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Radioactivity exploration from the Arctic to the Antarctic.
Part 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 ^{7}Be, ^{210}Pb. and ^{210}Po in the surface air, radioactive isotopes of Caesium (^{134}Cs, ^{137}Cs) and plutonium ($^{239+240}$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 ^{7}Be was 0.6±0.4 mBq.m^{-3} , ^{210}Pb 46±34 µBq.m^{-3} and ^{210}Po 37±23 µBq.m^{-3}

The activity concentration of ^{137}Cs in the surface of the Arctic Ocean was in the range of 8-12 Bq.m^{-3}. When crossing the Nansen basin the activity concentration of ^{137}Cs increased to about 18 Bq.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}Cs/^{137}Cs activity ratios was about 0.02 due to the contribution mainly from Sellafield and a few percent contribution from Chernobyl. The ^{134}Cs/^{137}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 $^{239+240}$Pu in the surface of the Arctic Ocean was in the range of 6 - 8 mBq.m^{-3}. But locally the activity concentration of $^{239+240}$Pu was found to be increased to 11 mBq.m^{-3} at 86°N 48-53°E, and to 16 mBq.m^{-3} at 83°N 10°E.

A. 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**.

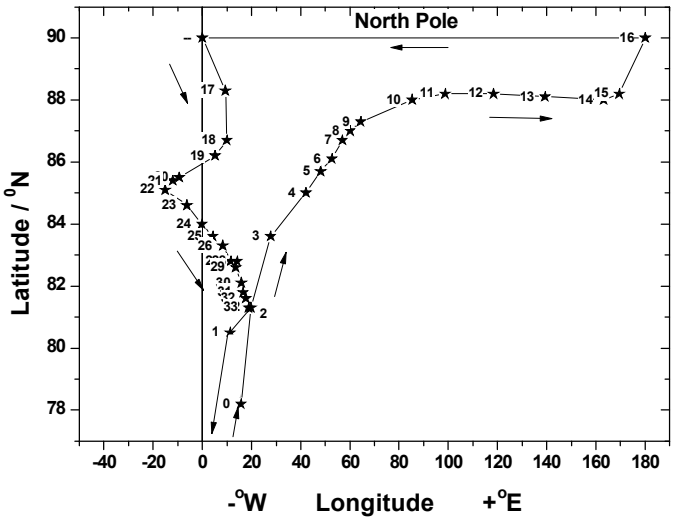

Figure 4-3
Route of the Arctic Ocean -91 expedition with station numbers

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}C 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
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

B. ^{7}Be, ^{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 ^{7}Be 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 m^3 were collected at each occasion on membrane filters (size 0.25×0.25 m) at a flow rate of 100 m^{3}h^{-1}. The Andersen sampler was compared with a FOA transportable reference high volume air sampler (Microsorban filter, 0.56x0.56 m^2, 1 100 m^{3}h^{-1}). The samplers were placed close together on deck of the cruise vessel as show in **Figure 4-4**. The ^{7}Be results for the Anderson sampler were normalized to that of the FOA sampler.

The activity concentrations of ^{7}Be (mBq.m^{-3}), ^{210}Pb. and ^{210}Po (µBq.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.**

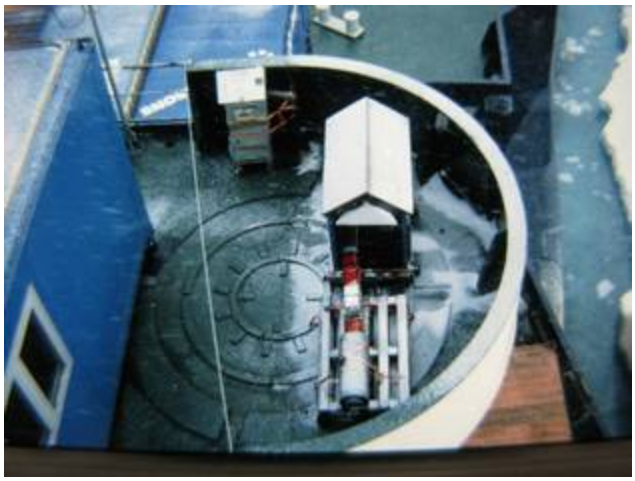

Figure 4-4
The Andersen air- sampler and the FOA transportable reference high volume air sampler placed close together

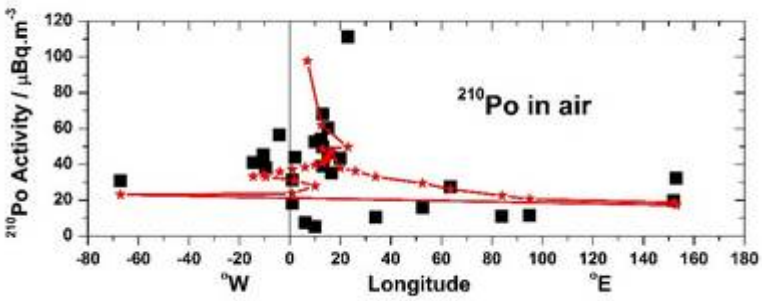

Figure 4-5a
Longitudinal distribution of ^{210}Po activity concentration in air over the Arctic Ocean during 1991.

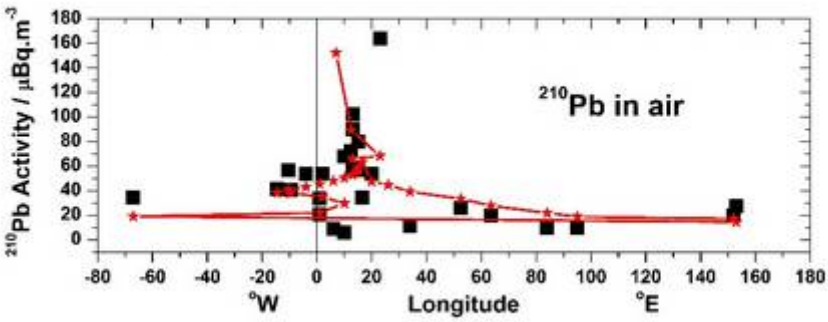

Figure 4-5b
Longitudinal distribution of ^{210}Pb activity concentration in air ^{7}Be, and over the Arctic Ocean during 1991.

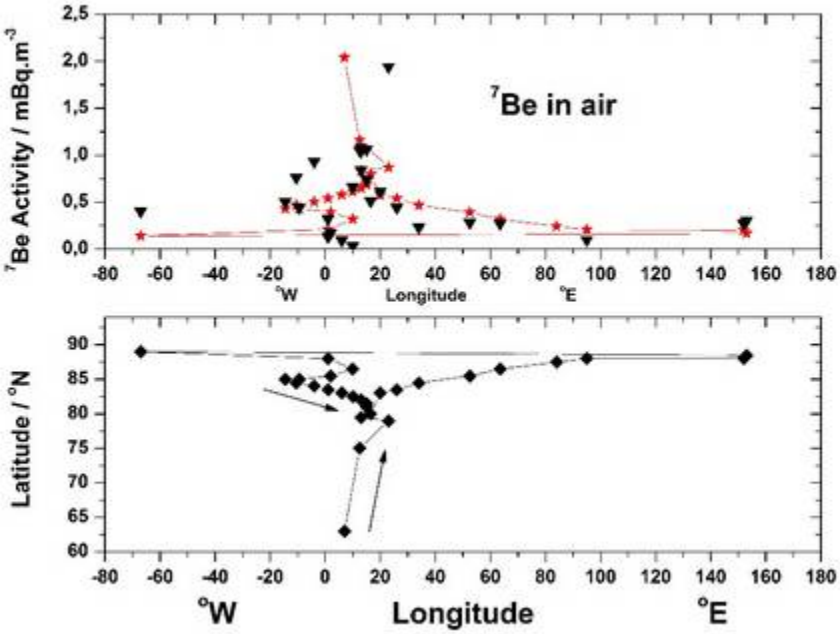

Figure 4-5c
Longitudinal distribution of ^{7}Be activity concentration in air and the route of Arctic Ocean expedition during 1991.

Equations of the PLS model:

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

Goodness of fit statistics $R^2 = 0.395$

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

Goodness of fit statistics $R^2 = 0.354$

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

Goodness of fit statistics $R^2 = 0.285$

Table AO91-2
Air concentrations of ^{7}Be ^{210Pb} 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 ^{7}Be 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 ^{7}Be ^{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

C. 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 ($Cu_2[Fe(CN)_6]$. 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 $^{134}Cs/^{137}Cs$ 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 cartright 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).

D. Results

D1. ^{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).

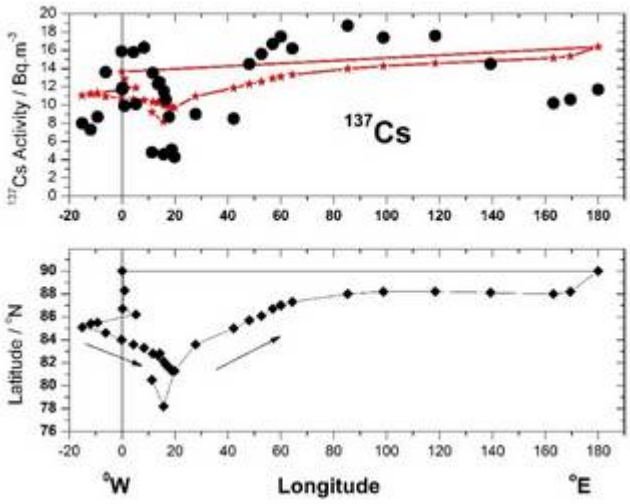

Figure 4-7
Activity concentration of ^{137}Cs in sea-water along the route (Roos et al., 1998). The red curve represent the estimated values of PLS modelling.

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

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

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

D2. $^{134+137}Cs$ isotopes in surface water

The Longitudinal distribution of $^{134}Cs/^{137}Cs$ 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 $^{134}Cs/^{137}Cs$ 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 Frams Strait the ^{137}Cs activity-concentration dropped to about 4.5-5.5 $Bq.m^{-3}$ with a much lower $^{134}Cs/^{137}Cs$ 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 $^{134}Cs/^{137}Cs$ activity ratio of about 0.02.

At the boundary between Nansen and Amundsen basin about 85 °N, a 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 basis 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 $Bq.m^{-3}$.

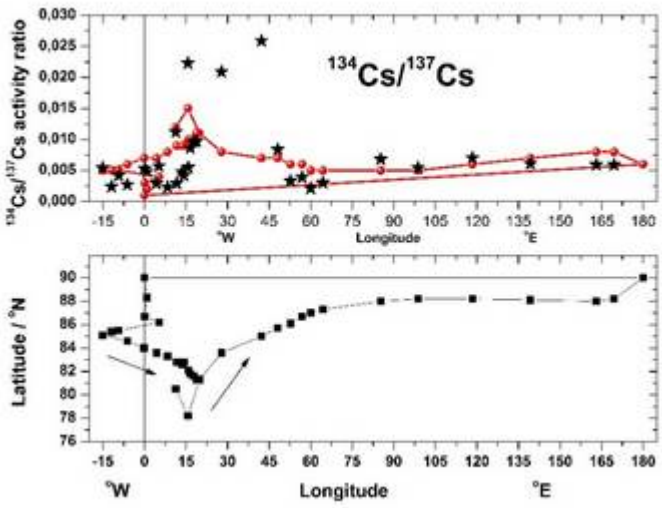

Figure 4-8
$^{134}Cs/^{137}Cs$ activity ratio in sea water along the route (Roos et al., 1998)

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

$^{134}Cs/^{137}Cs$-activity ratio = 0.111 – 0.00124×(Latitude°N) + $3.83 \cdot 10^{-5}$×(Longitude°E)
Goodness of fit statistics for $^{239+240}Pu$ resulted in a R^2 value of 0.219.

D3. Depth profiles of ^{137}Cs activity concentration

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

Nansen station 83.6°N, 30.29 °E profile :

$$A_{Cs\text{-}137}\ [Bq.m^{-3}] = 0.07 + 8.7 \cdot exp(-0.0012 \cdot d);$$
$$R^2=0.99;\ d½= 562\ m$$

Amundsen station 87.5°N, 106.44°E profile:

$$A_{Cs\text{-}137}\ [Bq.m^{-3}] = 0.37 + 14.9 \cdot exp(-0.0015 \cdot d)\ ;$$
$$R2=0.97;\ d½= 464\ m$$

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).

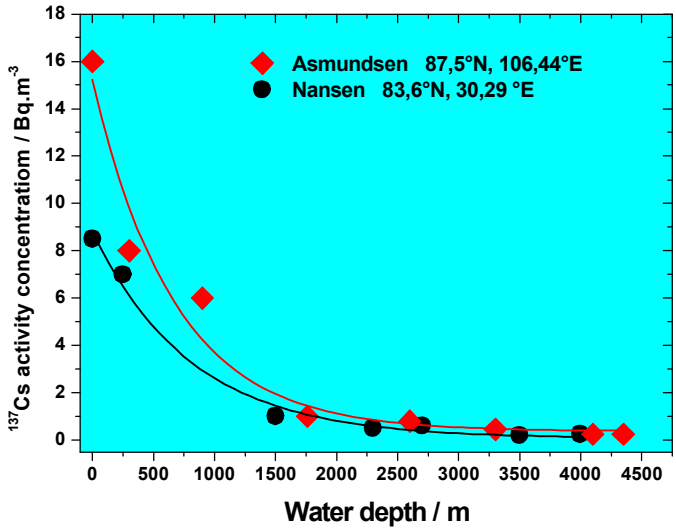

Figure 4-9
Depth profiles of ^{137}Cs activity concentration

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.

D.4 $^{239+240}$Pu isotopes in surface water

The Longitudinal distribution of $^{239+240}$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 $^{239+240}$Pu in the Norwegian Coastal Current (NCC) West Spitsbergen Current (WSC) to Frams Strait was in the range of 6 -7 mBq.m^{-3} . Entering the Polar mixed Layer (PML) at 81 °N the temperature drops from 5 to - 2 °C , and the $^{239+240}$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 $^{239+240}$Pu to about 7 -8 mBq.m^{-3}. The passing over the Lomonosov ridge at about 86 °N into the Markov basin, the activity concentration of $^{239+240}$Pu was about 8 mBq.m^{-3}. When crossing the Nansen basis the activity concentration of $^{239+240}$Pu was about 8 mBq.m^{-3} at 88 °N 80 °E. Further east towards the North pole the activity concentration of $^{239+240}$Pu was still about 6 m Bq.m^{-3}. But locally high activity of $^{239+240}$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 $^{239+240}$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.

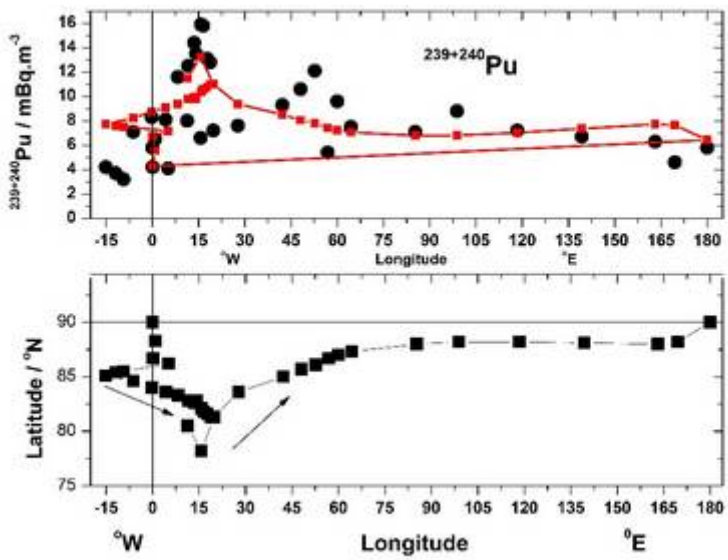

Figure 4-11
$^{239+240}$Pu activity concentration in sea water along the route. (Roos et al., 1998) The red curve represent the estimated values of PLS modelling.

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

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

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

D. Conclusions

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

The activity concentration of ^{137}Cs in the Arctic Ocean water was in the range of 8-12 Bq.m^{-3}. The activity concentration of ^{137}Cs increased to about 18 Bq.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 80-100 °E and locally high activity at 83 °N 10 °E.

The ^{134}Cs/^{137}Cs activity ratios was about 0.02 due to the contribution mainly from Sellafield and a few percent contribution from Chernobyl. The ^{134}Cs/^{137}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 $^{239+240}$Pu in the Arctic Ocean was in the range of 6-8 mBq.m^{-3} with locally high $^{239+240}$Pu activity concentration of 11 mBq.m^{-3} at 86 °N 48-53 °E, and 16 mBq.m^{-3} at 83 °N 10 °E.

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