43.8	21.4	5.7 μBq.m ⁻³
Po-210 B	910907-1004	37.6	17.2	4.6 μBq.m ⁻³

During the cruise in the Arctic Ocean during 1991-07-28 to 1991-10-04 the air concentrations of ^7Be was 0.6 ± 0.4 mBq.m⁻³, ^{210}Pb 46 ± 34 μBq.m⁻³ and ^{210}Po 37 ± 23 μBq.m⁻³.

Table AO91-3

Ratios of air concentrations of ^7Be , ^{210}Pb and ^{210}Po in Arctic 1991-07-28 to 1991-10-04

Isotope ratio	Date	Average Ratio	SE
Be-7/A / Pb-210 A	910728-0906	12	4
Be-7 A / Po-210 A	910728-0906	17	5
Be-7 B / Pb-210 B	910907-1004	12	3
Be-7 B / Po-210 B	910907-1004	13	3

4.3 Water Sampling and radioactivity measurements

By using pumps of the ship, samples of surface-water were taken and collected in 200 l vessels in our laboratory accommodated in a container on board. Caesium-134 was added as chemical yield determinant for Caesium. During several hours under continuous stirring with microcrystals of Ammonium Molybdo-Phosphate (AMP- ion exchange crystals, Bio-Rad Laboratories, Canada, Ltd) which had been added to adsorb the dissolved Caesium from the water in the vessels. After the stirring stopped, the AMP crystals were let to be settled over night in the funnel shaped bottom of the vessel. The bottom sediment was tapped into 10 l bottles for transport to Lund where the APM precipitate was separated and measured by high resolution gamma spectrometry (HPGE or Ge-Li) for 1-2 days.

Large volume samples (1000-2000 l) of surface water collected by our own pump by a tube hanging from the rail of the ship. Particulate matter was removed in a 1 μm pre-filter and dissolved Caesium was adsorbed in a cartridge cotton filter impregnated with Copper ferro-cyanide ($\text{Cu}_2[\text{Fe}(\text{CN})_6]$). The filters were dried and transported to Lund, where they were ashed in an oven at 450 °C. The ash was then measured by high-resolution gamma spectrometry (HPGE or Ge-Li) for 1-2 days in order to determine the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in the ocean water.



Figure 4-6

Kjell-Åke Carlsson in the water laboratory container with the 200 l precipitation vessels to his back and the cartridge filter holders on the wall.

After adding ^{242}Pu and ^{243}Am as radiochemical yield determinants, Plutonium and americium isotopes were precipitated by adding sodium hydroxide to 200 l seawater collected in another 200 litre vessel as above, The hydroxide precipitate of was settled over night and the bottom sediment was tapped in 10 l bottles for transport to Lund. Pu and Am isotopes was radio-chemically separated, and electro-deposited on stainless steel disks to be measured by alpha spectroscopy for 3-4 weeks (Holm, 1984).

4.4. Results

4.4.1 ^{137}Cs isotopes in surface water

The Longitudinal distribution of ^{137}Cs activity concentration in surface water and the route of Arctic Ocean expedition are displayed in **Figure 4-7** (Roos et al., 1998).

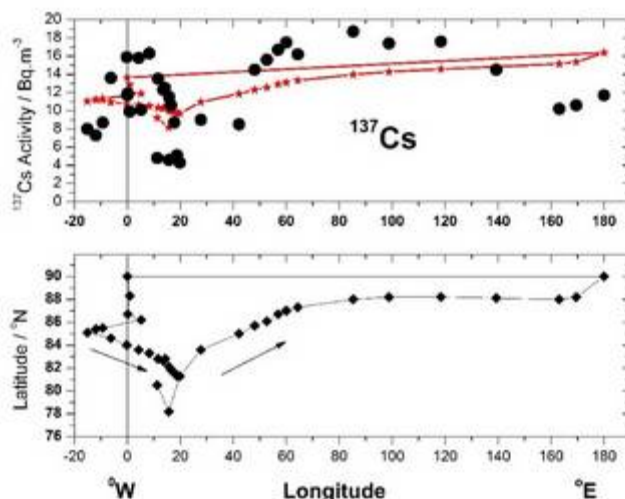


Figure 4-7

Activity concentration of ^{137}Cs in sea-water along the route (Roos et al., 1998). The red curve represent the estimated values of PLS modelling.

Equation of PLS modelling of ^{137}Cs - activity concentration in sea-water along the route of Arctic Ocean-91 expedition:

$$^{137}\text{Cs} [\text{Bq.m}^{-3}] = -30.097 + 0.486 \times (\text{Latitude}^\circ\text{N}) + 0.015 \times (\text{Longitude}^\circ\text{E})$$

Goodness of fit statistics for $^{239+240}\text{Pu}$ resulted in a R^2 value of 0.240

4.4.2 $^{134+137}\text{Cs}$ isotopes in surface water

The Longitudinal distribution of $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios in surface water and the route of Arctic Ocean expedition are displayed in **Figure 4-8** (Roos et al., 1998).

The activity concentration of ^{137}Cs in the Norwegian Coastal Current (NCC) was in the range of 7-8 Bq.m^{-3} with high $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios of 0.02 due to the contribution mainly from Sellafield origin with a few percent contribution from Chernobyl. When the Ship entered the West Spitsbergen Current (WSC) to Fram's Strait the ^{137}Cs activity-concentration dropped to about 4.5-5.5 Bq.m^{-3} with a much lower $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio of about 0.002-0.005. Entering the Polar mixed Layer (PML) at 81 °N the temperature drops from 5 to -2 °C, and the ^{137}Cs activity concentration increased to about 9 Bq.m^{-3} with a $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio of about 0.02.

At the boundary between Nansen and Amundsen basin about 85 °N, there was an increase of the activity concentration of ^{137}Cs up to about 15 Bq.m^{-3} . The passing over the Lomonosov ridge at about 86 °N into the Markov basin, the activity concentration of ^{137}Cs dropped to about 8 Bq.m^{-3} . When crossing the Nansen basin the activity concentration of ^{137}Cs decreased to about 6 Bq.m^{-3} at 88 °N 80 °E. Further east towards the North pole it decreased again to about 12 Bq.m^{-3} . Nevertheless, locally high activity about 16 mBq.m^{-3} was found at 83 °N 10 °E. On the route, southwards close to zero longitude the activity concentration of ^{137}Cs steadily decreased to about 4 Bq.m^{-3} .

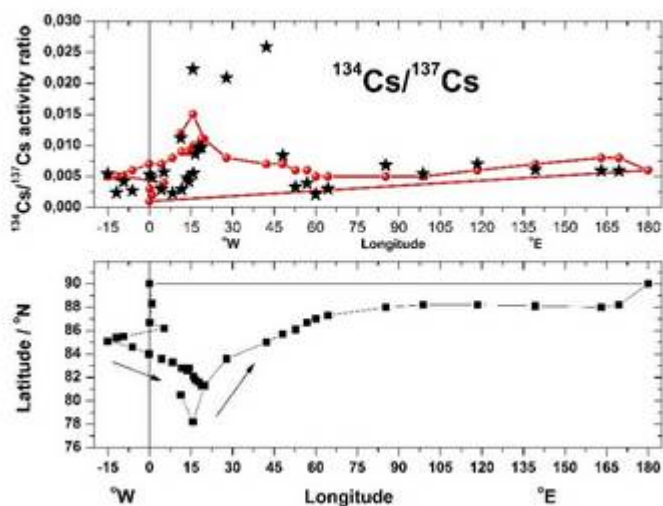


Figure 4-8

$^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in sea water along the route (Roos et al., 1998)

Equation of PLS modelling of $^{134}\text{Cs}/^{137}\text{Cs}$ -activity ratio in seawater along the route of Arctic Ocean-91 expedition:

$$^{134}\text{Cs}/^{137}\text{Cs}\text{-activity ratio} = 0.111 - 0.00124 \times (\text{Latitude}^\circ\text{N}) + 3.83 \cdot 10^{-5} \times (\text{Longitude}^\circ\text{E})$$

Goodness of fit statistics for $^{239+240}\text{Pu}$ resulted in a R^2 value of 0.219.

4.4.3 Depth profiles of ^{137}Cs activity concentration

The depth profiles of ^{137}Cs are displayed in Figure 4-9. The activity concentration ($\text{Bq}\cdot\text{m}^{-3}$) values are fitted to a first order exponential decrease with depth (d, m).

Nansen station 83.6°N , 30.29°E profile :

$$A_{\text{Cs-137}} [\text{Bq}\cdot\text{m}^{-3}] = 0.07 + 8.7 \cdot \exp(-0.0012 \cdot d);$$

$$R^2=0.99; d_{1/2}= 562 \text{ m}$$

Amundsen station 87.5°N , 106.44°E profile:

$$A_{\text{Cs-137}} [\text{Bq}\cdot\text{m}^{-3}] = 0.37 + 14.9 \cdot \exp(-0.0015 \cdot d);$$

$$R^2=0.97; d_{1/2}= 464 \text{ m}$$

The ^{137}Cs activity concentration at the surface of the Amundsen 87.5°N , 106.44°E profile is, however, slightly elevated compared to the Nansen station 83.6°N , 30.29°E profile. This is likely due to Atlantic water of high ^{137}Cs activity concentration flow into the Arctic interior. Concentrations in the Atlantic layer (at temperature maximum) are almost the same at both stations and very close to those reported for Atlantic water by others (Cochran et al., 1995). In samples taken in 1979, and 1983, however, values below 1 mBq/l has been reported (Livingston et al., 1984, Smith et al., 1998).

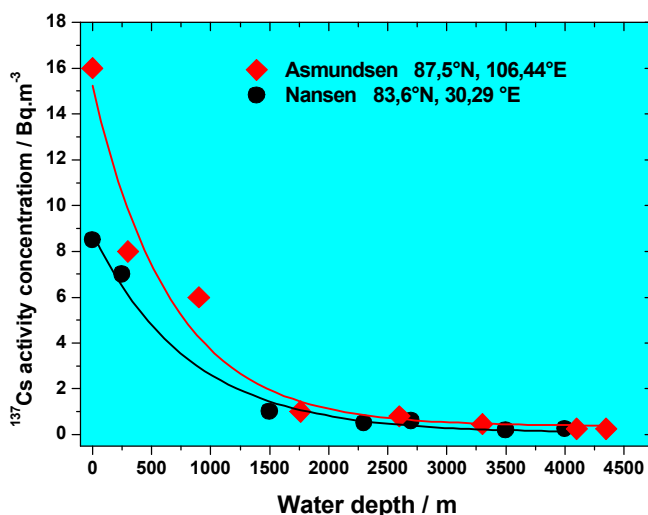


Figure 4-9

Depth profiles of ^{137}Cs activity concentration

The presently high values reported during later years in the Atlantic layer is thus likely an effect of Sellafield releases. In the bottom water at both stations the ^{137}Cs concentrations are about 0.3 Bq.m^{-3} . This is similar values as previously has been reported for the Norwegian Sea deep water (Wedekind et al., 1997).



Figure 4-10

Kjell-Åke Carlsson with M/S Oden at the North pole.

4.4.4 $^{239+240}\text{Pu}$ isotopes in surface water

The Longitudinal distribution of $^{239+240}\text{Pu}$ activity concentration in surface water and the route of Arctic Ocean expedition are displayed in **Figure 4-11** (Roos et al., 1998). The activity concentration of $^{239+240}\text{Pu}$ in the Norwegian Coastal Current (NCC) West Spitsbergen Current (WSC) to Frams Strait was in the range of $6 - 7 \text{ mBq.m}^{-3}$. Entering the Polar mixed Layer (PML) at 81°N the temperature drops from 5 to -2°C , and the $^{239+240}\text{Pu}$ activity concentration increased to about 10 Bq.m^{-3} . At the boundary between Nansen and Amundsen basin about 85°N , there is a decrease of the activity concentration of $^{239+240}\text{Pu}$ to about $7 - 8 \text{ mBq.m}^{-3}$. The passing over the Lomonosov ridge at about 86°N into the Markov basin, the activity concentration of $^{239+240}\text{Pu}$ was about 8 mBq.m^{-3} . When crossing the Nansen basis the activity concentration of $^{239+240}\text{Pu}$ was about 8 mBq.m^{-3} at 88°N 80°E . Further east towards the North pole the activity concentration of $^{239+240}\text{Pu}$ was still about 6 m Bq.m^{-3} . But locally high activity of $^{239+240}\text{Pu}$ up to 16 Bq.m^{-3} was found at 83°N 10°E . On the route southwards close to zero longitude the activity concentration of $^{239+240}\text{Pu}$ rapidly decreased to about 6 mBq.m^{-3} .

The high plutonium concentrations found when moving south across the Nansen Basin could be an effect of the convection when moving closer to Svalbard across the Nansen Basin (Rudels et al.,

1996). The convection brings up water with higher Pu-concentrations originating from the Atlantic layer mixing with surface water.

Equation of PLS modelling of $^{239+240}\text{Pu}$ activity concentration in sea water along the route of Arctic Ocean-91 expedition:

$$^{239+240}\text{Pu} [\text{mBq}\cdot\text{m}^{-3}] = 71.366 - 0.745 \times (\text{Latitude}^\circ\text{N}) + 0.012 \times (\text{Longitude}^\circ\text{E})$$

Goodness of fit statistics for $^{239+240}\text{Pu}$ resulted in a R^2 value of 0.279.

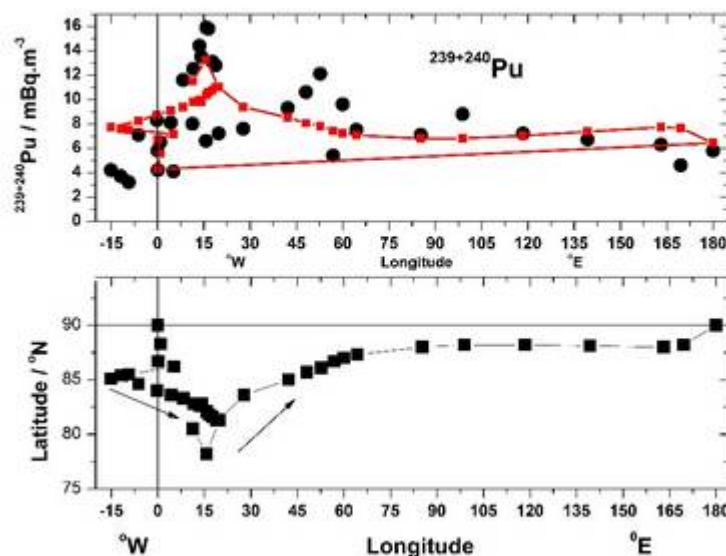


Figure 4-11

$^{239+240}\text{Pu}$ activity concentration in sea water along the route. (Roos et al., 1998) The red curve represent the estimated values of PLS modelling.

4.5 Conclusions

During the cruise in the Arctic Ocean during 1991-07-28 to 1991-10-04 the air concentrations of ^7Be was $0.6 \pm 0.4 \text{ mBq}\cdot\text{m}^{-3}$, ^{210}Pb $46 \pm 34 \text{ }\mu\text{Bq}\cdot\text{m}^{-3}$ and ^{210}Po $37 \pm 23 \text{ }\mu\text{Bq}\cdot\text{m}^{-3}$.

The activity concentration of ^{137}Cs in the Arctic Ocean water was in the range of $8\text{-}12 \text{ Bq}\cdot\text{m}^{-3}$. The activity concentration of ^{137}Cs increased to about $18 \text{ Bq}\cdot\text{m}^{-3}$ when crossing the Nansen basin at 88°N 80°E . There was an accumulation of ^{137}Cs in an area around at 88°N and $80\text{-}100^\circ\text{E}$ and locally high activity at 83°N 10°E .

The $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios was about 0.02 due to the contribution mainly from Sellafield and a few percent contribution from Chernobyl. The $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio decreased to about 0.002-0.005 in areas of high ^{137}Cs activity concentration that exclude an origin of nuclear reactor fuel.

The activity concentration of $^{239+240}\text{Pu}$ in the Arctic Ocean was in the range of 6-8 $\text{mBq}\cdot\text{m}^{-3}$ with locally high $^{239+240}\text{Pu}$ activity concentration of 11 $\text{mBq}\cdot\text{m}^{-3}$ at 86 °N 48-53 °E, and 16 $\text{mBq}\cdot\text{m}^{-3}$ at 83 °N 10 °E.

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Chapter 5: The Tundra-94 expedition

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Abstract

The joint Swedish-Russian “*Tundra Ecology-94*” expedition during 1994 used the large Russian ice-breaking research vessel R/V Akademik Fedorov a platform and went along a coastline of 3500 km—from the Kola Peninsula 10°E to Kolyuchinskaya Bay 173°E. Radioactivity in air, seawater and sediment was explored at various locations along the route.

The average of ⁷Be activity concentration in air over the Arctic Ocean was found to be only about 0.6 mBq.m⁻³, in air close to the Siberian coast-line, however, it was as high as 11 mBq.m⁻³. The activity concentration of ²¹⁰Pb in the air over the Arctic Ocean varies between 37 – 176 μBq.m⁻³. In the air close to the Siberian coastline 71°N 84°E, however, the activity concentration of ²¹⁰Pb in the air was much higher, about 2500 μBq.m⁻³.

Anthropogenic radioactivity in the Arctic originate from nuclear weapons fallout, release from nuclear fuel reprocessing plant, and from the Chernobyl accident. The minimum values of the ¹³⁷Cs activity concentration water along the route of the Tundra were found in South-eastern Barents Sea: 5.3 Bq.m⁻³ of surface-water, and of bottom-water 6.4 Bq.m⁻³. Maximum values were found in the Western Laptevsea: 12.8 Bq.m⁻³ of surface-water, and of bottom-water 5.1 Bq.m⁻³. East of 150 °E the ¹³⁴Cs / ¹³⁷Cs ratios are less than 0.003, indicating that less than 6% of the ¹³⁷Cs originated from the Chernobyl accident.

¹³⁷Cs levels are reduced to values of about 1.4 Bq.m⁻³ in the low salinity water near the mouths of the Ob and Yenisey Rivers. The ¹³⁴Cs / ¹³⁷Cs activity ratio of 0.014 in the freshwater indicates that the Chernobyl component in the river systems is the same (30%) as in the marine waters west of 150 °E.

In surface water the ⁹⁰Sr activity concentration range from 2 to 4 Bq.m⁻³, Maximum values about 3.5 Bq.m⁻³ were found between 100-140 °E. But east of 150 °E the values decreased to about 0.5 Bq.m⁻³ at 170 °E. In bottom water the ⁹⁰Sr activity concentration range from 1.5 at 40 °E to maximum values about 4 Bq.m⁻³ between 100-120 °E. The measured ⁹⁰Sr/¹³⁷Cs ratios in surface water close to a value of 0.14 over a wide range of stations from the Barents to the Laptev Seas. The ¹²⁹I concentration in sea-water along the route of the Tundra expedition decrease from about 20·10¹¹ atoms.l⁻¹ at 40 °E, to about 1·10¹¹ atoms.l⁻¹ east of 160 °E.

The ²³⁹⁺²⁴⁰Pu activity concentration in surface seawater decrease from about 10 mBq.m⁻³ to about 1 mBq.m⁻³ east of 160 °E. In bottom seawater it is more evenly distributed between 10⁻⁴ mBq.m⁻³, with minimum at 60-80 °E and maxima at 40°E and 160 °E. Measured ²³⁸Pu/^{239,240}Pu activity ratios in the water column yield no evidence of any leakage of plutonium from dumped nuclear wastes in the Kara and Barents Seas.

5.1 Introduction

The joint Swedish-Russian “*Tundra Ecology-94*” expedition during 1994 along a coastline of 3500 km—from the Kola Peninsula 10°E to Kolyuchinskaya Bay 173°E, used the large Russian ice-breaking research vessel R/V Akademik Fedorov a platform (**Figure 5-1**). In **Table 5-1** is given locations of the various places where we were transferred a shore with helicopters (**Figure 5-6**) or escorted by on Russian atomic Icebreaker (**Figure 5-2**). In **Figure 5-3** is given a diagram of the route of the expedition.



Figure 5-1a.

R/V Akademik Fedorov in the harbour of Gothenburg loading of the Tundra-94 expedition



Figure 5-1b.

R/V Akademik Fedorov, ready to leave for the Arctic

Radioactivity exploration from the Arctic to the Antarctic.

Table 5-1

Expedition route and research sites of the "Tundra Ecology-94" expedition

Visiting date 1	On return date 2	Site No.	Name	Position
06-04	09-08		Gothenburg	57.43°N 11.98°E
06-08>>09	09-02		Murmansk	68.57°N 44.10°E
06-10	08-31	1(27)	Kachkovsky Bay. Kola Peninsula	67.30°N 41.00°E
06 12>>13	08-29>>30	2(26)	North-Eastern Kanin Peninsula	68.15°N 6.00°E
06-14>>15	08-26>>28	3(25)	Kolguyev Island	69.15°N 50.00°E
06-15>>16	08-25>>26	4(24)	Pechora Bay	68.5°N 54.00°E
06-17>>20	08-22>>08-23	5(23)	Western Yamal Peninsula	70.45°N 67.00°E
06-21>>22	08-20>>21	6 (22)	Belyi Island. Northern Yamal Peninsula	73.00°N 70.00°E
06-22>>23	08-18>>19		Dickson	
06-23		7	Arctic Institute Islands	75.00°N. 82.00°E
06-24>>26	08-15>>17	8(21)	North West of Taymyr Peninsula	76.00°N 94.00°E
06-27>>28	08-13>>14	9(20)	Chelyuskin Peninsula	77.20°N 102.00°E
08-29>>30	08-10>>11	10(19)	North-east of Taymyr Peninsula	76.00°N 112.00°E
07-03>>05			Khatanga rotation point 1	74.00°N 110.00°E
07-05>>06		11	Olenekskiy Bay	73.15°N 120.00°E
	08-08>>08-08		Tiksi rotation point 2	74.00°N 110.00°E
	08-03>>08-04	12	Yana Delta	71.30°N 136.00°E
07-10>>11	07-31>>08-02	13	New Siberian Islands	75.00°N 140.00°E
07-14>>15		14	Lopatka Peninsular, N-W Indigirka	71.45°N 149.00°E
07-17>>18		15	North-east of Kolyma Delta	71.45°N 158.00°E
07-20>>21		16	Ayon Island	69.50°N 168.00°E
07-22>>26		17	South-western Wrangels Island	70.50°N 179.00°E
07-25	point of return	18	Kolyuchinskaya Bay	67.00°N 173.45°E



Figure 5-2.

The Russian atomic icebreaker approaching to assist Akademik Fedorov in the heavy ice.

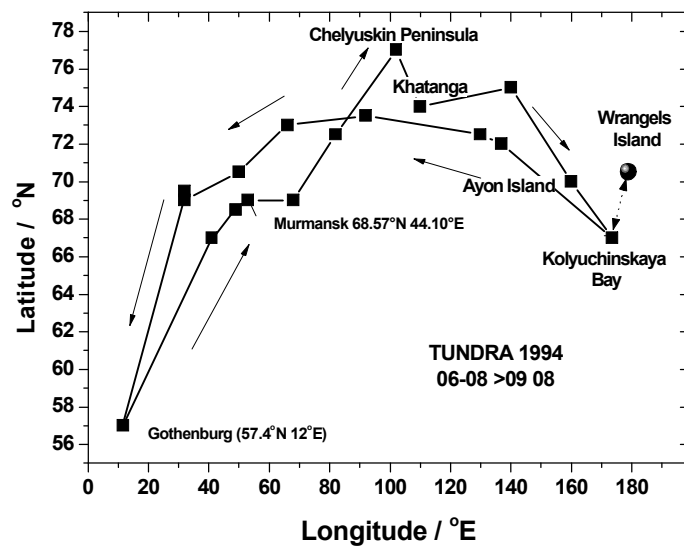


Figure 5-3.

Route of the Tundra-94 expedition with R/V Akademik Fedorov along the Siberian Coastline, with a helicopter tour to Wrangles Island.

5.2 Material and Methods.

5.2.1 Air sampling

Air filter samples were taken by using an Andersen sampler with the capacity of $100 \text{ m}^3\text{h}^{-1}$ (filter size $0.25 \times 0.25 \text{ m}$ membrane filter). An air volume of about $1\,500 \text{ m}^3$ were collected at a rate of $100 \text{ m}^3\text{h}^{-1}$ during each sampling occasion. The Andersen sampler was placed close together with a FOA transportable reference high volume air sampler (filter size $0.56 \times 0.56 \text{ m}$. microsorban filter. $1\,100 \text{ m}^3\text{h}^{-1}$), previously taken part in an inter calibration of samplers (Vintersved, 1994). The filters were stored on board and then taken the institute at Lund for analysis. The results of the Anderson sampler were normalized to those of the calibrated FOA sampler based on ^7Be measurements. FOA nowadays FOI, is a Swedish research institute in the areas of defence and security.



Figure 5-4.

The FOA air sampler

5.2.2 Analysis of the air filters

The filters were measured for ^7Be by gamma spectrometry using a high performance Germanium detector (HPGe Canberra). After adding ^{209}Po as radiochemical yield determinant, the samples were wet-ashed by using a mixture of concentrated nitric and per-chloric acids. Polonium was spontaneously

deposited on nickel discs, and measured by alpha spectrometry, using surface ion implanted silicon detectors.

Remaining trace of Polonium, was removed by anion exchange. The solution was then stored for about 8 months to allow equilibrium in-growth of ^{210}Po from ^{210}Pb . The activity of ingrown ^{210}Po , was then analysed as described above, and finally the activity concentrations of ^{210}Po and ^{210}Pb in air were calculated.

5.2.3 Water sampling

Large volume (200 litre) water samples, were taken from the water cooling system of the ship, and collected in special vessels for precipitation of ^{137}Cs and $^{239+240}\text{Pu}$. Continuous sampling of caesium also took place with a separate pump and a pipe hanging from the rail of the ship to about 2 m depth. An in-line system with filters impregnated with Copper-Ferro-Cyanide ($\text{Cu}_2\text{Fe}(\text{CN})_6$) was used to collect Caesium isotopes from the seawater. The filters were dried and brought to Lund for radiochemical analysis. After ashing the filters at 420 °C, the residues were analysed for ^{134}Cs and ^{137}Cs by using a Ge (Li) gamma spectrometer.



Figure 5-5.

Interior of the water laboratory container, with the two 200 litre precipitation vessels to the right and the cartridge filters on the wall to the left

Radioactivity exploration from the Arctic to the Antarctic.



Figure 5-6.

Bertil Persson and Kjell-Åke Carlsson landed on the tundra after a tour with the Russian helicopter in the back.



Figure 5-6a.

View of the tundra



Figure 5-6b.

Sampling of the tundra



Figure 5-6a.

Closer view



Figure 5-7a.

Chelyuskin Peninsula 77.20°N; 102.00°E

Summer lake of the waste accumulated during the winter.



Figure 5-7a.

Road at Chelyuskin Peninsula



Figure 5-7a.

A mound of flat stones raised by Adolf Erik Nordenskiöld's expedition in 1878 as a memorial of the visit.



Figure 5-7b.

An anchor left by Adolf Erik Nordenskiöld's expedition in 1878.

5.3 Results

5.3.1 ^{210}Pb and ^7Be in air 1994- June 08 > September 08

In the **Figures 5-8a** and **b** are given the activity concentrations of ^{210}Pb ($\mu\text{Bq.m}^{-3}$), and ^7Be (mBq.m^{-3}) in air, measured during 1994-June 8 < September 8 at the joint Swedish-Russian Tundra Ecology-94 expedition.

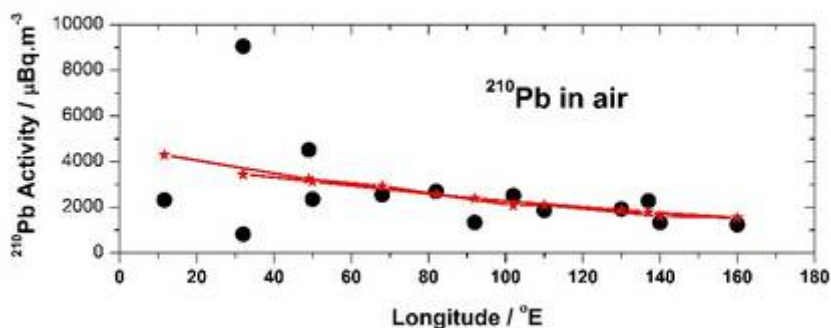


Figure 5-8 a

Longitudinal distribution of ^{210}Pb air concentration ($\mu\text{Bq.m}^{-3}$) during 1994-June 8 < September 8 at the Tundra Ecology-94 expedition. The red line shows the predicted

Radioactivity exploration from the Arctic to the Antarctic.

values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

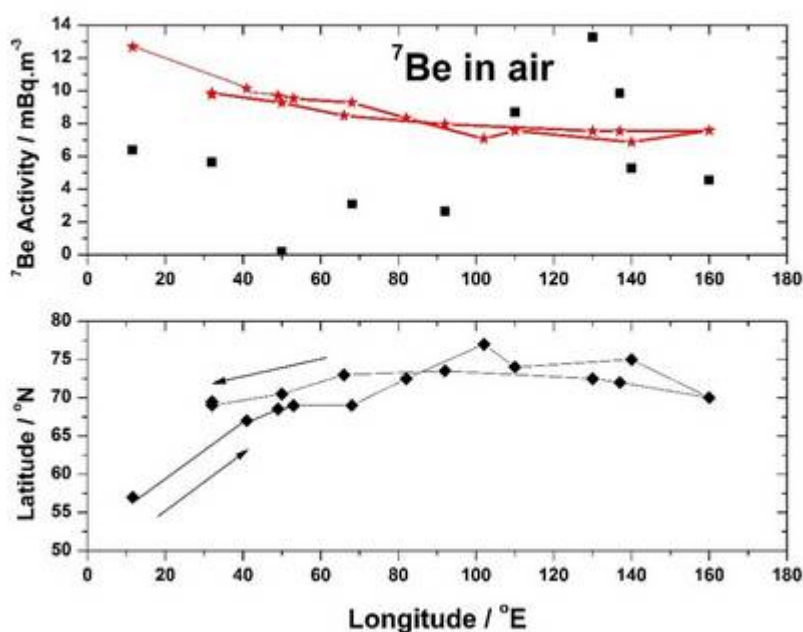


Figure 5-8 b Longitudinal distribution of ⁷Be air concentration (mBq.m⁻³) during 1994-June 8 < September 8 and the route of the Tundra Ecology-94 expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

Equations of the PLS models for the air concentrations displayed in red in Figure 5-8:

$$C_{7\text{Be}} = 24.65 - 0.207 \cdot \text{Latitude} - 0.016 \cdot \text{Longitude}; [\text{mBq} \cdot \text{m}^{-3}]$$

Goodness of fit statistics (Variable $C_{7\text{Be}}$): $R^2 = 0.036$

$$C_{210\text{Pb}} = 6992 - 44.45 \cdot \text{Latitude} - 14.61 \cdot \text{Longitude}; [\mu\text{Bq} \cdot \text{m}^{-3}]$$

Goodness of fit statistics (Variable $C_{210\text{Pb}}$): $R^2 = 0.176$

Table 5-2a

Average air concentrations of ⁷Be and ²¹⁰Pb measured during 1994-June 8 > September 8 at the joint Swedish-Russian Tundra Ecology-94 expedition.

Isotope	Date	Average	SD	SE	
Latitude		71	4	1	N
Longitude		84	47	11	E
⁷ Be	1994-0605 > 0719	11.4	9.0	3.2	mBq/m ³
²¹⁰ Pb	1994-0605 > 0719	2373	1029	364	μBq/m ³
⁷ Be	1994-07-19 > 0908	7.2	5.4	2.0	mBq/m ³
²¹⁰ Pb	1994-07-19 > 0908	2712	2854	1079	μBq/m ³

During the Swedish-Russian Tundra Ecology-94 expedition along the Siberian coastline, the average air concentrations of ^7Be and ^{210}Pb measured during May-July were 11 ± 3 and $2.4 \pm 0.4 \text{ mBq.m}^{-3}$ respectively and during July-September they were 7.2 ± 2 and $2.7\pm 1.1 \text{ mBq.m}^{-3}$ respectively.

Table 5-2 b

Ratios of average air concentrations of ^7Be and ^{210}Pb measured during 1994-June 8 > September 8 at the joint Swedish-Russian Tundra Ecology-94 expedition.

Isotope ratio	Date	Average	SE
Latitude		71	1
Longitude		84	11
$^7\text{Be} / ^{210}\text{Pb}$	1994-0605 > 0719	5	2
$^7\text{Be} / ^{210}\text{Pb}$	1994-07-19 > 0908	3	1

5.3.2. ^{137}Cs activity concentration in seawater

The ^{137}Cs activity concentration water along the route of the Tundra expedition is shown in figures 5-9a and b respectively. The minimum values were found in South-Eastern Barents Sea: 5.3 Bq.m^{-3} of surface-water, and of bottom-water 6.4 Bq.m^{-3} . Maximum values were found in the Western Laptevsea: 12.8 Bq.m^{-3} of surface-water, and of bottom-water 5.1 Bq.m^{-3} .

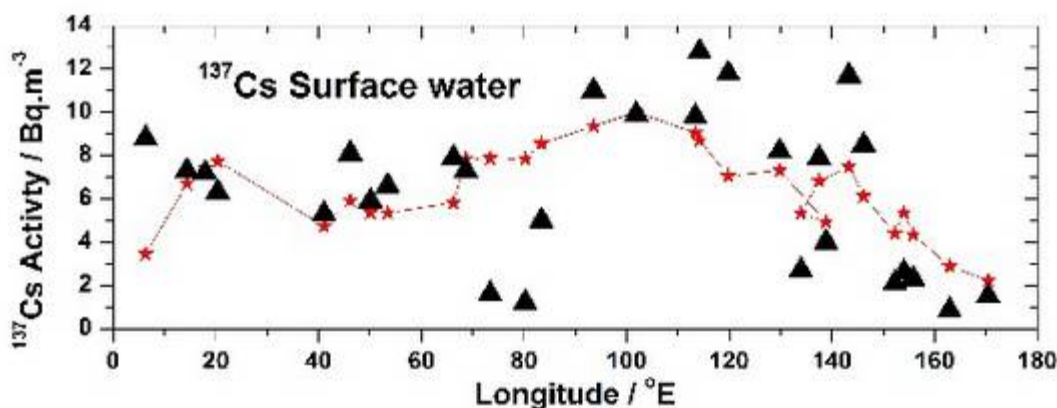


Figure 5-9a. Longitudinal distribution of ^{137}Cs activity concentration in surface seawater along the route of Tundra expedition. Predicted values in read. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

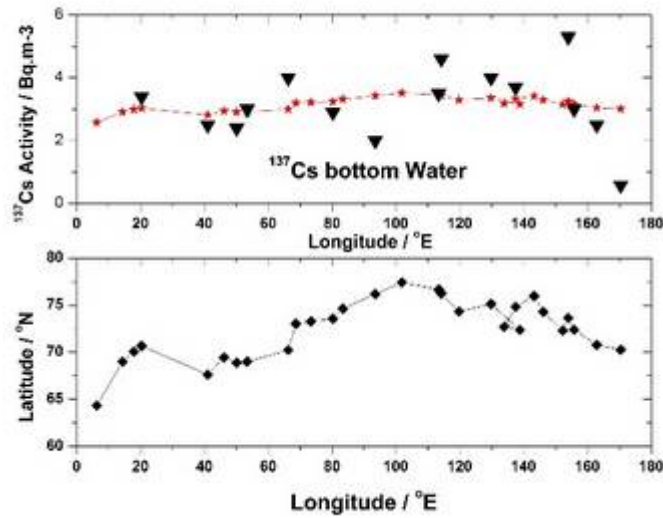


Figure 5-9b. Longitudinal distribution of ^{137}Cs activity concentration in bottom seawater and the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

Equation of the PLS model for activity concentration in surface water (SW), displayed in red in **Figure 5-9**:

$$^{SW}\text{C}_{137\text{Cs}} = -44.93 + 0.756 \cdot (\text{Latitude}^\circ\text{N}) - 0,035 \cdot (\text{Longitude}^\circ\text{E}); [\text{Bq.m}^{-3}]$$

Goodness of fit statistics : $R^2 = 0,312$

Equation of the PLS model for bottom water (BW):

$$^{BW}\text{C}_{137\text{Cs}} = -2.005 + 0.071 \cdot (\text{Latitude}^\circ\text{N}) + 4,49 \cdot 10^{-5} \cdot (\text{Longitude}^\circ\text{E}); [\text{Bq.m}^{-3}]$$

Goodness of fit statistics (Variable $^{BW}\text{C}_{137\text{Cs}}$ bottom water): $R^2 = 0,076$



Figure 5-10a.
View of the Arctic sea



Figure 5-10b.
View of the Arctic sea

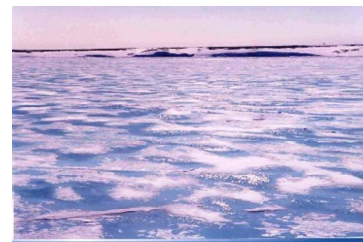


Figure 5-10c.
View of the Arctic sea

5.3.3 $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in sea water

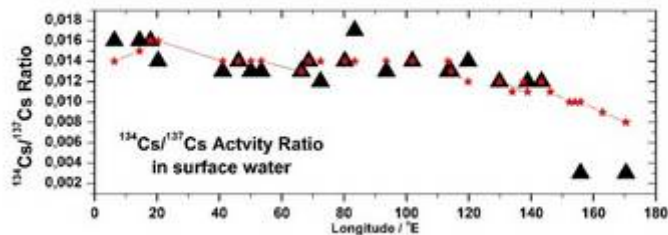


Figure 5-11a.

Longitudinal distribution of $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in surface sea-water along the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

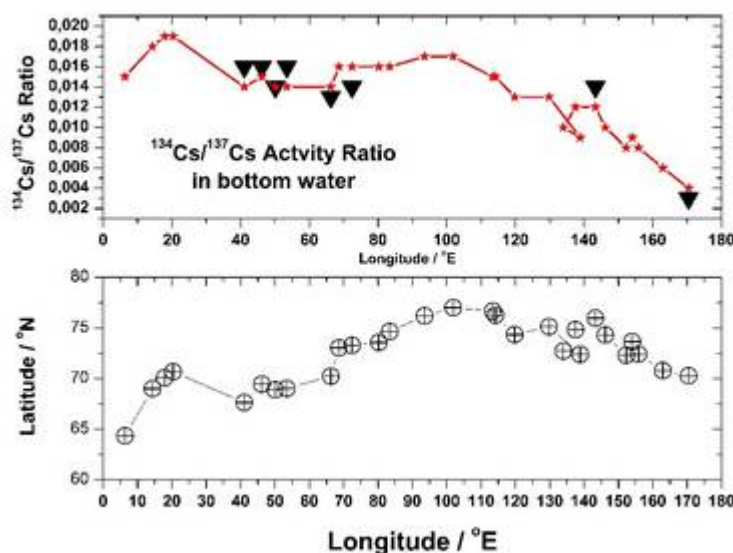


Figure 5-11b.

Longitudinal distribution of $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in bottom sea water along the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

Equation of the PLS model for $^{134}\text{Cs}/^{137}\text{Cs}$ ratio in surface water (SW):

$$(^{134}\text{Cs}/^{137}\text{Cs})_{\text{SW}} = -0.011 + 3.98 \cdot 10^{-4} \cdot (\text{Latitude } ^\circ\text{N}) - 4.85 \cdot 10^{-5} \cdot (\text{Longitude } ^\circ\text{E})$$

Goodness of fit statistics (Variable $^{134}\text{Cs}/^{137}\text{Cs}$ Surface water): $R^2 = 0.447$

Equation of the model for bottom water (BW):

$$(^{134}\text{Cs}/^{137}\text{Cs})_{\text{BW}} = -0.042 + 8.89 \cdot 10^{-4} \cdot (\text{Latitude } ^\circ\text{N}) - 9.37 \cdot 10^{-5} \cdot (\text{Longitude } ^\circ\text{E})$$

Goodness of fit statistics (Variable $^{134}\text{Cs}/^{137}\text{Cs}$ Bottom water): $R^2 = 0.913$

5.3.4 ^{90}Sr activity concentration in seawater

The ^{90}Sr activity concentration in seawater along the route of the Tundra expedition is shown in **figures 5-13a and b** respectively. In surface water the ^{90}Sr activity concentration range from 2 to 4 $\text{Bq}\cdot\text{m}^{-3}$. Maximum values about 3.5 $\text{Bq}\cdot\text{m}^{-3}$ were found between 100-140 $^\circ\text{E}$. But east of 150 $^\circ\text{E}$, the values decreased to about 0.5 $\text{Bq}\cdot\text{m}^{-3}$ at 170 $^\circ\text{E}$.

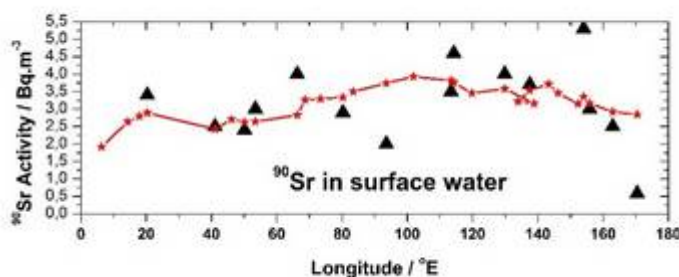


Figure 5-12a Longitudinal distribution of the ^{90}Sr activity concentration in surface seawater along the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

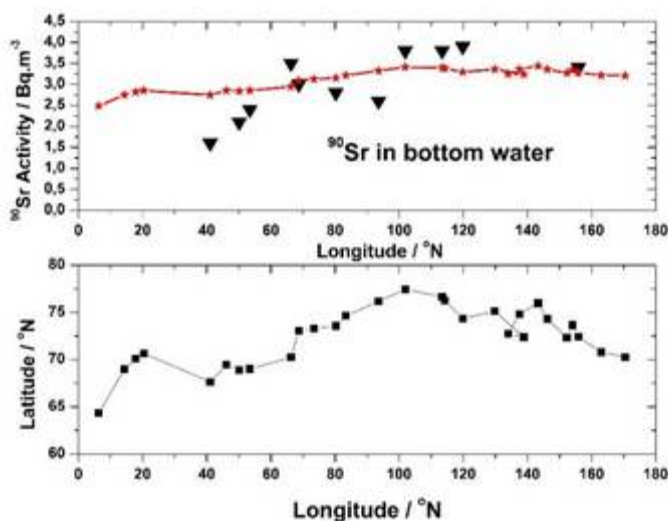


Figure 5-12b Longitudinal distribution of the ^{90}Sr activity concentration in surface and bottom seawater along the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

In bottom water the ^{90}Sr activity concentration range from 1.5 at 40 $^\circ\text{E}$ to maximum values about 4 $\text{Bq}\cdot\text{m}^{-3}$ between 100-120 $^\circ\text{E}$.

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Equation of the PLS model for ^{90}Sr activity concentration in Surface water (SW):

$$^{SW}C_{90\text{Sr}} = -7.910 + 0.153 \cdot (\text{Latitude}^\circ\text{N}) + 9.6 \cdot 10^{-5} \cdot (\text{Longitude}^\circ\text{E}); [\text{Bq}\cdot\text{m}^{-3}]$$

Goodness of fit statistics: $R^2 = 0.16$

Equation of the PLS model ^{90}Sr activity concentration in Bottom Water (BW):

$$^{BW}C_{90\text{Sr}} = -0.873 + 0.052 \cdot (\text{Latitude}^\circ\text{N}) + 0.0025 \cdot (\text{Longitude}^\circ\text{E}); [\text{Bq}\cdot\text{m}^{-3}]$$

Goodness of fit statistics: $R^2 = 0.21$

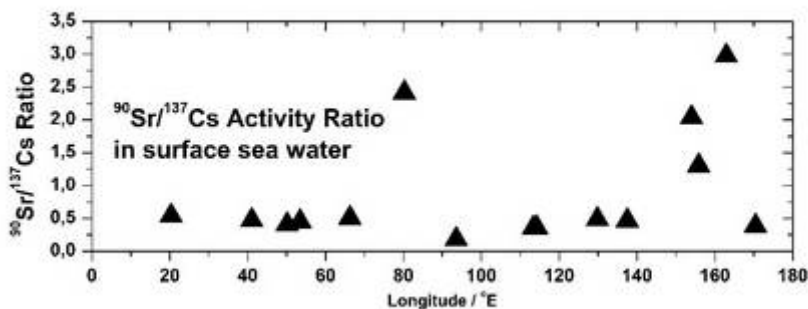


Figure 5-13a Longitudinal distribution of the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratio in surface seawater along the route of Tundra expedition.

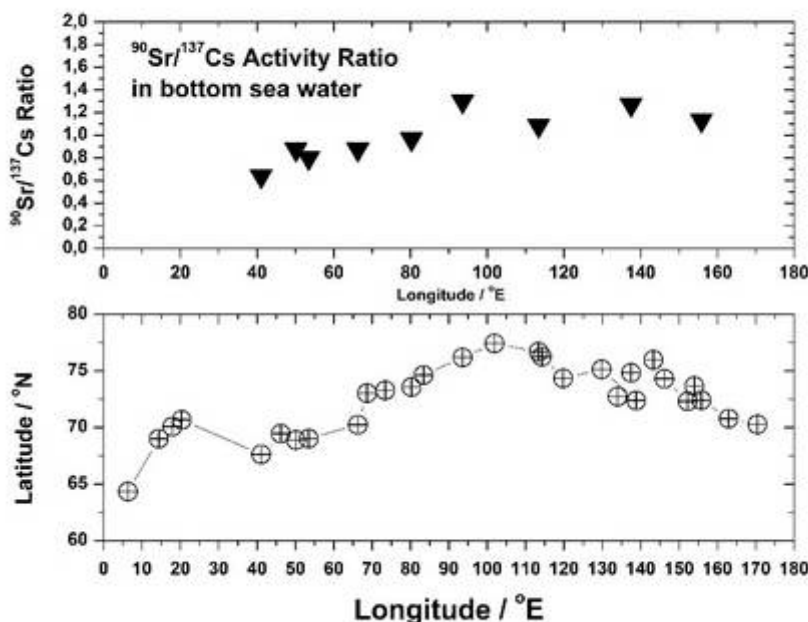


Figure 5-13b Longitudinal distribution of the $^{90}\text{Sr}/^{137}\text{Cs}$ activity ratio in bottom seawater and the route of Tundra expedition.

5.3.5 ^{129}I concentration in seawater

The ^{129}I concentration in sea-water along the route of the Tundra expedition is shown in **Figure 5-14**. The concentration decrease from about $20 \cdot 10^{11} \text{ atoms.l}^{-1}$ ($2 \cdot 10^{15} \text{ atoms.m}^{-3}$), ≈ 3 pico-molar, to about $1 \cdot 10^{11} \text{ atoms.l}^{-1}$ east of 160°E .

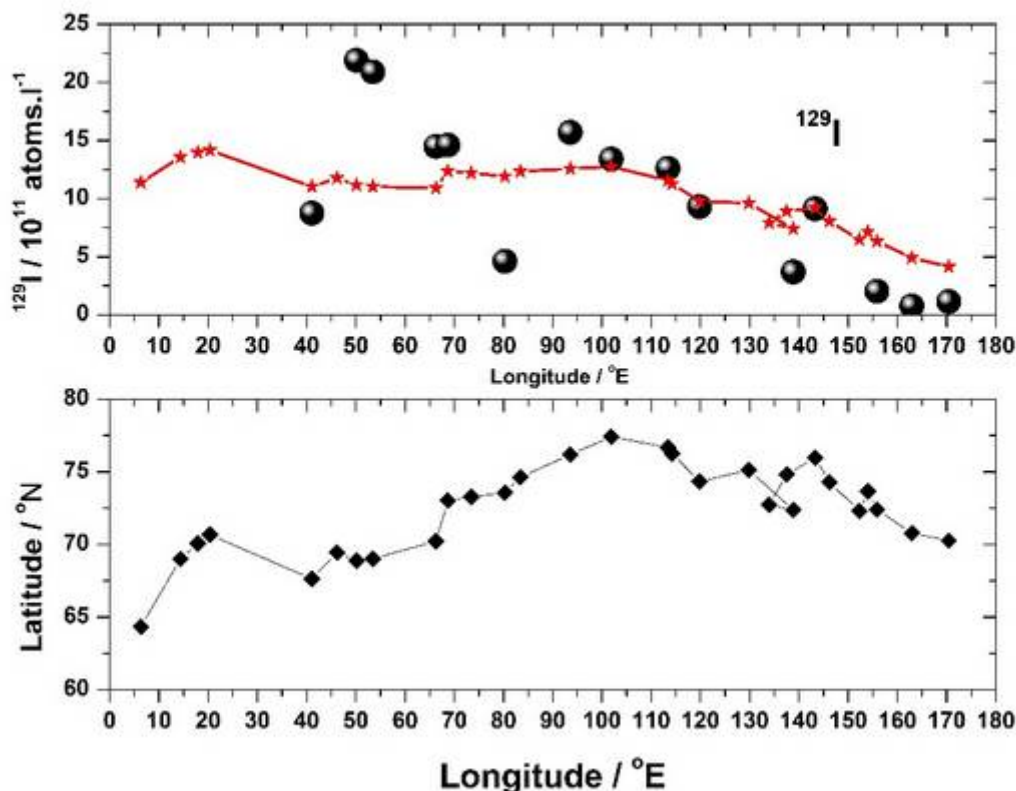


Figure 5-14 Longitudinal distribution of ^{129}I concentration in surface seawater and the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

PLS Model parameters for ^{129}I concentration in surface seawater :

$${}^{\text{SW}}\text{C}_{129\text{I}} = -25.529 + 0.580 \cdot (\text{Latitude } ^\circ\text{N}) - 0.065 \cdot (\text{Longitude } ^\circ\text{E}); [10^{14} \text{ atoms.m}^{-3}]$$

Goodness of fit statistics: $R^2 = 0.311$

5.3.6 $^{239+240}\text{Pu}$ activity concentration in seawater

The $^{239+240}\text{Pu}$ activity concentration in surface and bottom seawater along the route of Tundra expedition are given in **Figure 3-15a** and **b** respectively. In surface seawater the $^{239+240}\text{Pu}$ activity concentration decrease from about 10 $\text{mBq}\cdot\text{m}^{-3}$ to about 1 $\text{mBq}\cdot\text{m}^{-3}$ east of 160 °E.

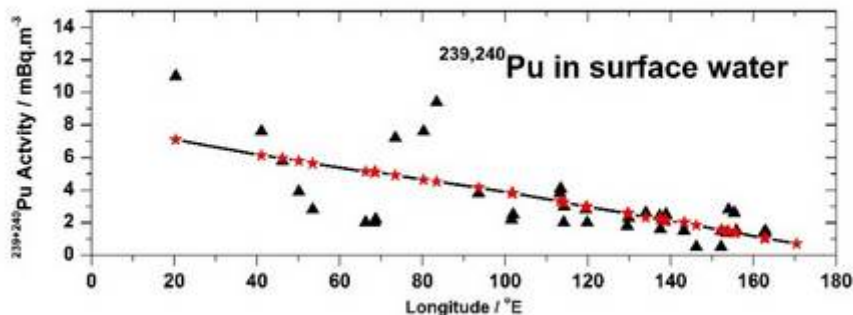


Figure 5-15a.

$^{239+240}\text{Pu}$ activity concentration in surface seawater along the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

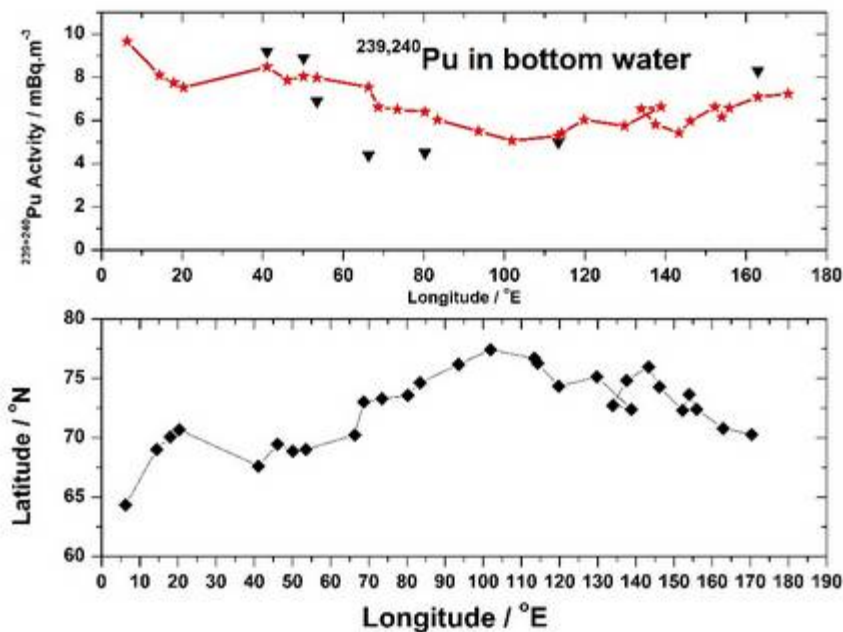


Figure 5-15b.

$^{239+240}\text{Pu}$ activity concentration in bottom seawater and the route of Tundra expedition. The red line shows the predicted values from PLS regression modelling of the data with Longitude and Latitude as explanatory X-values.

In bottom seawater the $^{239+240}\text{Pu}$ activity concentration is more evenly distributed between 10 - 4 mBq.m⁻³, with the minimum at 60-80 °E and maxima at 40°E and 160 °E.

Equation of the model PLS model for activity concentration of $^{239,240}\text{Pu}$ in Surface water (SW):

$${}^{\text{SW}}C_{239,240\text{Pu}} = 5.948 + 0.028 \cdot (\text{Latitude}^\circ\text{N}) - 0.043 \cdot (\text{Longitude}^\circ\text{E}) ; [\text{mBq}\cdot\text{m}^{-3}]$$

Goodness of fit statistics (Variable ${}^{\text{SW}}C_{239,240\text{Pu}}$): $R^2 = 0.471$

Equation of the PLS model for $^{239,240}\text{Pu}$ Bottom water (BW):

$${}^{\text{BW}}C_{239,240\text{Pu}} = 30.96 - 0.331 \cdot (\text{Latitude}^\circ\text{N}) - 0.0028 \cdot (\text{Longitude}^\circ\text{E}) ; [\text{mBq}\cdot\text{m}^{-3}]$$

Goodness of fit statistics (Variable ${}^{\text{BW}}C_{239,240\text{Pu}}$): $R^2 = 0.432$

5.3.7 ^{137}Cs and $^{239+240}\text{Pu}$ activity concentration in sediment

The integrated sediment activity of $^{234+240}\text{Pu}$ was measured in samples taken at sampling sites specified in **Table 5-3**, and the results are displayed in **Figure 5-16a** and **b**.

Table 5-3

Sampling location and integrated sediment activity of ^{137}Cs and $^{234+240}\text{Pu}$ at the Tundra

Station	Latitude		Longitude		Water depth m	Activity penetration depth cm	^{137}Cs		$^{234+240}\text{Pu}$	
	°N	min	°E				SD	[Bq/m ²]	SD	[Bq/m ²]
9	70	14	66	17	20	19	556	67	28.8	3.3
14	76	11	93	34	55	8	217	11		
18	75	8	129	50	58	9	393	17		
21	74	50	137	32	20	14	836	60	34.3	3.4
28	70	16	170	26	30	7	119	8	4.94	0.4

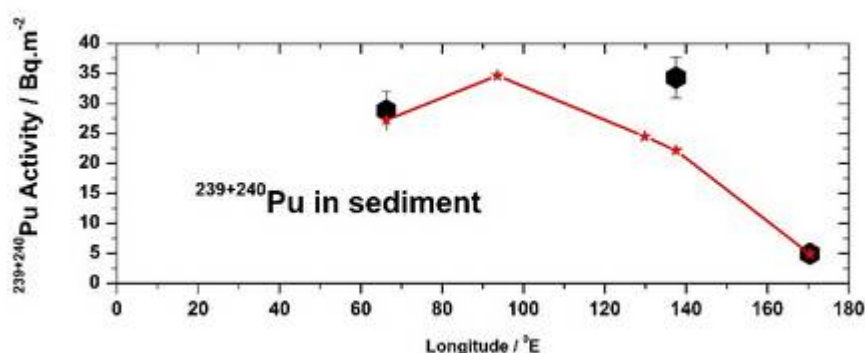


Figure 5-16a

Measured and predicted, integrated sediment activity of $^{234+240}\text{Pu}$ at specific sampling stations (see below).
Predicted values in red

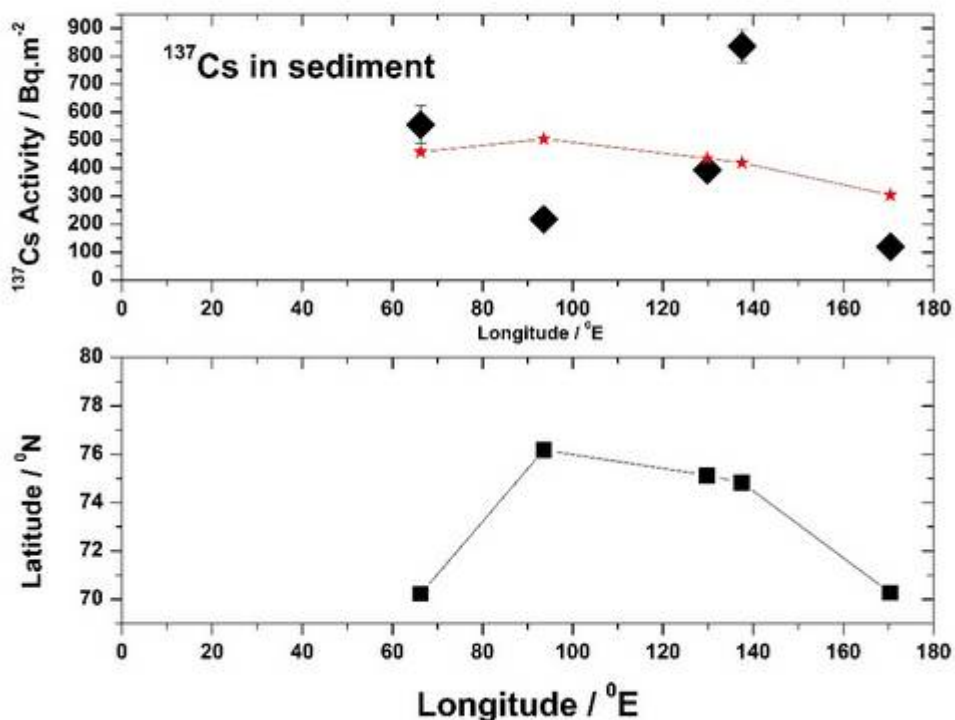


Figure 5-16

Measured and predicted integrated sediment activity of ^{137}Cs at specific sampling stations, given in the bottom diagram with latitude versus longitude. Predicted values in read

Equation of the model for predicted integrated sediment activity, $^{\text{SED}}A$, of $^{239+240}\text{Pu}$ [Bq/m^2] in sediment:

$$^{\text{SED}}A_{239+240\text{Pu}} = -115.2 - 0,215 \cdot (\text{Longitude}^{\circ}\text{E}) + 2.23 \cdot (\text{Latitude}^{\circ}\text{N})$$

Goodness of fit statistics ($^{\text{SED}}A_{239+240\text{Pu}}$): $R^2 = 0,852$

Equation of the PLS model for predicted integrated sediment activity, $^{\text{SED}}A$, of ^{137}Cs [$\text{Bq}\cdot\text{m}^{-2}$] in sediment (SED):

$$^{\text{SED}}A_{137\text{Cs}} = -473.3 - 1.486 \cdot (\text{Longitude}^{\circ}\text{E}) + 14.66 \cdot (\text{Latitude}^{\circ}\text{N}); [\text{Bq}\cdot\text{m}^{-2}]$$

Goodness of fit statistics (Variable ^{137}Cs [Bq/m^2]): $R^2 = 0.069$

5.4. Discussions

5.4.1. Beryllium-7 activity concentrations in the Arctic air

The activity-concentration of ^7Be in air in the Arctic air as summarized in **Table 5-4** varies between 2 - 4.9 mBq.m^{-3} with average $2.8 \pm 0.3 \text{ mBq.m}^{-3}$ (Buraglio et al., 2001, Kulan, 2006, Paatero and Hatakka, 2000, Baskaran and Shaw, 2001, Dibb and Jaffrezo, 1993). The average of ^7Be activity concentration in air over the Arctic Ocean was, however, only about 0.6 mBq.m^{-3} . In contrast the activity concentration of ^7Be in air close to the Siberian coast-line as high as 11 mBq.m^{-3} (Persson, 2013).

Table 5-4. Summary of atmospheric ^7Be concentrations in Arctic and sub-Arctic air

Time	Location	Lat	Long	Be-7		Reference
		N+; S-	E+; W-	Arithm. Mean	SD	
910728-0906	Arctic Ocean	82.07	51.00	0.62	0.52	This work ^{*)}
910907-1004	Arctic Ocean	84.36	-2.32	0.51	0.33	This Work ^{*)}
940605-0908	N Siberian coast	71	84	11.4	9.0	This work ^{*)}
2000	Uppsala. Sweden	59.88	17.63	4.7	2.3	(Buraglio et al., 2001)
1972-1995	Sweden	59.88	17.63	4.8	2.4	(Buraglio et al., 2001)
1972-2003	Sweden. Kiruna	67.84	20.34	1.9	1.0	(Kulan, 2006)
1972-2003	Sweden. Grindsjön	59.07	17.82	2.3	1.2	(Kulan, 2006)
1972-2003	Sweden. Ljungbyhed	56.08	13.23	2.5	1.3	(Kulan, 2006, Aldahan et al., 2008)
1995- 1997	Finland Sodankyla	67.37	26.65	2.5	2.0	(Paatero, 2000)
1996	Alaska USA:Poker Flat	65.13	-147.48	3.0	2.0	(Baskaran and Shaw, 2001)
1996	Alaska USA Eagle	65.9	-141.20	2.2	1.0	(Baskaran and Shaw, 2001)
1988-1989	Dye3	65.18	43.82	2.6	1.1	(Dibb and Jaffrezo, 1993)
1988-1990	Barrow	71.30	-156.77	1.9	1.1	(Dibb and Jaffrezo, 1993)
1988-1991	Kap Toban	70.42	-21.97	2.4	1.3	(Dibb and Jaffrezo, 1993)
1988-1992	Nord	81.36	-16.40	2.5	1.4	(Dibb and Jaffrezo, 1993)
1988-1993	Thule	77.50	-69.33	3.7	1.9	(Dibb and Jaffrezo, 1993)
1997-2004	Summit. Greenland	72.575	-27.55	2.0	0.5	(Dibb, 2007)

^{*)}(Persson, 2013).

5.4.2 ^{210}Pb activity concentrations in the Arctic air

Observations of the activity concentration of ^{210}Pb in the air over the Arctic ocean as summarized in **Table 5-5**, varies between 37 – 176 $\mu\text{Bq.m}^{-3}$ (Persson and Holm, 2013, McNeary and Baskaran, 2003, Dibb and Jaffrezo, 1993, Dibb, 2007, Paatero et al., 2003, Samuelsson et al., 1986). In 1991 we found the average activity concentration of ^{210}Pb over the Arctic Ocean to be $40 \pm 4 \mu\text{Bq.m}^{-3}$. In the air close to land masses the activity concentration of ^{210}Pb in the air increase to 269- 2712 $\mu\text{Bq.m}^{-3}$ (McNeary

and Baskaran, 2003, Baskaran and Shaw, 2001, Dibb and Jaffrezo, 1993); with the highest values of about 2500 $\mu\text{Bq.m}^{-3}$ at the Siberian coastline (Persson and Holm, 2013).

Table 5-5. Activity concentrations ($\mu\text{Bq.m}^{-3}$) of ^{210}Pb recorded at different locations during the Tundra-94 expedition.

Time	Location	Lat °N	Long °E	Pb-210		Po-210		Reference
				Average $\mu\text{Bq.m}^{-3}$	SD	Average $\mu\text{Bq.m}^{-3}$	SD	
940605>0703	Siberian Tundra	71	84	2373	364	2044	870	This work
940704>0908	Siberian Tundra	71	84	2712	1079	2336	994	This work

5.4.3 ^{137}Cs activity distribution

The minimum values of the ^{137}Cs activity concentration water along the route of the Tundra were found in South-eastern Barents Sea: 5.3 Bq.m^{-3} of surface-water, and of bottom-water 6.4 Bq.m^{-3} . Maximum values were found in the Western Laptev sea: 12.8 Bq.m^{-3} of surface-water, and of bottom-water 5.1 Bq.m^{-3} .

East of 150 °E the $^{134}\text{Cs} / ^{137}\text{Cs}$ ratios are less than 0.003, indicating that less than 6% of the ^{137}Cs originated from the Chernobyl accident.

The $^{134}\text{Cs} / ^{137}\text{Cs}$ activity ratio of 0.014 in the freshwater indicates that the Chernobyl component in the river systems is the same (30%) as in the marine waters.

5.4.4 ^{90}Sr activity distribution

The relative magnitudes of ^{90}Sr inputs to the Arctic Ocean differ from those for ^{137}Cs for the same sources. The $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratio of 35 reported for Chernobyl fallout was sufficiently high that ^{90}Sr inputs from this source can be considered to be negligible (Aarkrog, 1988). The present fallout concentration in the oceans is assumed to be about 1.6 Bq/m^3 (Dahlgard, 1995). Sellafield represents a major ^{90}Sr source term, which similar to ^{137}Cs , attained a maxima in the late 1970's and has decreased substantially since that time. An important additional source of ^{90}Sr to the Siberian seas is associated with river runoff from fallout, discharges from nuclear reprocessing plants and inputs from accidental releases of ^{90}Sr , such as the Khystym accident on 29 September 1957 at Mayak, USSR (Lollino et al., 2014). The greater mobility of ^{90}Sr compared to ^{137}Cs in freshwater environments results in reduced ^{90}Sr residence times in soils and more rapid transport through the drainage basin to marginal seas. By using the record of reported ^{90}Sr discharges, transport times of less than 10 y and transfer factors of 10 Bq.m^{-3} per PBq.a^{-1} the Sellafield contribution to Barents Sea water is estimated to be approximately 0.5 Bq.m^{-3} in 1994 (Gray et al., 1995). The addition of a fallout component of approximately 1.6 Bq.m^{-3} is not sufficient to give the values ($> 3 \text{ Bq.m}^{-3}$) measured in the Kara and Laptev Seas. These results suggest an additional contribution of the order of 1-2 Bq.m^{-3} to ^{90}Sr concentrations on the Siberian shelves. Contributions from riverine sources will generally only play a minor role since most the salinities are too high.

The $^{90}\text{Sr}/^{137}\text{Cs}$ fallout ratio in seawater is approximately 0.7 while Sellafield discharge results give an average cumulative decay corrected $^{90}\text{Sr}/^{137}\text{Cs}$ ratio of 7 between 1980 and 1992 (Gray et al., 1995, Dahlgaard et al., 1995). The measured $^{90}\text{Sr}/^{137}\text{Cs}$ (non-Chernobyl) ratios in surface water (**Figure 5-13**) are also close to a value of 0.14 over a wide range of stations from the Barents to the Laptev Seas, despite the observation above that much of this signal is from Sellafield. Clearly, the ^{90}Sr input from the Russian river systems has been sufficiently large to reduce the $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratio to values similar to fallout levels. Bottom waters show slightly higher ratios indicating a Sellafield contribution. Calculations as above applying known transfer factors and transport times reveal, however, that direct transport will not notably effect the fallout ratio. Instead the Sellafield activity must be of an older date, reflecting a longer half-life on the shelf than expected, or recirculated from the central Arctic Ocean.

5.4.5 ^{129}I distribution

The decreasing gradient in ^{129}I activity, east of the Barents Sea to the Laptev Sea, reflects the general increase in ^{129}I discharges since the 1980's. The sharp decrease in ^{129}I concentrations at 150 °E indicates that the front between Atlantic and Pacific origin water has been encountered. In the 1980s, this front was located over the Lomonosov Ridge, but has shifted to its present position over the Mendelyev Ridge (McLaughlin et al., 1996, Smith et al., 1998). The relatively low radionuclide levels measured in the East Siberian Sea are typical of fallout values associated with Pacific-origin water transported into this region from the Bering Sea.

5.4.6 $^{239+240}\text{Pu}$ activity distribution

Plutonium activity concentrations and isotopic ratios, measured along the Siberian Shelf and in the Central Arctic Ocean, indicate that it mainly originates from global fallout of atmospheric nuclear weapons tests. This demonstrate that plutonium fallout of atmospheric nuclear weapons tests, deposited at mid-latitudes in the North Atlantic in the late-1950s and early-1960s, have found their way to the Arctic interior and beyond (Kershaw and Baxter, 1995, Josefsson, 1998);Herrmann, 1998 #473}.

Measured $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratios in the water column yield no evidence of any leakage of plutonium from dumped nuclear wastes in the Kara and Barents Seas. Were leakage of plutonium to occur in the future from dumped nuclear wastes in the Kara and Barents Seas, it is likely that some of it will be transported through the Eurasian Shelf and into the Central Arctic with the Transpolar Drift, on a timescale of one to two decades, eventually exiting the Arctic through Fram Strait.

The geographical distribution of plutonium indicate that a broad peak that appears to have passed through the North Pole recently. We attribute this peak to the plutonium 'signal' that entered the Arctic following the period of maximal fallout deposition referred to above. The distribution is consistent with the well-established pattern of water-mass circulation in the Arctic, bearing in mind the limited number of plutonium observations available.

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Chapter 6: The Arctic Ocean-96 expedition

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Abstract

The Swedish Arctic Ocean-96 expedition arranged by the Swedish Polar Research Committee took place during 1996 with the Swedish icebreaker M/S Oden. The expedition focused on studying the distribution of radionuclides in different water masses in the central Arctic Ocean. The expedition crossed the Barents Sea, entered the Nansen Basin at the St. Anna Through, and continued north across the Amundsen Basin. The main part of the expedition was concentrated on the north Lomonosov Ridge and the return route passed the North Pole and went south along 10 °E towards Svalbard. In both legs, water samples in the surface and subsurface layers were collected for the determination of fission products and transuranic elements in seawater and sediment.

The highest ¹³⁷Cs, ⁹⁰Sr and ¹²⁹I activities and ¹³⁴Cs/¹³⁷Cs activity ratios are found in a band stretching from the northern Lomonosov Ridge, over the North Pole and south along 10-15 °E to 85 °N. The releases from European nuclear fuel reprocessing plants and Chernobyl fallout traced by the ¹²⁹I and ¹³⁴Cs/¹³⁷Cs signals respectively have approximately the same spatial distribution in the Arctic Ocean surface layer. The activity concentration of ⁹⁰Sr in surface water is about 2 Bq.m⁻³. For ²³⁹⁺²⁴⁰Pu the lowest concentrations in the surface water of 4 mBq.m⁻³ was found on the Lomonosov Ridge and in the Makarov Basin. In the Eurasian Basin, the concentrations ²³⁹⁺²⁴⁰Pu were about 13 mBq.m⁻³.

For all the radionuclides analysed, the water profiles generally show activities decreasing with depth. An exception is the high ¹³⁷Cs activity concentrations found in the lower halocline layer at the Lomonosov Ridge and Makarov Basin stations. It is assumed to be due to a contribution of Chernobyl fallout to the Arctic Ocean surface layers in the years around 1990. Such maxima were not observed in the ⁹⁰Sr or ¹²⁹I measurements.

Inventories down to 900 m depth reveal that between 60-70 % of the ¹³⁷Cs, ⁹⁰Sr and ¹²⁹I in the Arctic Ocean water, are present in the surface layer inflow from the Atlantic. The total inventories down to 900 m in the Eurasian Basin of the Arctic Ocean estimate to 6.7 PBq for ¹³⁷Cs, 3.4 PBq for ⁹⁰Sr and 5.3x10²⁸ atoms for ¹²⁹I.

6.1 Introduction

The Swedish Arctic Ocean-96 expedition took place during 1996 with the Swedish icebreaker M/S Oden and arranged by the Swedish Polar Research Secretariat. Our part of the expedition projects focused on studying the distribution of radionuclides in different water masses in the central Arctic Ocean. The expedition crossed the Barents Sea, entered the Nansen Basin at the St. Anna Through, and continued to the North across the Amundsen Basin. The main part of the expedition was concentrated on the north Lomonosov Ridge and the return route passed the North Pole and went south along 10 °E towards Svalbard. Samples for the determination of fission products and transuranic elements in seawater and sediment, were collected in both the surface and subsurface layers. The route of the expedition and the sampling locations at the Arctic Ocean-96 expedition is displayed in **Figure 6-1**.

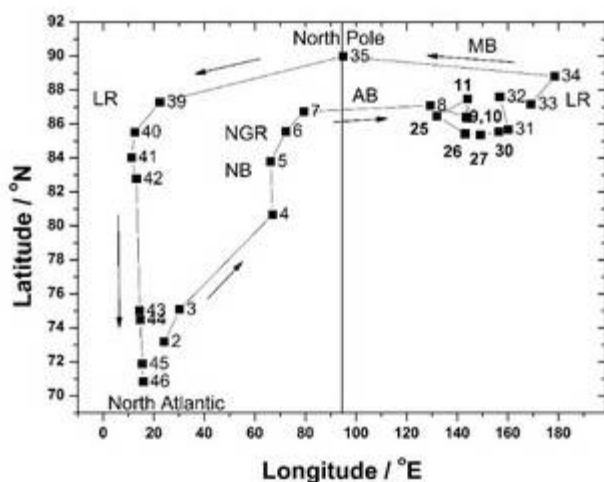


Figure 6-1.

The sampling point along the route of the Arctic Ocean 96 expedition Makarov Basin MB, Lomonosov Ridge LR, Amundsen Basin AB, Nansen Gakkel Ridge NGR, Nansen Basin NB.



Figure 6-2

The Swedish icebreaker M/S Oden stuck in the ice in the Arctic Ocean. *(Photo Bertil Persson)*

6.2 Material and Methods

6.2.1 Sampling and radioactivity measurements of ^{134}Cs and ^{137}Cs

Samples of surface-water, taken by pumps of the ship, were collected in 200 l vessels in our laboratory accommodated in a container on board. Caesium-134 was added as chemical yield determinant for caesium. Then microcrystals of Ammonium molybdo-phosphate (AMP-1 Ion Exchange Crystals, Bio-Rad Laboratories, Canada, Ltd) added to the water in the vessels under continuous stirring during several hours adsorb the dissolved Caesium. The AMP crystals settled over night in the funnel shaped bottom of the vessel. The sediment was then tapped into 10 l bottles for transport to Lund, where the APM crystals were separated and measured by high resolution gamma spectrometry (HPGe or Ge-Li) for 1-2 days.



Figure 6-3
The Arctic Ocean water and ice
(Photo Bertil Persson)



Figure 6-4
Dan Josefsson managing the Go Flow bottle with 100 l seawater sample on the back of M/S Oden.
(Photo Bertil Persson)

Large volume samples (1000-2000 l) of surface water collected by our own pump by a hose hanging from the railing of the ship. Particular matter were adsorbed in a 1 µm pre-filter and dissolved Caesium in a cartridge cotton filter impregnated with Copper Ferro-cyanide ($\text{Cu}_2[\text{Fe}(\text{CN})_6]$). The filters were dried and for transport to Lund, where they turned to ash in an oven at 450 °C. The ash measured by high-resolution gamma spectrometry (HPGe or Ge-Li) for 1-2 days determined the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in the Ocean water.

After adding ^{242}Pu and ^{243}Am as radiochemical yield determinants, all Pu and Am isotopes were precipitated by adding sodium hydroxide pellets to 200 l seawater. The precipitate settled over night, and then tapped in 10 l bottles for transport to Lund. Pu and Am isotopes was radio-chemically separated, and deposited on stainless steel disks to be measured by alpha spectroscopy for 3-4 weeks (Holm, 1984).

In the central Arctic Ocean, subsurface samples were collected at various depths with a 100 litre Go-Flow bottle. One sample was taken at depths between 235-340 m where temperature maximum occur, and another at depths between 850-1000 m. In order to study the particulate matter in the shelf seas, large volumes of water passed through cotton-wound cartridge filters with a pore size of approximately one µm.

6.2.2 Sampling and radioactivity measurements of ^{90}Sr

A volume of 100 litre sea water for ^{90}Sr analysis was collected in the 200 l vessels on board the ship. Trace amounts of the gamma ray emitting ^{85}Sr added as chemical yield determinant co-precipitated as Sr-oxalate at pH 4-5. The precipitate settled overnight, and was then collected in small bottles to be analysed in Lund. In highly concentrated nitric-acid Ca-oxalate precipitate dissolve, while strontium forms insoluble nitrates. Two re-precipitation steps combined with an acetone wash yield a very pure strontium salt which is suitable for gravimetric recovery (Bojanowski and Knapinska-Skiba, 1990).

6.2.3 Sampling and radioactivity measurements of ^{129}I

Sea water samples for ^{129}I analysis were collected in plastic bottles in a volume of 1 l. The samples were sent to Iso-Trace Laboratory, Toronto, Canada for mass spectrometry analysis of ^{129}I , by using a 3 MV tandem accelerator mass spectrometer.

6.2.4 Method of analysis of Trans uranium elements ($^{239+240}\text{Pu}$, ^{241}Am)

A volume of 200 litre sea water for $^{239+240}\text{Pu}$ and ^{241}Am analysis was collected in the 200 l the precipitation vessels in our laboratory on board. Concentrated HCl was added to adjust the pH-value to be < 2 . Yield determinants (^{242}Pu and ^{243}Am) were then added to the water samples in the vessels. To obtain isotopic equilibrium the mixture stirred for about 30 minutes by air-injection. Then NaOH in pellets was added to adjust the pH-value to be > 10 . Trans-uranium elements co-precipitated in the precipitation of various hydroxides. The precipitation settled over night to the bottom of the funnel shaped vessels and was then collected in 10 l bottles for transfer to the laboratory in Lund. At the laboratory in Lund the precipitates were dissolved in HCl. By the adding ammonia to the solution Pu and Am were co-precipitated with iron hydroxide at pH > 10 . Pu and Am isotopes were then separated, following the methods described in IAEA Technical Report 295 (1989), and electroplated onto

stainless steel discs (IAEA, 1989). The alpha activity on the discs was measured, either by surface barrier detectors or passivated ion implanted silicon detectors (PIPS).

The filters turned into ashes at 550 °C during night, and the residue leached for approximately 12 hours with *Aqua Regia* (a mixture of concentrated HNO₃ and HCl in a volume ratio of 1:3). After filtering to remove the unsolved material, the procedure continued as described above for the water samples.

6.3 Results and Discussion

6.3.1 ¹³⁴Cs and ¹³⁷Cs in Arctic Ocean water

¹³⁷Cs in Surface water

The distribution of ¹³⁷Cs in the surface layer at a depth of 8 m of the Arctic Ocean water along the route of the expedition is displayed in **Figure 6-5**.

The highest concentrations are found in a band stretching from north of the Lomonosov Ridge and its close vicinity across the North Pole and south along 15 °E in the Amundsen Basin. Lower activity levels are found in the Nansen and eastern Amundsen Basins. At the Gakkel ridge at about 10 °E, a front separates the high activities to the north from the significantly lower to the south. ¹³⁷Cs concentration falls from 10.7 to 5.2 Bq.m⁻³ and the ¹³⁴Cs/¹³⁷Cs activity ratio from 0.005 to 0.003.

Also in 1994 this front was observed in ¹³⁷Cs activities in the same area, decreasing from 11.3 to 5.1 Bq.m⁻³ (Ellis et. al., 1995). The activity-gradient reflects the input fallout from Chernobyl and release from reprocessing plants in the surface layer of the Arctic Ocean. The frontal zone between surface water from the Barents Sea and that originating further east on the shelf was in this area situated at approximately 85 °N in 1987 (Anderson and Jones, 1992), i.e. at the same latitude as the 1996 activity gradient. This implies that the radioactivity derived from European reprocessing sources enters the Arctic Ocean surface layer well east of the Barents Sea.

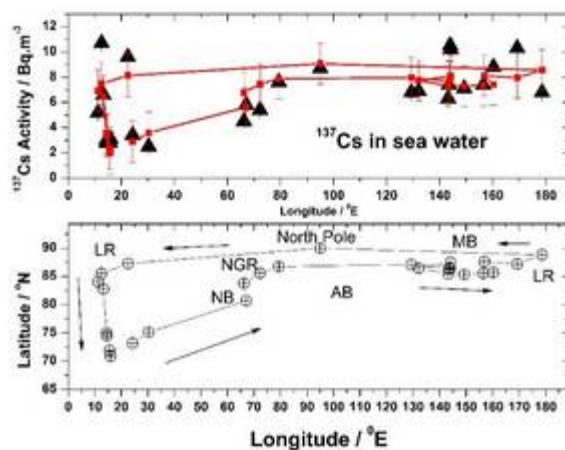


Figure 6-5

The distribution of ¹³⁷Cs in the surface layer at a depth of 8 m of the Arctic Ocean water at the various sampling locations along the route of the expedition. The values predicted by PLS regression modelling given in red.

The overall ^{137}Cs distribution is similar to the 1991 results, with the exception of the western Nansen Basin, where a local maximum was observed in 1991 (Roos et. al., 1998). This maximum may also be discerned in the 1987 results, although only results from few stations were reported (Cochran et. al., 1995). In the area close to the North Pole, ^{137}Cs surface activities were lower than earlier in the 1990's, with maximum activities in 1991 and 1994 of $18 \text{ Bq}\cdot\text{m}^{-3}$ (Roos et. al., 1995) and $15 \text{ Bq}\cdot\text{m}^{-3}$ (Ellis et. al., 1995) respectively.

^{134}Cs in Surface water

The distribution of $^{134}\text{Cs} / ^{137}\text{Cs}$ -activity ratio in the surface layer at a depth of 8 m of the Arctic Ocean water along the route of the expedition is displayed in **Figure 6-6**.

Equation of the PLS model:

$$^{134}\text{Cs}/^{137}\text{Cs} = 0.0849 + 1.53 \cdot 10^{-3} \cdot (\text{Latitude}^\circ\text{N}) + 1,74 \cdot 10^{-4} \cdot (\text{Longitude}^\circ\text{E})$$

Goodness of fit statistics (Variable Cs-134/): $R^2 = 0,143$

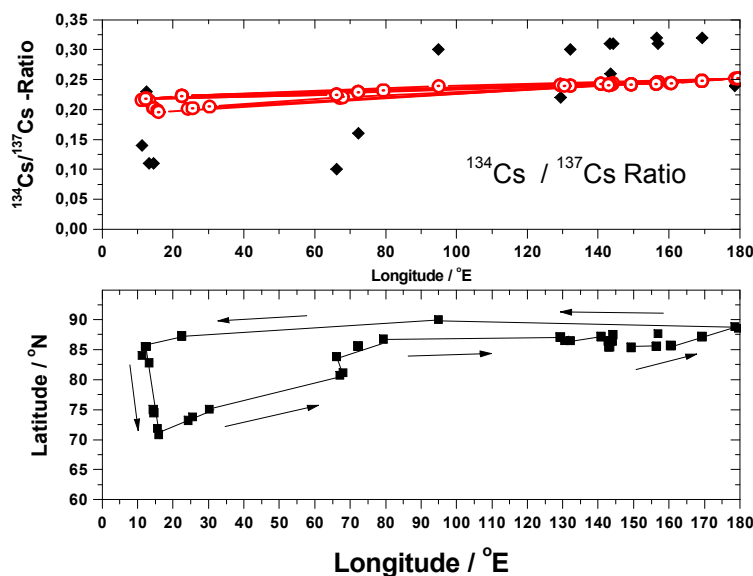


Figure 6-6

The distribution of $^{134}\text{Cs}/^{137}\text{Cs}$ -activity Ratio in the surface layer at a depth of 8 m of the Arctic Ocean water at the various sampling locations along the route of the expedition. The values predicted by PLS regression modelling given in red.

The ^{134}Cs activity originates partly from release from Sellafield reprocessing plant and debris from the Chernobyl accident deposited during 1986 in the north-west Norwegian Sea (Mitchell and Steele, 1988), which compared to reprocessing activity, may result in a different distribution in the central Arctic Ocean. The Chernobyl activity deposited in the Baltic Sea follows the NCC on its way to the Arctic. During the 1970's and early 1980's, the release from Sellafield reprocessing plant could be traced by the increased $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio. But since 1980 the release of ^{134}Cs from Sellafield

has become less significant (Aarkrog et al., 1987). By 1996, the initial Chernobyl activity ratio of approximately 0.5 had decreased to about 0.2. Thus the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio, could be used to trace the fallout from the Chernobyl accident due to its high content of ^{134}Cs . The contribution of ^{134}Cs from Chernobyl is thus estimated to be in the order of 30 % at the Northern Lomonosov Ridge, and at the North Pole. The ratios decrease towards the Amundsen and Nansen Basins, which contain roughly 23-25 and 10-16 % Chernobyl derived ^{134}Cs respectively. In 1987 20-30 % Chernobyl caesium was found at the continental slope north of Svalbard down to about 500 m (Cochran et al., 1995) and in 1991 Chernobyl (Josefsson, 1998).

6.3.2 Strontium-90 in Arctic Ocean water

The distribution of ^{90}Sr in the surface layer at a depth of 8 m of the Arctic Ocean water along the route of the expedition is displayed in **Figure 6-7**. The values predicted by PLS regression modelling of the data with Longitude and Latitude as explanatory X-values are also displayed in the figure.

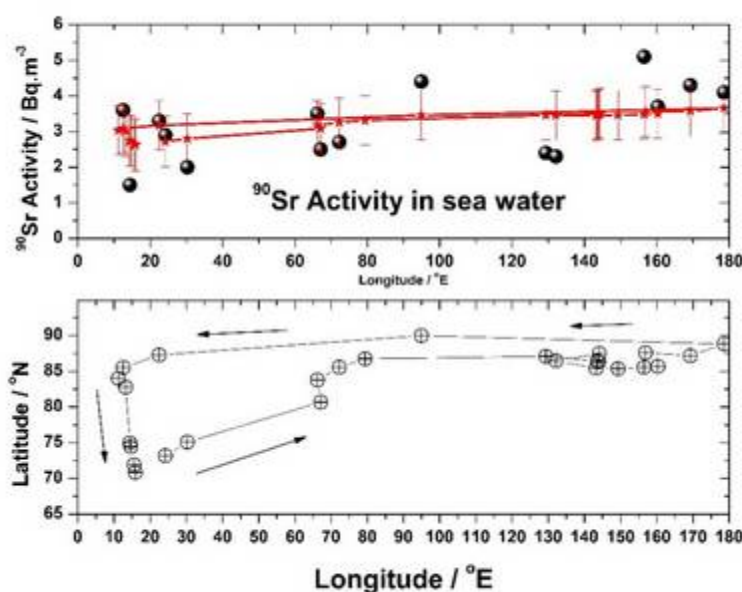


Figure 6-7

The distribution of ^{90}Sr in the surface layer at a depth of 8 m of the Arctic Ocean water along the route of the expedition. The values predicted by PLS regression modelling given in red.

Equation of the model:

$$^{90}\text{Sr} [\text{Bq.m}^{-3}] = 0.263 + 0.326 \cdot (\text{Latitude}^{\circ}\text{N}) + 2.75 \cdot 10^{-3} \text{Longitude}^{\circ}\text{E}$$

Goodness of fit statistics (Variable $^{90}\text{Sr} [\text{Bq.m}^{-3}]$): $R^2 = 0,196$

Mean: $^{90}\text{Sr} [\text{Bq.m}^{-3}] = 3.22 \pm 0.74$

The ^{90}Sr fallout background is estimated to be 1.6 Bq.m^{-3} , and an additional source is needed to reach the recorded levels of $2.3\text{-}5.1 \text{ Bq.m}^{-3}$ in the central Arctic Ocean (Dahlgard et al., 1995). This

source is assumed to be associated with releases from the Russian rivers, since the contributions from Sellafield and Chernobyl are small (Aarkrog, 1988). The mean concentrations of ^{90}Sr in the Ob and Yenisey rivers were reported to be about $10\text{-}20\text{ Bq}\cdot\text{m}^{-3}$ in the 1990's with a $^{90}\text{Sr}/^{137}\text{Cs}$ run-off ratio above 2 (Chumichev, 1997, Dahlgaard et al., 1995). In 1994, indications of additional river input of ^{90}Sr were recorded in the Laptev Sea surface water. But in the low salinity waters outside Yenisey, the concentrations were not larger than at surrounding stations (Josefsson, 1998).

Atlantic layer circulation is reflected in the ^{137}Cs and ^{90}Sr activity distributions, with activities increasing along the path as an effect of the decreasing input. Thus, lower ^{137}Cs and ^{90}Sr activities are found in the Nansen Basin north of Frans Josef Land, higher above the Lomonosov Ridge and highest above the western Gakkel Ridge. The latitudinal distribution of the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio is displayed in **Figure 6-8**. The $^{137}\text{Cs}/^{90}\text{Sr}$ ratio in the fallout from nuclear weapons tests is about 1.6.

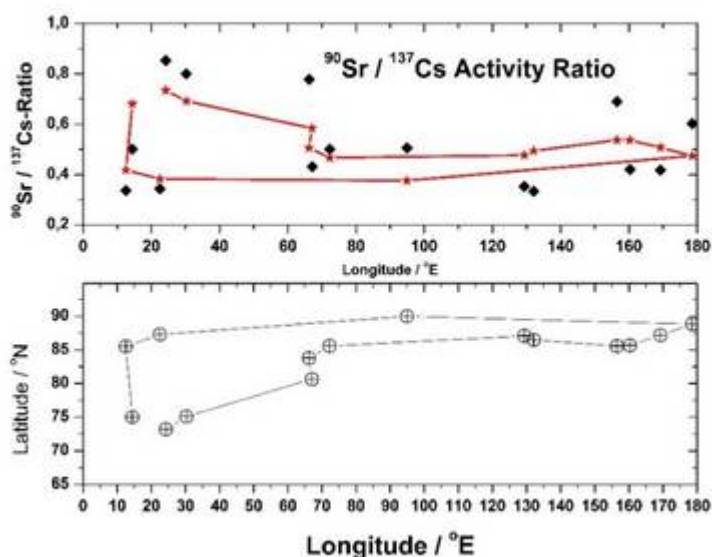


Figure 6-8

The distribution of $^{90}\text{Sr}/^{137}\text{Cs}$ -ratio in the surface layer at a depth of 8 m of the Arctic Ocean water along the route of the expedition. The values predicted by PLS regression modelling given in red.

Equation of the model:

$$\text{Sr}^{90/137}\text{Cs-Ratio} = 2.530 - 0.248 \cdot (\text{Latitude}^\circ \text{N}) + 8.31 \cdot 10^{-4} \cdot (\text{Longitude}^\circ \text{E})$$

Goodness of fit statistics (Variable Ratio): $R^2 = 0.365$. Average: 0.52 ± 0.18

6.3.3 Iodine-129 in Arctic Ocean water

The distribution of ^{129}I in the surface layer at a depth of 8 m of the Arctic Ocean water along the route of the expedition is displayed in **Figure 6-9**

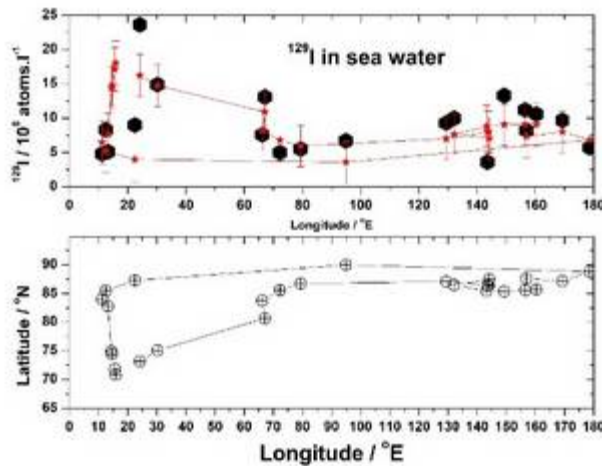


Figure 6-9

The distribution of ¹²⁹I in the surface layer at a depth of 8 m of the Arctic Ocean water along the route of the expedition. The values predicted by PLS regression modelling given in red.

Equation of the model:

$$^{129}\text{I} [\times 10^8 \text{ atoms.l}^{-1}] = 78,939 - 0,866 \cdot (\text{Latitude}^\circ\text{N}) + 0.0268 \cdot (\text{Longitude}^\circ\text{E})$$

Goodness of fit statistics: $R^2 = 0.568$

Average: $(9.260 \pm 4,591) \cdot 10^8 \text{ atoms.l}^{-1} = 1.5 \pm 0.7 \text{ femto-molar(fM)}$

A comparison of the distribution of the ¹²⁹I activity and the ¹³⁴Cs/¹³⁷Cs activity ratio shows that the Sellafield, La Hague and Chernobyl accident releases have almost the same surface distribution in the Eurasian part of the Arctic Ocean. The border between the Beaufort Gyre and the transpolar drift is possibly reflected by slightly lower ¹²⁹I concentrations at Station 34 in the Makarov Basin and at the North Pole Station. As an effect, both the ¹²⁹I activity and the ¹²⁹I/¹³⁷Cs atom ratio on the Lomonosov Ridge are higher to the south, i.e. upstream of the transpolar drift, reflecting the smaller time elapsing since the release.

6.3.4 ¹³⁷Cs, ⁹⁰Sr, and ¹²⁹I in Subsurface water layers

The average depth distribution for ¹³⁷Cs at all stations is illustrated in **Figure 6-10**. In **Figure 6-11**, the depth distributions of ⁹⁰Sr and ¹²⁹I in the Arctic Ocean waters are displayed as averages of all sampling stations.

The estimated mean residence times for the surface sea water layer are 2-6 years and up to 15 years in the high salinity gradient layer i.e. the halocline (Schlosser et al., 1995). This is compatible with our relative age signal from the ¹²⁹I concentrations and the ¹²⁹I/¹³⁷Cs ratios in the halocline, which are significantly lower than in the surface layer. A temporary input of higher activity to both the surface and halocline layers will traverse the surface layer much faster than the halocline one, and will after sufficient time, give a relative peak in halocline activity. The lack of a corresponding peak in the ¹²⁹I and ⁹⁰Sr concentrations exclude Sellafield and global fallout as the sources and it may instead be Chernobyl activity introduced into the central Arctic Ocean around 1990. Higher values of ⁹⁰Sr in the

surface layer are an effect of the riverine output, which enhances surface activity as compared to subsurface layers.

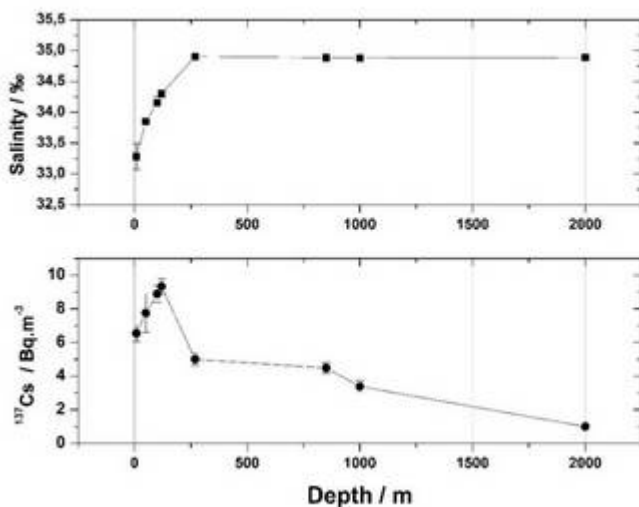


Figure 6-10

The average depth distributions for ¹³⁷Cs and salinity ‰ of all stations.

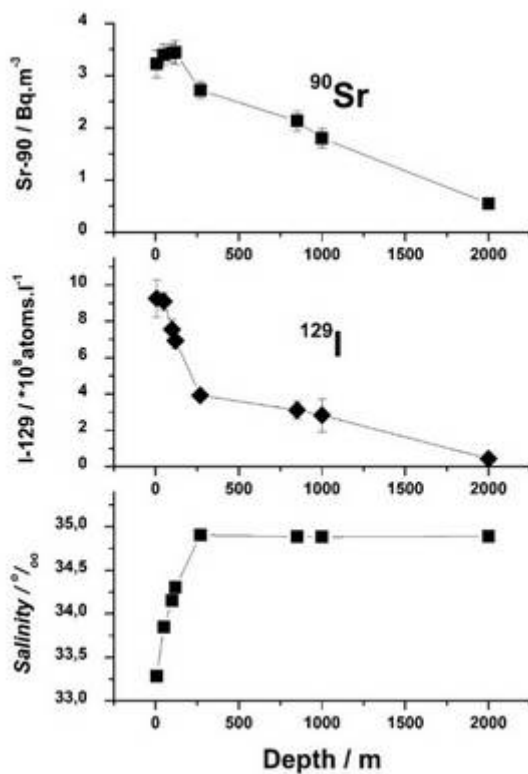


Figure 6-11

The average depth distributions of ⁹⁰Sr, ¹²⁹I and salinity ‰ of all stations.

Concentration of ^{129}I also follow the Atlantic layer circulation, but with concentrations decreasing along the path as an effect of the increasing Sellafield and La Hague releases. These patterns are the same as for the lower Atlantic layer at 850-1000 m depth with only slightly lower concentrations of all three radionuclides. The ^{129}I concentrations at stations 33 and 34 in the Makarov Basin are lower than on the Lomonosov Ridge, both in the Atlantic core samples and the lower Atlantic layer samples. Comparisons with the corresponding surface water results discussed above indicate that the reprocessing signal in the surface layer seems to extend a little farther into the Canadian Basin. In the North Pole area, water column profiles obtained earlier reveal a decrease in the ^{137}Cs content since 1991 and 1994 throughout the Atlantic layer (Smith et al., 1998).

In subsurface samples, no ^{134}Cs activity was determined and thus no exclusive Chernobyl tracer is available. Elevated $^{137}\text{Cs}/^{90}\text{Sr}$ ratios may both be an effect of a higher amount of Chernobyl activity or of older Sellafield activity, and lower $^{129}\text{I}/^{137}\text{Cs}$ ratios may be an effect of old Chernobyl caesium. These ratios are almost constant throughout the Atlantic layer at each station, showing that the source composition and thus possibly the origin of the water mass are the same. In the water sample of 2000 m depth at station 34, the activity level is lower than in the 850 m sample at the same station. The ratios between the nuclides are, however, almost the same, which indicate that the source composition is approximately the same, although diluted with low-activity deep water.

6.3.5 Transuranic elements ($^{239+240}\text{Pu}$, ^{241}Am) in the central Arctic Ocean

The activity concentrations of $^{239+240}\text{Pu}$, ^{241}Am in surface water of the central Arctic Ocean in 1996 are presented in **Figure 6-12**. The average activity concentrations of $^{239+240}\text{Pu}$ is $7.91 \pm 0.65 \text{ mBq.m}^{-3}$, of ^{238}Pu $0.47 \pm 0.07 \text{ mBq.m}^{-3}$ and, of ^{241}Am $0.38 \pm 0.11 \text{ mBq.m}^{-3}$.

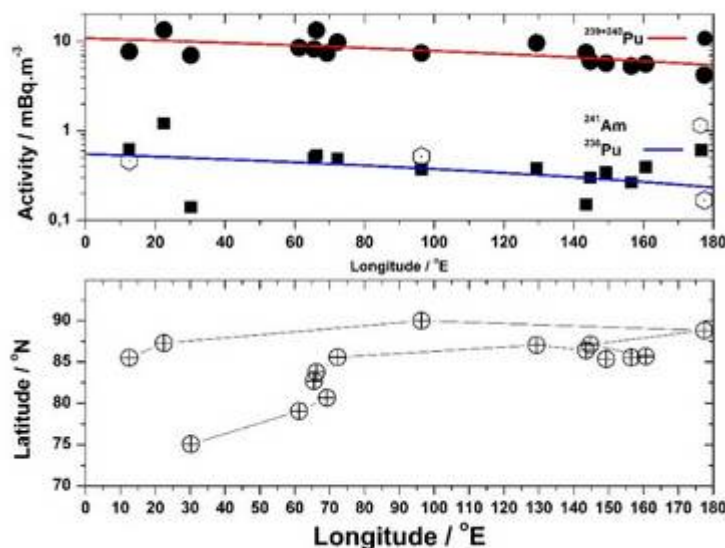


Figure 6-12

The activity concentrations of $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am in surface water of the central Arctic Ocean in 1996.

$^{239,240}\text{Pu}$ concentrations in near-surface seawater of the Eurasian Shelf has been studied in the course of the Arctic '96 and Tundra Ecology '94 expeditions. Samples were, however, also been taken from aboard the F.S. Polarstern in July–August 1996 (Vintro et al., 2002, Persson et al., 2015a, Persson et al., 2015b). The Latitudinal distribution of $^{239-240}\text{Pu}$ activity concentration in surface water measured by these expeditions and the sampling sites is displayed in **Figure 6-13a b**. The results of the Tundra Ecology '94 and F.S. Polarstern expeditions exhibit the same pattern with an increasing activity concentration toward higher latitudes. This is probably because as there sampling sites cover the same longitudinal region. The sampling sites of the Arctic Ocean-96 are spread over the whole Arctic Ocean with a wide spread in the $^{239,240}\text{Pu}$ concentrations. Although at 86°N 160°E where the sampling sites of Polarstern and Arctic Ocean-96 coincide, the values of the $^{239,240}\text{Pu}$ concentrations are in good agreement about $5 \text{ mBq}\cdot\text{m}^{-3}$.

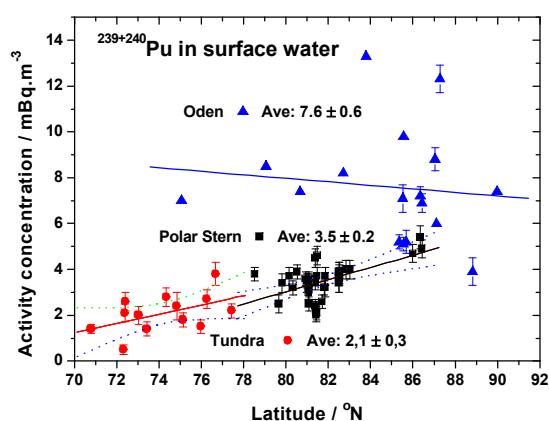


Figure 6-13a

The Latitudinal distribution of $^{239-240}\text{Pu}$ activity concentration in surface water measured by 3 different expeditions Tundra 1994, Polarstern and Arctic Ocean-96

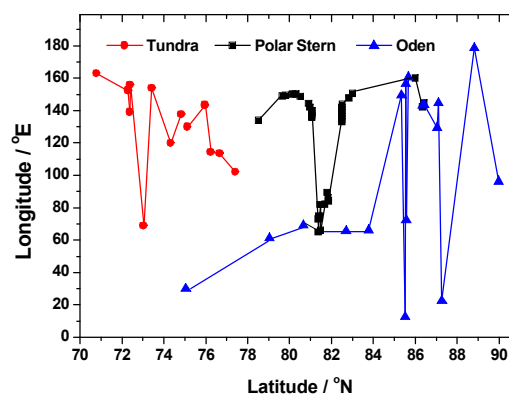


Figure 6-13b

The surface water sampling sites of the 3 different Arctic expeditions Tundra 1994, Polar Stern and Arctic Ocean-96.

The lowest concentrations of $^{239+240}\text{Pu}$ in the surface water (4.2 to $7.5 \text{ mBq}\cdot\text{m}^{-3}$) was found on the Lomonosov Ridge and in the Makarov Basin. In the Eurasian Basin, the concentrations of $^{239+240}\text{Pu}$ are somewhat higher, about $13.4 \text{ mBq}\cdot\text{m}^{-3}$. The depth distribution of the Pu and Am isotopes displayed in **Figure 6-14** follow the same pattern as the salinity.

River water injected into the central Arctic Ocean from the eastern Kara or the Laptev Seas is assumed to be the dominant source of fresh water in the Amundsen and Makarov basins (Anderson and Jones, 1992). The depletion of plutonium is probably due to sediment scavenging on the continental shelf and at the shelf break. Comparisons with data obtained earlier for plutonium in the Arctic Ocean surface layer, reveal concentrations changing with time.

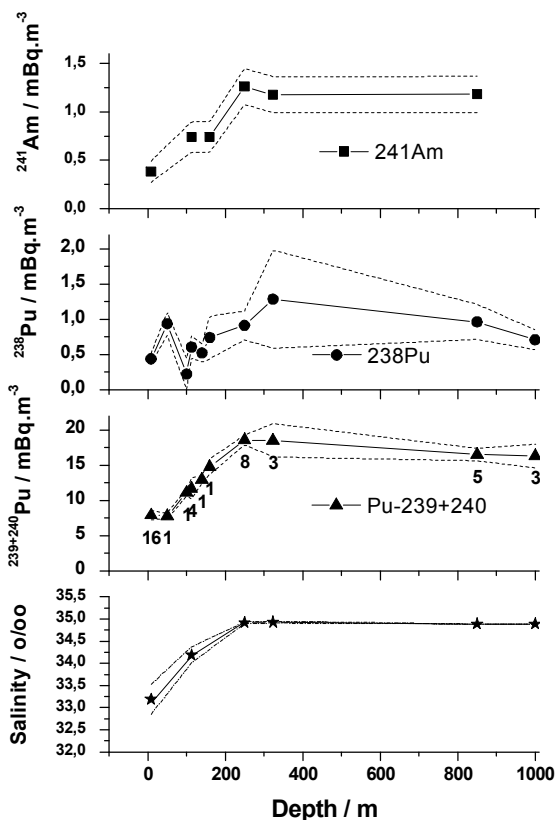


Figure 6-14.

Depth distribution of salinity and the activity concentrations of $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am in the Arctic Ocean 1996. The figures represent the number of samples for each dot.

A large part of ^{214}Am is produced in situ from ^{241}Pu . Because ^{241}Am has a greater association with particles in the water than Pu-isotopes, the $^{241}\text{Am} / ^{239+240}\text{Pu}$ ratio increase with depth as shown in Figure 6-15.

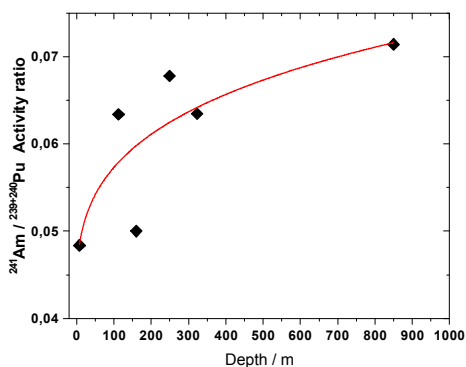


Figure 6-15

The variation of the $^{241}\text{Am} / ^{239+240}\text{Pu}$ activity ratio variation with water depth.

At the LOREX station in the northern Makarov Basin, the surface concentration was 14 mBq.m^{-3} in 1979 and had reached $17\text{-}18 \text{ mBq.m}^{-3}$ in the Nansen Basin in 1987 (Livingston et al., 1984, Cochran et al., 1995). However, current activity concentrations are approximately the same as recorded during the 1991 Arctic Ocean expedition in the same area. The lower concentration 1996 is likely the effect of the depletion of plutonium in the North Atlantic surface layer, which is the current supply of transuranic elements to the Arctic. An effective half-life of 7-8 years has been estimated for surface plutonium in the north-east Atlantic Ocean which is in good agreement with the Nansen Basin decrease from 1987 to 1996 (Holm et al., 1991).

No obvious contributions of the high activities released from Russian rivers have been observed, although at Station A5, the value of 13.1 mBq.m^{-3} might indicate an influence of high concentration riverine outflow.

The measurements of ^{238}Pu , yield a mean $^{238}\text{Pu} / ^{239+240}\text{Pu}$ activity ratio of 0.05 ± 0.03 in central Arctic Ocean surface water. This is close to the fallout ratio of 0.04, hence only a minor fraction of the plutonium may be attributed to Sellafield.

The activity concentration of ^{241}Am was measured only at three surface water stations, yielding a mean activity of 0.4 mBq.m^{-3} and a mean activity $^{238}\text{Pu} / ^{239+240}\text{Pu}$ activity ratio of 0.06. A similar ratio was measured in the north-west Atlantic in 1984, and a slightly higher value, 0.1, in the Nansen Basin in 1987 (Cochran et al., 1987, Cochran et al., 1995).

The samples collected during the expedition were used to trace releases of ^{134}Cs , ^{137}Cs and ^{129}I from the Chernobyl accident and Sellafield reprocessing plant. From the NCC input, the surface pathway, as traced by enhanced $^{134}\text{Cs}/^{137}\text{Cs}$ ratios and ^{129}I concentrations, led east along the Eurasian shelf seas to the Laptev Sea, then north, following approximately the Lomonosov Ridge, past the North Pole and towards the East Greenland Current. Compared to this, the different distribution pattern of $^{239+240}\text{Pu}$ is another evidence that plutonium is not, to any significant extent, derived from Sellafield releases.

As a result of the Chernobyl and Sellafield input of radio caesium, the average surface $^{239+240}\text{Pu} / ^{137}\text{Cs}$ activity ratio is reduced by a factor 30 from the fallout level of 0.03 to approximately 0.001. The subsurface layers show higher plutonium concentrations than the surface layer. The Lomonosov Ridge and north Makarov Basin profiles are very uniform, with the highest concentrations, $17.3 - 22.0 \text{ mBq/m}^3$ observed at the temperature maxima at 235-340 m depth and slightly lower ones, $14.2 - 17.0 \text{ mBq/m}^3$, in deep Atlantic layer samples. In comparison, the concentrations at Stations A4 and A5 in the Nansen Basin in both layers are lower, but in level at Station A40 in the western Amundsen Basin. The observed depth distribution may be compared to the mean depth profile in the West Spitsbergen current (WSC) and the Barents Sea in 1995 (Herrmann et al., 1998), which showed an activity maximum at about 100 m followed by a slow decrease with depth. When the WSC enters the Arctic Ocean, it sinks below the Arctic surface water and the maximum plutonium concentrations move to lower depths. The major Arctic Ocean inflow across the Barents Sea enters the Nansen Basin mainly below the surface layer. North of Svalbard and Frans Josef land, Atlantic water is observed to penetrate down to approximately 500 m, while the inflow through the St. Anna Trough distributes the Atlantic water down to 1000 m (Schauer et al., 2002). The combined WSC and Barents Sea inflow will then result in a plutonium maximum deeper than 100 but above 1000 m.

Apart from our data, this is also confirmed in the 1987 when $^{239+240}\text{Pu}$ maxima on the continental slope north of Barents Sea was found at 350-1300 m depth and at approximately 600 m depth in the central Nansen Basin (Herrmann et al., 1998). The mean $^{238}\text{Pu} / ^{239+240}\text{Pu}$ ratio in both layers is 0.05.

At some stations, the ratio rises to above 0.1, but the large standard deviations in the ^{238}Pu results make it difficult to draw any specific conclusions, other than fallout being the dominating source also in the Atlantic layer.



Figure 6-16
The sediment core sampler

(Photo Bertil Persson)

6.3.6 $^{239+240}\text{Pu}$ and ^{137}Cs in sediments of the Arctic Ocean.

During the Arctic Ocean -96 expedition sediment samples were taken by using a multiple corer (10 cm diameter) and the cores sliced immediately on board, with the upper two cm sliced in 0.5 cm slices and the rest in 1 cm slices. The samples were transferred to Lund where they were dried and grinded. The ^{137}Cs activity measured by Ge(Li) gamma-spectroscopy. For the alpha measurements, the sediment was ashed in 550 °C over night, and then oxidised in aqua regia for 12 hours. The undissolved fraction was removed by filtering and the plutonium separated following the methods described in IAEA Technical Report 295 (1989) (IAEA, 1989). Plutonium was electroplated onto stainless steel discs and the alpha activity was measured on either surface barrier detectors or passivated ion implanted silicon detectors (PIPS).

The results of, the integrated sediment activity of ^{137}Cs and $^{234+240}\text{Pu}$ at specific sampling station are given in **Table 6-1** and displayed in **Figure 6-17**.

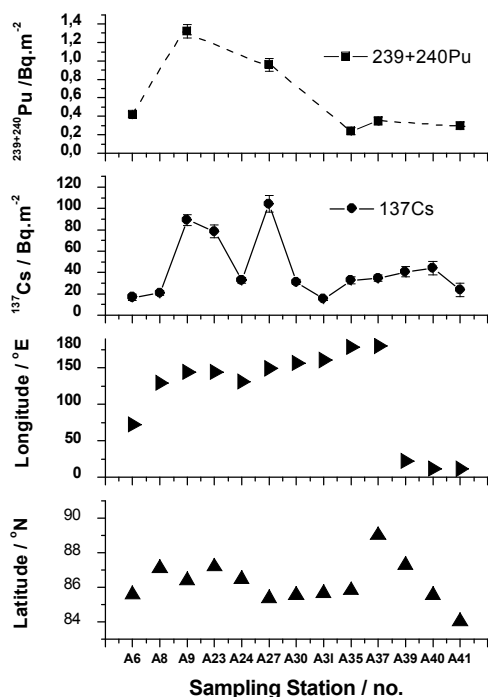


Figure 6-17 Sediment

The integrated sediment activity of ^{137}Cs and $^{234+240}\text{Pu}$ at specific sampling station with Latitude and longitude.

In the central Arctic Ocean, the integrated sediment activity of ^{137}Cs varied between 119 and 836 and of $^{234+240}\text{Pu}$ between 5-30 $\text{Bq}\cdot\text{m}^{-2}$. Highest inventories of both radionuclides are found at the shallow (-1000 m) stations of the Lomonosov Ridge, and significantly lower in the Eurasian Basin. There is a correlation between the sediment inventories and water-column concentrations of the European-derived radionuclides in the upper 1000. The main surface pathway of the releases from Sellafield, LaHague and the Chernobyl accident to the Arctic Ocean surface layer is along the Eurasian shelf seas, entered the Nansen Basin from the Laptev Sea and followed approximately the Lomonosov Ridge north. The main outflow of suspended particles occur from the Laptev Sea, and deposit on the eastern Lomonosov Ridge.

The depth distribution of the ^{137}Cs activity was rather uneven but with a penetration depth down to about 9-19 cm. The $^{234+240}\text{Pu}$ activity, however, shows a more steady decrease. It has passed approximately 35-40 years since the first anthropogenic radionuclides was introduced to the Arctic Ocean, and the sedimentation in the central Arctic Ocean during this period is well below 1 mm. The activity penetration to greater depths may be an effect of sediment mixing caused by currents or biological activity or from diffusion in the pore water.

The $^{234+240}\text{Pu}/^{137}\text{Cs}$ activity ratio in the sediment inventories was 0.045 ± 0.006 . This value is higher than in the global fallout for which the deposition ratio in 1994 was estimated to 0.024 (UNSCEAR, 1993). The generally elevated ratios in the shelf sediment compared to fallout levels, is likely an effect of additional ^{137}Cs input from Sellafield and the Chernobyl accident. The larger particle affinity of plutonium compared to caesium removes a larger fraction of the water column plutonium

before reaching the Nansen Basin, and this explains the lower ratios in the central Arctic Ocean sediment.

Table 6-1

Sampling location and integrated sediment activity of ^{137}Cs and $^{239+240}\text{Pu}$ at the Arctic Ocean-96.

Station	Latitude	Longitude	Water depth [m]	Pene- tration [cm]	^{137}Cs		$^{239+240}\text{Pu}$	
	$^{\circ}\text{N}$	$^{\circ}\text{E}$			SD		SD	
	degree	degree			[Bq.m ⁻²]		[Bq.m ⁻²]	
A6	85.57	72.28	3780	1	16.9	2.8	0.42	0.03
A8	87.08	129.50	3762	3	20.6	2.2		
A9	86.38	144.33	995	4	89.1	5.1	1.32	0.07
A23	87.18	144.57	980	4	78.7	5.9		
A24	86.47	130.92	4150	3	32.9	2.6		
A27	85.35	149.25	1110	4	104.4	7.7	0.96	0.07
A30	85.53	156.48	2388	2	31.1	2.4		
A31	85.65	160.58	3586	1	15.2	1.7		
A35	85.82	178.70	1113	2	32.6	3.5	0.24	0.04
A37	89.00	179.92	2271	3	34.5	3	0.35	0.04
A39	87.28	22.47	4180	0.5	40.7	4.9		
A40	85.52	12.05	2360	2	44.2	6.1		
A41	84.03	11.32	3270	2	23.6	6.3	0.3	0.04

Regarding the possibility that the Arctic Ocean acts as a sink for particle reactive elements, we have roughly calculated the time-integrated water column inventory for the time period when the Arctic Ocean has been exposed to ^{137}Cs and $^{239+240}\text{Pu}$ (i.e. approximately 35 years, 1961-1996) and compared with the sediment inventory. In the Kara and Laptev Seas we assumed the mean activity concentration over time to be 5 Bq.m⁻³ for ^{137}Cs and 10 mBq.m⁻³ for $^{239+240}\text{Pu}$ respectively. The residence time of fresh water in the Kara Sea has been estimated to 5 years and in the Laptev Sea to 3 years (Ostlund, 1994). We assume that the same values hold for the total water column, which during 35 years results in an activity renewal 7 and 12 times for the Kara and Laptev Seas respectively. The mean activity concentration of ^{137}Cs and $^{239+240}\text{Pu}$ during this period in the central Arctic Ocean have used reported data of concentration (Josefsson, 1998, Cochran et al., 1995, Livingston et al., 1984) and calculated.

The mean ^{137}Cs activities during 1961-1996 becomes 6, 3 and 1.1 Bq.m⁻³ for the 0-200, 200-1000 and 1000-3000 m layers respectively, and the corresponding $^{239+240}\text{Pu}$ concentrations are 1.1, 17.2, and 8.0 mBq.m⁻³. From the residence time of 10 years for the upper 200 m (Ostlund and Hut, 1984, Schlosser et al., 1995) it is assumed that this layer have been renewed 3.5 times during the period. Regarding residence-times of approximately 35 years below 200 m depth, it is assumed that the mean activity also represent the time integrated activity in those layers. The estimated mean depth of the Kara Sea is 111 m (Pavlov and Pfirman, 1995), in the Laptev Sea 100 m, and in the central Arctic Ocean 3000 m.

The results are presented in **Table 6-2** together with mean integrated sediment activity and the fraction deposited on the sea-floor in the three different areas. The long residence time in the deep waters of the central Arctic Ocean may increase the relative sediment content.

Regarding the possibility that the Arctic Ocean acts as a sink for particle reactive elements, we have roughly calculated the time-integrated water column inventory for the time period when the Arctic Ocean has been exposed to ^{137}Cs and $^{239+240}\text{Pu}$ (i.e. approximately 35 years, 1961-1996) and compared with the sediment inventory. In the Kara and Laptev Seas we assumed the mean activity concentration over time to be 5 Bq.m^{-3} for ^{137}Cs and 10 mBq.m^{-3} for $^{239+240}\text{Pu}$ respectively.

Table 6-2. Coarse estimate of fraction deposited on the shelf and in the arctic interior, as compared to total time-integrated activity.

	^{137}Cs			$^{239+240}\text{Pu}$		
	Kara Sea	Laptev Sea	Deap Arctic Ocean	Kara Sea	Laptev Sea	Deap Arctic Ocean
Time- integrated water activity [Bq.m^{-2}]	3900	6000	8800	8	12	46
Mean integrated sediment activity [Bq.m^{-2}]	400	800	43	30	34	0,6
Fraction deposited %	9	12	0,5	79	74	1,3

The residence time of fresh water in the Kara Sea has been estimated to 5 years and in the Laptev Sea to 3 years (Ostlund, 1994). We assume that the same values hold for the total water column, which during 35 years results in an activity renewal 7 and 12 times for the Kara and Laptev Seas respectively. The mean activity concentration of ^{137}Cs and $^{239+240}\text{Pu}$ during this period in the central Arctic Ocean have used reported data of concentration (Josefsson, 1998, Cochran et al., 1995, Livingston et al., 1984) and calculated.

6.4 CONCLUSIONS

The highest ^{137}Cs , ^{90}Sr , ^{129}I activities, and $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratios are found in a band stretching from the Norther Lomonosov Ridge, over the north Pole and south along $10\text{-}15^\circ\text{E}$ to 85°N . The European reprocessing and Chernobyl releases, as traced by the ^{129}I and $^{134}\text{Cs}/^{137}\text{Cs}$ signals respectively have approximately the same spatial distribution in the Arctic Ocean surface layer. Fallout-derived activity is advected to the central Arctic Ocean via the NAC. But the fallout runoff of mainly ^{90}Sr is also significant and estimated to contribute with up to 2 Bq.m^{-3} to the surface layer. For $^{239+240}\text{Pu}$, however, the lowest concentrations of in the surface water in the range 4.2 to 7.5 mBq.m^{-3} was found on the Lomonosov Ridge and in the Makarov Basin. In the Eurasian Basin, the concentrations $^{239+240}\text{Pu}$ are somewhat higher, up to 13.4 mBq.m^{-3} .

From the results presented here and reported earlier, the pathways for discharges to the Northern European seas are outlined. Via the NCC, the releases are transported to the Barents Sea. One-third enters the central Arctic Ocean through the Fram Strait and the remaining two-thirds across the Barents Sea. About half the inflow across the Barents Sea enters the Atlantic layer, primarily through the St Anna Through. The activity remaining on the shelf is transported across the Kara Sea and into the Laptev Sea. From there, the major fraction follows the Lomonosov Ridge to the North Pole and then into the east Greenland current. From the $^{129}\text{I}/^{137}\text{Cs}$ ratio with measured Chernobyl and assumed fallout

contributions subtracted, the transport time from Sellafield to the central Arctic Ocean surface layer is estimated to be 13-14 years.



Figure 6-18

Bertil Persson standing at the North Pole with the flag of Scania and the flags of each participant's country.

For all the radionuclides analysed, the water profiles generally show activities decreasing with depth. An exception is the high ^{137}Cs activity concentrations found in the lower halocline layer at the Lomonosov Ridge and Makarov Basin stations. The maxima were not observed in the ^{90}Sr or ^{129}I measurements, and is assumed to be an effect of a temporary Chernobyl activity input to the Arctic Ocean surface layers in the years around 1990.

Inventories down to 900 m reveal that between 60-70 % of the ^{137}Cs , ^{90}Sr , and ^{129}I in the Arctic ocean water, are present in the surface layer inflow from the Atlantic. The total inventories down to 900 m in the Eurasian Basin of the Arctic Ocean are estimated to 6.7 PBq of ^{137}Cs , 3.4 PBq of ^{90}Sr and $5.3 \cdot 10^{28}$ atoms of ^{129}I .

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The three authors and radioactivity explorers during one of the expeditions
From left: Kjell-Åke Carlsson, Bertil Persson and Elis Holm

This book is dedicated to to professor emeritus Bengt Forkman (Nuclear Physics, Lund University) who in 1979 took the initiative to arrange an environmental radioactivity research program for the Ymer-80 expedition. He engaged his old friend Bertil Persson whom he during the 1960th inspired to university studies in Lund and just been promoted to professor of Radioecology at the Swedish University of Agricultural Sciences in Uppsala. In 1980, Bertil Persson became professor of Medical Radiation Physics at Lund University and was tutor for Elis Holm at Lund University, who for his thesis was engaged in radiochemical analysis of plutonium isotopes in the environment. Elis Holm became deeply involved in management of Ymer-80 and all the following expeditions as well. We found a superior talent for the logistics in Kjell-Åke Carlsson (who was mechanical engineer at the department of radiation physics at Lund University). Without him, we would not have been able to solve all the thousands of practical issues and contacts with authorities and sponsors. He also contributed with the diaries extensive photographic and video documentation of all the expeditions. We have together compiled this book although several others were partly engaged in the various expeditions to whom we are deeply thankful for their contributions. They will appear as co-authors in the separate chapters referring to the expedition in question.