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Persson, Bertil R; Holm, Elis; Josefsson, Dan; Eriksson, Mats; Roos, Per; Carlsson, Kjell-Åke

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Corresponding author:

Bertil R.R. Persson, PhD. MDhc, professor emeritus

Lund University, Dept. of medical radiation physics,

Barngatan 2, S-22185 Lund Sweden

E-mail: bertil_r.persson@med.lu.se

Radioactivity Exploration from the Arctic to the Antarctic.

Part 6: The Arctic Ocean-96 expedition

Persson, B. R. R., Holm, E., Josefsson, D. Eriksson, M., Roos, P. & Carlsson, K.-Å.

Medical Radiation Physics, Lund University, S-22185 Lund, Sweden

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 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. 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 ^{137}Cs, ^{90}Sr and ^{129}l activities and ^{134}Cs/^{137}Cs 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 ^{129}I and ^{134}Cs/^{137}Cs signals respectively have approximately the same spatial distribution in the Arctic Ocean surface layer. The activity concentration of ^{90}Sr in surface water is about 2 Bq.m^{-3}. For $^{239+240}$**Pu** 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 $^{239+240}$**Pu** are about 13 mBq.m^{-3} .

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}l measurements. It is assumed to be due to a contribution of Chernobyl fallout to the Arctic Ocean surface layers in the years around 1990.

Inventories down to 900 m death reveal that between 60-70 % of the ^{137}Cs, ^{90}Sr and 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 ^{137}Cs, 3.4 PBq for ^{90}Sr and 5.3x10^{28} atoms for ^{129}l.

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 Programme. Our project in 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 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,

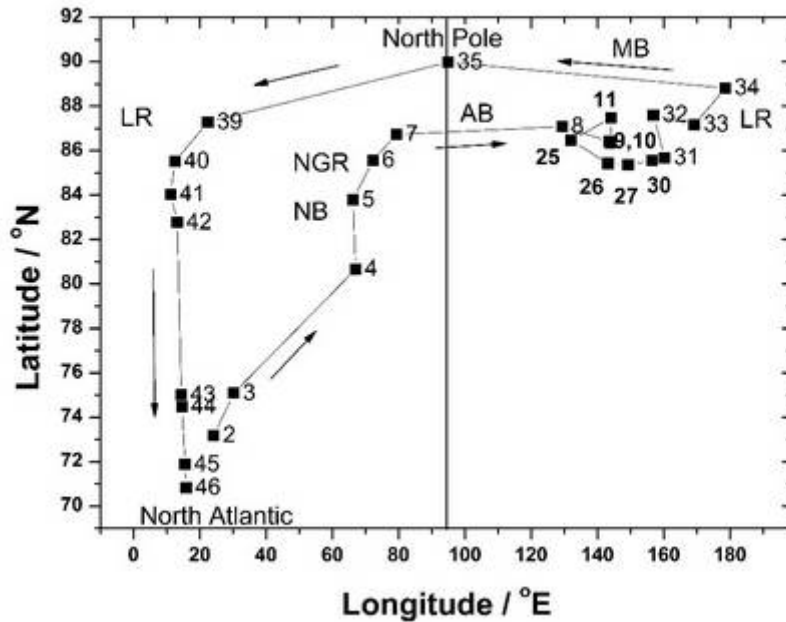**Figure 6-1**. The sampling point along the route of the Arctic Ocean 96 expedition Makarov Basin MB, Lomonosov Ridge LR, Amundsen Basin AB, Nansen Gakkel Ridge NGR, Nansen Basin NB.

The route of the expedition and the sampling locations at the Arctic Ocean-96 expedition is displayed in **Figure 6-1.**

Figure 6-2
The Swedish icebreaker M/S Oden 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) was added to the water in the vessels under continuous stirring during several hours to adsorb the dissolved Caesium. The AMP crystals were settled over night in the funnel shaped bottom of the vessel and 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.

Figure 6-3
Dan Josefsson manage the Go Flow bottle with 100 seawater sample. *(Photo Bertil Persson)*

Large volume samples (1000-2000 l) of surface water collected by our own pump by a tube hanging from the rail 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 ($Cu_2[Fe(CN)_6]$). The filters were dried and for transport to Lund, where they were ashed in an oven at 450 °C. The ash measured by high-resolution gamma spectrometry (HPGE or Ge-Li) for 1-2 days determined the $^{134}Cs/^{137}Cs$ activity ratio in the ocean water.

Plutonium and Americium was precipitated by adding sodium hydroxide to 200 l seawater collected in another 200 l vessel as above, after adding ^{242}Pu and ^{243}Am as radiochemical yield determinants. The hydroxide precipitate of was settled over night and tapped in 10 l bottles for transport to Lund where Pu and Am 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 l Go-Flow bottle . One sample was taken at temperature maximum at depths between 235-340 m, and another at depths between 850-1000 m. In order to study the particulate matter in the shelf seas, large volumes of water were passed through cotton-wound cartridge filters with a pore size of approximately 1 μm.

6.2.2   Sampling and radioactivity measurements of ^{90}Sr

Sea water samples for ^{90}Sr analysis were collected in a volume of 100 l 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 was allowed to settle overnight, and collected for in small bottles to be analysed in Lund. The oxalate precipitate was dissolved in high concentrated nitric acid in the weight to volume proportion 1:7. Strontium forms insoluble nitrates while calcium remains in the solution. 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 ($^{239+240}Pu$, ^{241}Am)

The precipitation vessels in our laboratory on board were filled with 200 l seawater and conc. 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. The NaOH in pellets was added to adjust the pH-value to be >l0. 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

collected in 10 l bottles, for transfer to the laboratory in Lund. At the laboratory the precipitates were dissolved in HCI and Pu and Am were co-precipitated with iron hydroxide at pH> l0 by the addition of ammonia. Plutonium and americium were then separated, following the methods described in IAEA Technical Report 295 (1989) and electroplated onto stainless steel discs (IAEA, 1989). The alpha activity of the disc 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 HNO_3 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.

Figure 6-4
The Arctic Ocean water and ice　　　　　　　　　　　　　　*(Photo Bertil Persson)*

6.3　Results and Discussion

6.3.1　^{134}Cs and ^{137}Cs in Arctic Ocean water

^{137}Cs in Surface water
The distribution of ^{137}Cs 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** a and b.

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 l5 °E in the Amundsen Basin. Lower activity levels are found in the Nansen and eastern Amundsen Basins. At the Gakkel ridge at about l0 °E, a front separates the high activities to the north from the significantly lower to the south. ^{137}Cs concentration falls from 10.7 to 5.2 Bq.m^{-3} and the ^{134}Cs/^{137}Cs activity ratio from 0.005 to 0.003.

Also in 1994 this front was observed in ^{137}Cs 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 sources enters the Arctic Ocean surface layer well east of the Barents Sea.
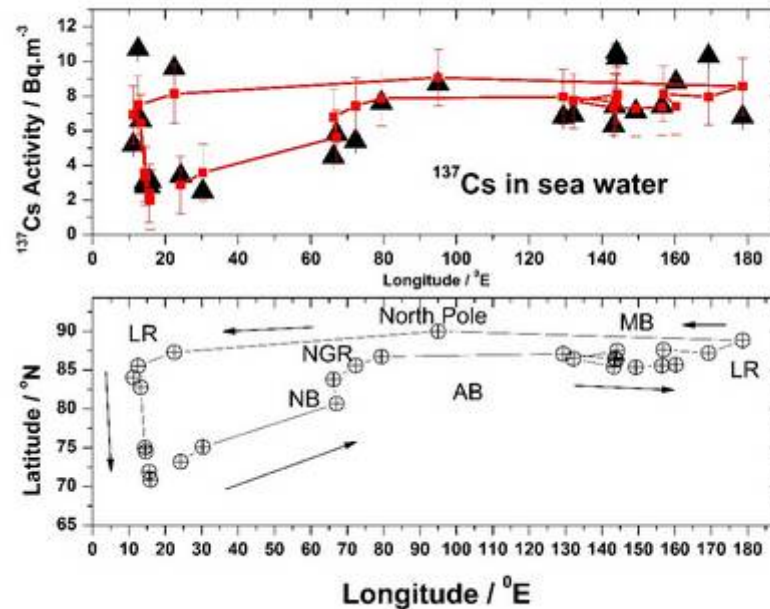

Figure 6-5
The distribution of ^{137}Cs in the surface layer at a depth of 8 m of the Arctic Ocean water at the various sampling locations along the route of the expedition. The predicted values given in red.

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. a1.,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 Bq.m^{-3} (Roos et. a1.,199S) and l5 Bq.m^{-3} (Ellis et. al., 1995) respectively.

^{134}Cs in Surface water
The distribution of ^{134}Cs / ^{137}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}Cs/^{137}Cs = 0.0849 + 1.53·10^{-3}·(Latitude°N) + 1,74·10^{-4}· (Longitude°E)
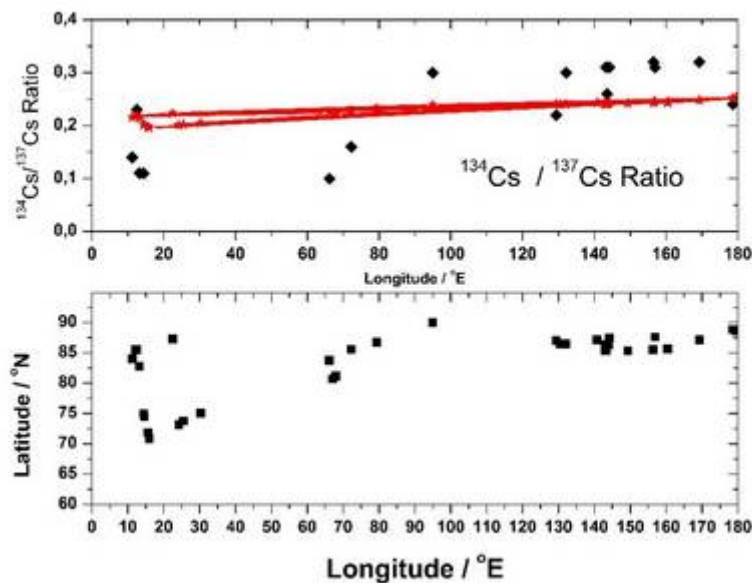Goodness of fit statistics (Variable Cs-134/): R² = 0,143

Figure 6-6
The distribution of ^{134}Cs/^{137}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 predicted values 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}Cs/^{137}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 0.02. The ^{134}Cs/^{137}Cs activity ratio, could therefore 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.**
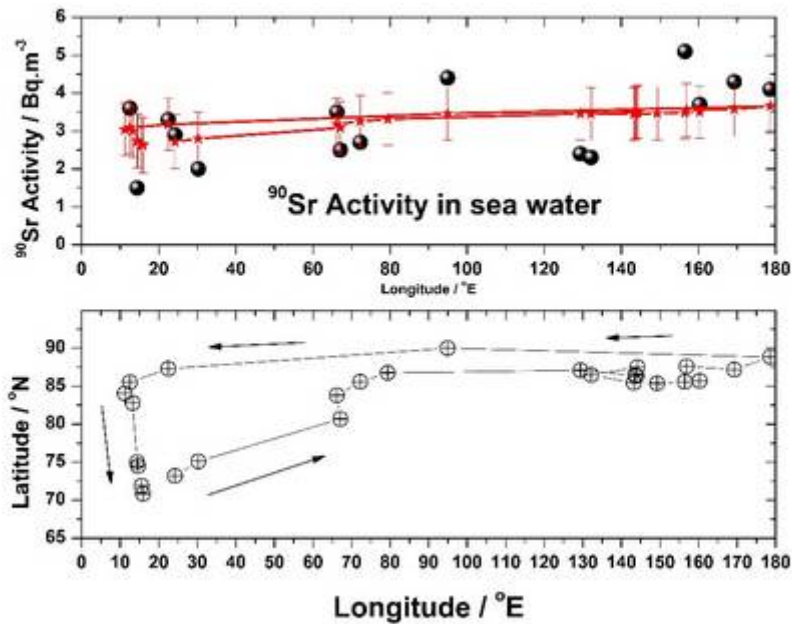
Figure 6-7
The distribution of ^{90}Sr in the surface layer at a depth of 8 m of the Arctic Ocean water
along the route of the expedition. The predicted values given in red.

Equation of the model:

^{90}Sr [Bq.m-3] = 0.263 + 0.326·(Latitude°N) + 2.75·10^{-3}Longitude°E)
Goodness of fit statistics (Variable ^{90}Sr [Bq.m^{-3}]): R² = 0,196

Mean: ^{90}Sr [Bq.m^{-3}] = 3.22 ± 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 2.3-5.1 Bq.m^{-3} in the central Arctic Ocean (Dahlgaard 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 10-20 Bq.m^{-3} in the 1990's with a ^{90}Sr/ ^{137}Cs/ run-off ratio above 2 (Chumichev, 1997, Dahlgaard et al., 1995). ln 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}Sr/^{137}Cs ratio is displayed in **Figure 6-8**.

Figure 6-8
The distribution of $^{90}Sr/^{137}Cs$-ratio in the surface layer at a depth of
8 m of the Arctic Ocean water along the route of the expedition.
The predicted values given in red.

Equation of the model:

$Sr^{90}/^{137}Cs$-Ratio = 2.530 - 0.248·(Latitude° N)+8.31·10⁻⁴ ·(Longitude °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
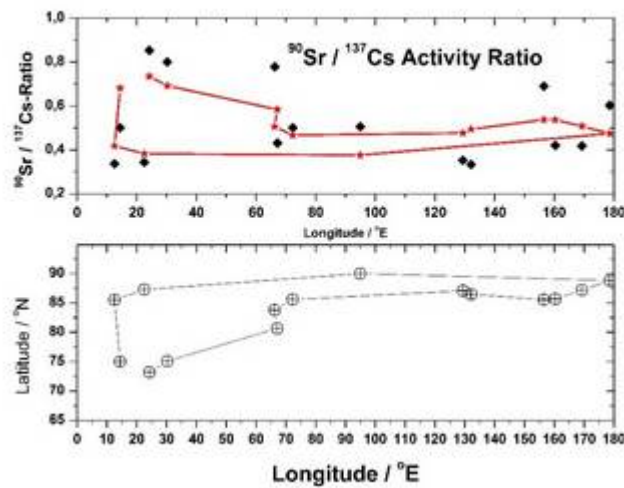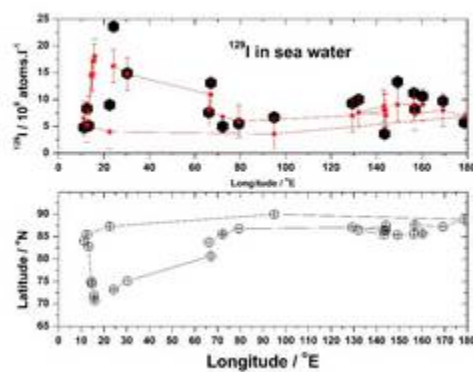the route of the expedition is displayed in **Figure 6-9**

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

Equation of the model:

^{129}I [x10⁸ atoms.l⁻¹] = 78,939 - 0,866· (Latitude°N) + 0.0268· (Longitude°E)

Goodness of fit statistics: R² = 0.568
Average: (9.260 ± 4,591)·10^8 atoms.l^{-1} = 1.5 ±0.7 femto-molar(fM)

A comparison of the distribution of the ^{129}I activity and the ^{134}Cs/^{137}Cs 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 ^{129}I concentrations at Station 34 in the Makarov Basin and at the North Pole Station. As an effect, both the ^{129}I activity and the ^{129}I/^{137}Cs 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 ^{137}Cs, ^{90}Sr, and ^{129}I in Subsurface water layers

The average depth distribution for all stations is illustrated in **Figure 6-10.**
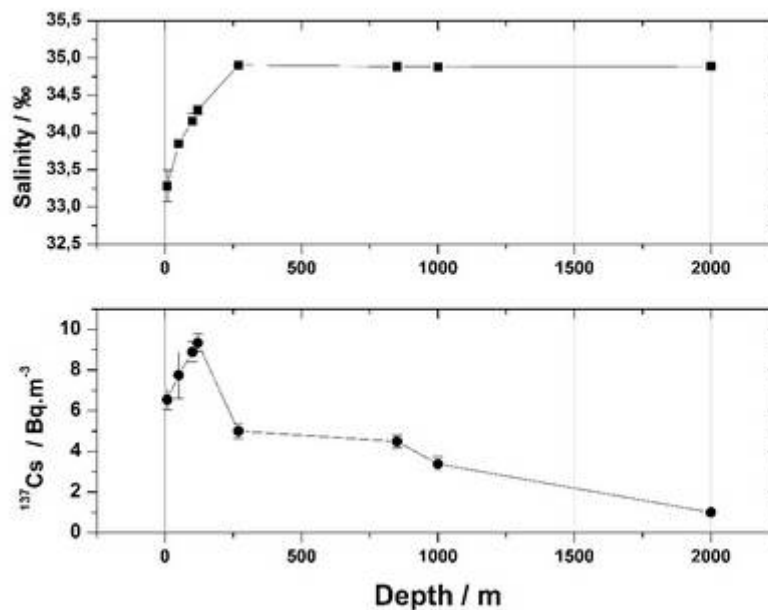
Figure 6-10
The average depth distributions of ^{137}Cs and salinity ‰ of all stations.

The depth distributions of ^{90}Sr and ^{129}I in the Arctic Ocean waters are displayed in **Figure 6-11** as averages of all sampling stations**.**

The estimated mean residence times for the surface layer are 2-6 years and up to l5 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 ^{129}I concentrations and the ^{129}I /^{137}Cs 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 ^{129}I and ^{90}Sr 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 ^{90}Sr in the surface layer are an effect of the riverine output, which enhances surface activity as compared to subsurface layers.

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).
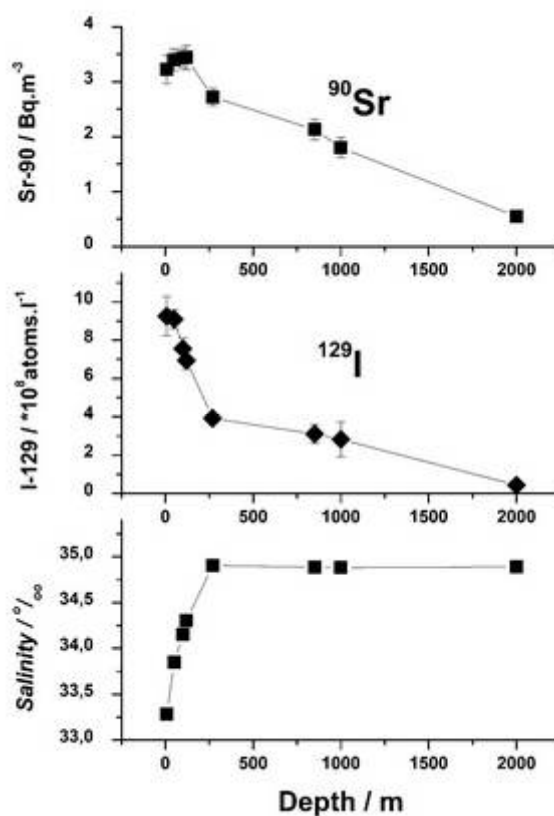
Figure 6-11
The average depth distributions of ^{90}Sr, ^{129}I and salinity $^o/_{oo}$
of all stations.

In subsurface samples, no ^{134}Cs activity was determined and thus no exclusive Chernobyl tracer is available. Elevated ^{137}Cs/^{90}Sr ratios may both be an effect of a higher amount of Chernobyl activity or of older Sellafield activity, and lower ^{129}I/^{137}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, however, are almost the same, indicating that the source composition is approximately the same, although diluted with low-activity deep water.

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

The activity concentrations of $^{239+240}$Pu, ^{241}Am in surface water of the central Arctic Ocean in 1996 are presented in **Figure 6-12. The a**verage activity concentrations of $^{239+240}$Pu 7.91 ±0,65 mBq.m^{-3}, of ^{238}Pu 0.47 ±0.07 mBq.m^{-3} and of ^{241}Am 0.38 ±0.11 mBq.m^{-3}.
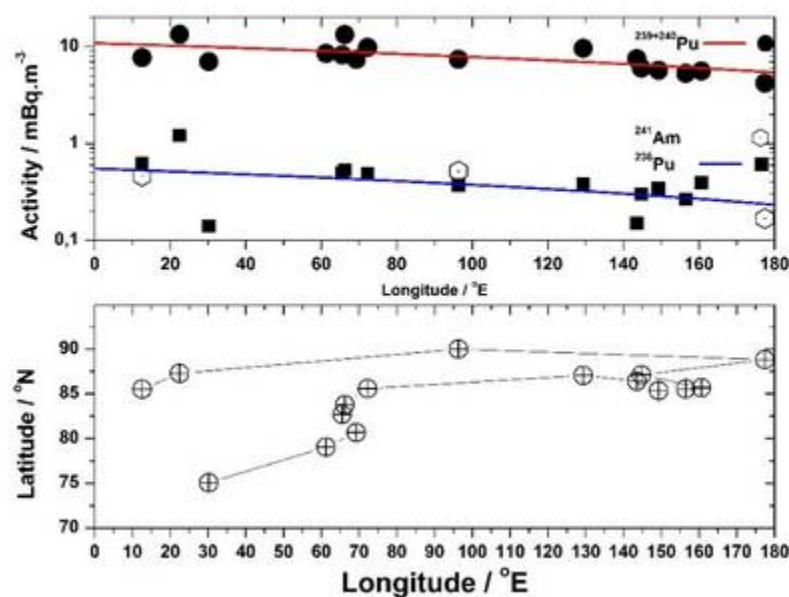
Figure 6-12
The activity concentrations of $^{239+240}$Pu, ^{238}Pu and ^{241}Am in surface water of the central Arctic Ocean in 1996.

^{239,240}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 has, 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 $^{239-240}$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}Pu concentrations. Although at 86ºN 160ºE where the sampling sites of

Polarstern and Arctic Ocean-96 coincide, the values of the ^{239,240}Pu concentrations are in good agreement about 5 mBq.m^{-3}.
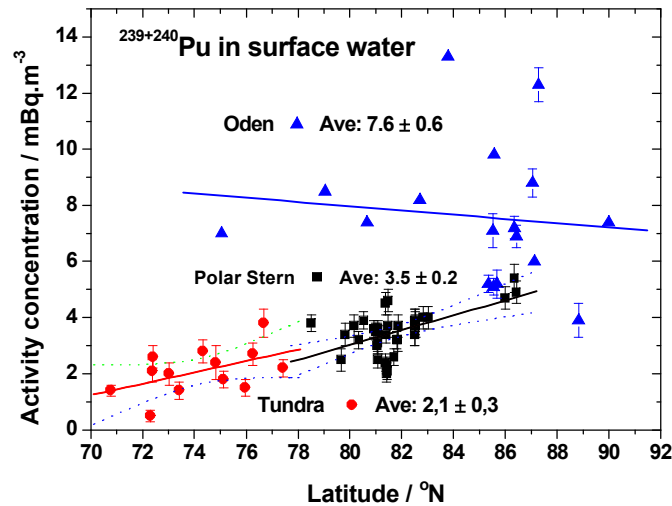
Figure 6-13a
The Latitudinal distribution of $^{239-249}$Pu activity concentration of in surface water measured by 3 different expeditions Tundra 1994, PolarStern and Artic Ocean-96
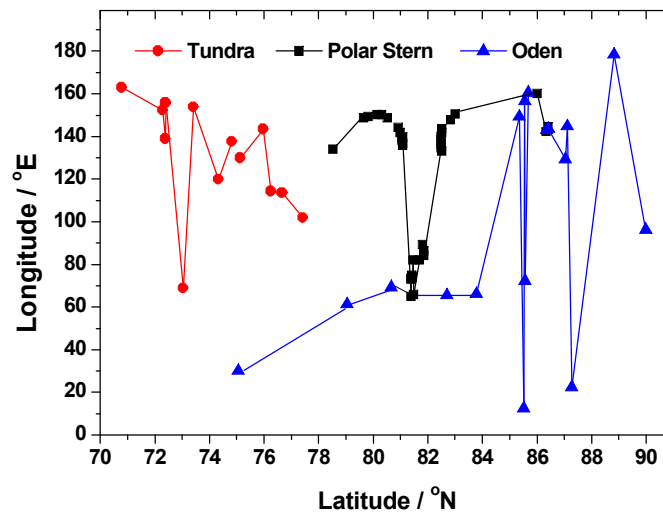
Figure 6-13b
The surface water sampling sites of the 3 different Arctic expeditions Tundra 1994, Polar Stern and Artic Ocean-96.

The lowest concentrations of $^{239+240}$**Pu** in the surface water (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 off $^{239+240}$**Pu** are somewhat higher, about 13.4 mBq.m^{-3}. The depth distribution of the Pu and Am isotopes displayed in **Figure 6-14** follow the same pattern as the salinity.
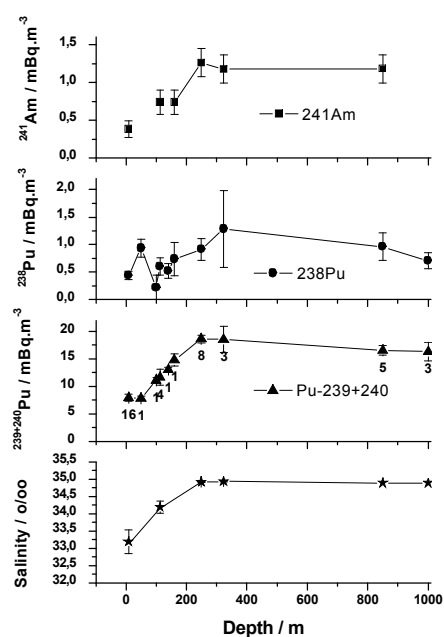
Figure 6-14.
Depth distribution of salinity and the activity
concentrations of $^{239+240}$**Pu,** ^{238}**Pu** and ^{241}**Am**
in the Arctic Ocean 1996.

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 shelf and at the shelf break. Comparisons with data obtained earlier for plutonium in the Arctic Ocean surface layer, reveal concentrations changing with time.

At the LOREX station in the northern Makarov Basin, the surface concentration was 14 mBq.m^{-3} in 1979 and had reached 17-18 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}Pu / $^{239+240}$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}Pu / $^{239+240}$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}Cs/^{137}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 **$^{239+240}$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 $^{239+240}$Pu /^{137}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 mBq/m^3 observed at the temperature maxima at 235-340 m depth and slightly lower ones, 14.2-17.0 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 **$^{239+240}$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}Pu/ $^{239+240}$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. Compared to the surface values, **^{241}Am/ $^{239+240}$Pu** ratios in the Atlantic layer at Stations A34 and A40 are increasing slowly with depth. The Norwegian and Barents Sea data showed a subsurface ratio of 0.15-0.25 in 1995 which, by sedimentation on the Barents Sea shelf, is assumed to get depleted on introduction into the central Arctic Ocean.

Figure 6-15
The sediment core sampler   *(Photo Bertil Persson)*

6.3.6 $^{239+240}$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 I 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 $^{234+240}$Pu at specific sampling station are given in **Table 6-1** and displayed in **Figure 6-17**. In the central Arctic Ocean, the integrated sediment activity of ^{137}Cs varied between 119 and 836 and of $^{234+240}$Pu between 5-30 Bq.m^{-2}. Highest inventories of both radiocaesium and plutonium 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.
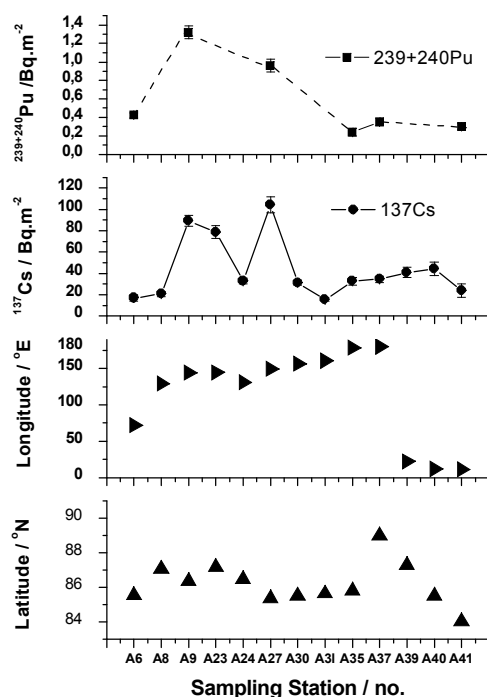
Figure 6-16 Sediment
The integrated sediment activity of ^{137}Cs and $^{234+240}$Pu at
specific sampling station with Latitude and longitude.

The depth distribution of the ^{137}Cs activity was rather uneven but with a penetration depth down to about 9-19 cm. The $^{234+240}$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 $^{234+240}$Pu/^{137}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 in1994 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.

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 $^{239+240}$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 l0 mBq.m^{-3} for $^{239+240}$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 $^{239+240}$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.

Table 6-1
Sampling location and integrated sediment activity of ^{137}Cs and $^{234+240}$Pu at the Arctic Ocean 96.

Station	Latitude °N degree	Longitude °E degree	Water depth lml	Pene-tration [cm]	^{137}Cs [Bq/m^2]	SD	$^{239+240}$Pu [Bq/m^2]	SD [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		
A3l	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

The mean ^{137}Cs activities during 196I-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 $^{239+240}$Pu concentrations are l.l.17.2 and 8.0 mBq/m^3. From the residence time of l0 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 the long residence times below 200 m depth (approximately above 35 years). 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 6-1** 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 $^{239+240}$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 l0 mBq.m^{-3} for $^{239+240}$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 $^{239+240}$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.

Figure 6-17
The North Pole with the flag of each participant´s country. Bertil Persson standing at
the old flag of Scania.

6.4 CONCLUSIONS

The highest ^{137}Cs. ^{90}Sr and ^{129}l activities and ^{134}Cs/^{137}Cs activity ratios are found in a band stretching from the Norther Lomonosov Ridge, over the north Pole and south along 10-15 °E to 85 °N. The European reprocessing and Chernobyl releases, as traced by the 129I and ^{134}Cs/^{137}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 **$^{239+240}$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 **$^{239+240}$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 Trough. 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 129l/137Cs 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.

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 $5.3\cdot10^{28}$ atoms of ^{129}I.

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