ICES Journal of Marine Science, 54: 333–340. 1997

Distribution and sedimentation of radionuclides in the Barents Sea

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Føyn, L., and Sværen, I. 1997. Distribution and sedimentation of radionuclides in the Barents Sea. – ICES Journal of Marine Science, 54: 333–340.

Analysis of sediment samples for cesium-137 from a wide range of positions has demonstrated an uneven distribution. The pathways and the possible origin of the radionuclide are discussed. The importance of sedimentation processes for the distribution pattern is demonstrated. The mean values of the upper centimetre of the bottom sediment are between non-detectable and 8 Bq · kg^{-1} (dry).

Key words: Barents Sea, radioactive cesium, distribution, sedimentation processes.

Received 18 January 1996; accepted 16 May 1996.

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Introduction

The Chernobyl accident in April 1986 and the consequent speculations about possible contamination of the marine environment, similar to the pronounced contamination observed in mountain lakes and on land in some areas of the Nordic countries, initiated a monitoring programme at the Institute of Marine Research (IMR) aimed at establishing the degree of contamination in areas important for Norwegian fisheries.

The sinking of the nuclear submarine ''Komsomolets'' in April 1989 combined with information about the dumping of radioactive material in the Barents and Kara Seas by the former USSR (Anon, 1993a) increased public and political concern about the possible problems of radioactive contamination in the marine environment.

In 1991 we started sampling of sediments, water and biota from the Barents Sea. The programme was later included in IMR's part of the AMAP (Arctic Monitoring and Assessment Programme). We decided, initially, to concentrate our measurements on sediments. The ability of sedimentary particles to remove a large proportion of the radionuclides from the water phase is referred to by, among others, Duursma and Gross (1971) and Meili (1994). As a first approach to determine a possible radioactive contamination of the Barents Sea, analysis of sediments was therefore chosen as most likely to give the best estimate of the degree of contamination.

Materials and methods

Sediment samples were collected from a set of 102 stations during cruises in the years of 1991, 1992 and 1993 (Fig. 1). Sampling procedure followed the guidelines established for monitoring in the North Sea (ICES, 1992). At each station three boxcorer shots were taken. The boxcore had an inner dimension of 30 cm × 30 cm. From each boxcore approximately 100 cm^2 of the upper 1 cm was collected. The samples from each station were mixed and stored at − 20°C until further preparation took place.

In the laboratory, the samples were freeze dried, homogenized, and filled in 215 ml polyethylene counting boxes before gamma-counting. The weight of the samples varied between 100 g and 200 g. The gamma-counting was performed on a Canberra HpGe-detector with 30% efficiency, an electric cryostat cooling system, and a 10 cm lead shielding. Counting time was approximately 22 h for each sample.

To establish the particle composition of the sediments, analyses were done to give the weight percentage of each sample in the following fractions: <63 μm, 63–125 μm, 125–250 μm, 250–500 μm, 500–1000 μm, 1000–2000 μm, and >2000 μm.

Results and discussion

Figure 2 presents the results of our determination of ^{137}Cs in sediments as becquerel per kg dry weight

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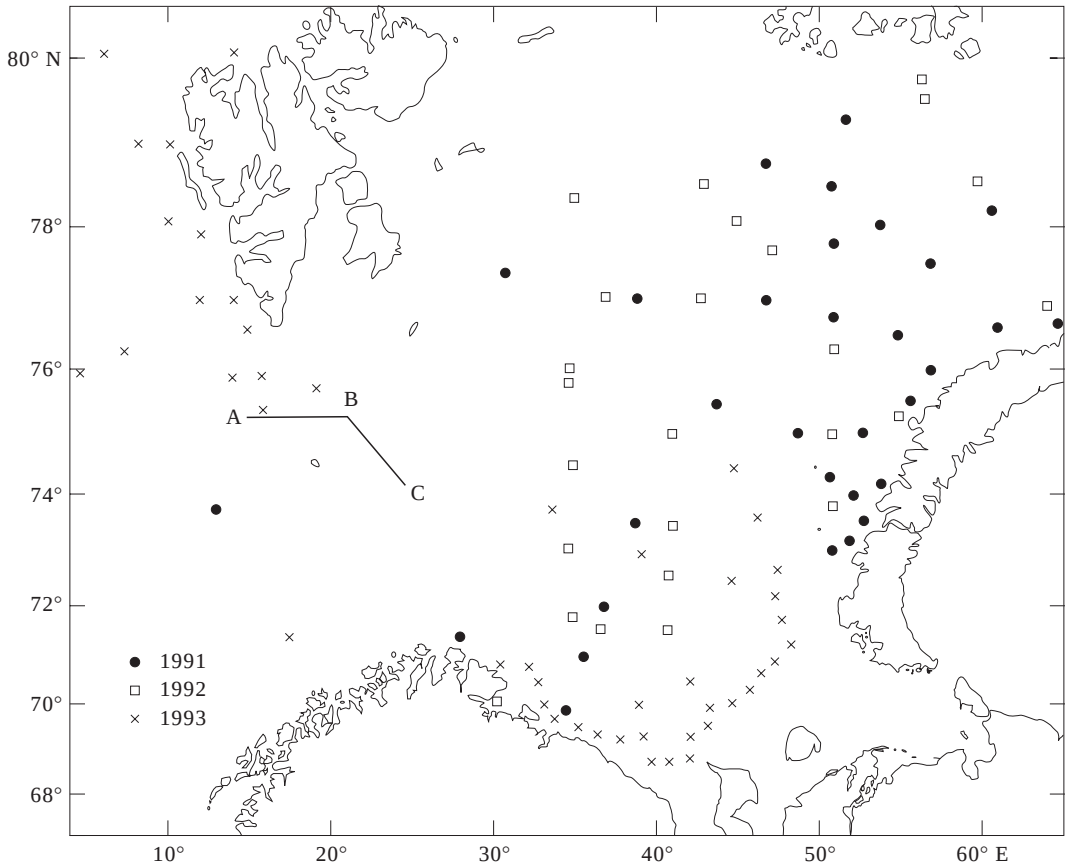

Figure 1. Sediment sampling stations in the Barents Sea and the hydrographic section A, B, C.

($Bq \cdot kg^{-1}$ d.w.). The overall mean value of the 102 stations in the Barents Sea and at the west coast of Spitsbergen is 3.2 $Bq \cdot kg^{-1}$ d.w., ^{134}Cs was not detected. The ^{137}Cs values varied between <1.0 and 8.6 $Bq \cdot kg^{-1}$ d.w. with the highest values found close to Spitsbergen.

For comparison, values in sediments of the North Sea, as reported in the North Sea Quality Status Report 1993 (Anon, 1993b), were found, in 1987, to be highest along the North Sea coast of the UK with values of 65 $Bq \cdot kg^{-1}$ d.w. and 6.0 $Bq \cdot kg^{-1}$ d.w. for ^{137}Cs and ^{134}Cs respectively. Sediment surface values for ^{137}Cs in the German Bight (in 1990) and the Skagerrak were measured up to 20 and 30 $Bq \cdot kg^{-1}$ d.w. respectively (Anon, 1993b). The Quality Status Report (Anon, 1993b) states that there was a decrease in the sediment values of radiocesium between 1986 and 1991 in the North Sea as a major source of contamination was the Chernobyl fall-out in 1986.

In the Barents Sea, the main sources of anthropogenic radioactive contaminants are:

- atmospheric fall-out (including Chernobyl fall-out),
- discharges from the nuclear industry in Europe,
- assumed contribution from land-based sources in northern Russia,
- dumped radioactive material of the former USSR,
- contribution from the Chernobyl accident via the Baltic.

As can be seen from Figure 2, there is an uneven distribution of ^{137}Cs in the sediments of the Barents Sea. Most pronounced are the relatively high values close to Spitsbergen and the low values in the south-eastern part of the Barents Sea. From our observations, it may be interpreted that of the many sources contributing to the radioactive contamination of the Barents Sea sediments, some have to be more important than others.

Global fall-out originates from the nuclear bomb tests beginning in the 1940s with a pronounced peak period in 1961 and 1962. The major tests took place in the northern hemisphere and especially in the Barents Sea area where there was observed an immediate response in the radioactive contamination of fish (Føyn, 1991; Føyn and Sværen, 1995). It may be assumed that the fall-out contribution from the nuclear bomb tests at Novaya Zemlya is evenly distributed throughout the Barents

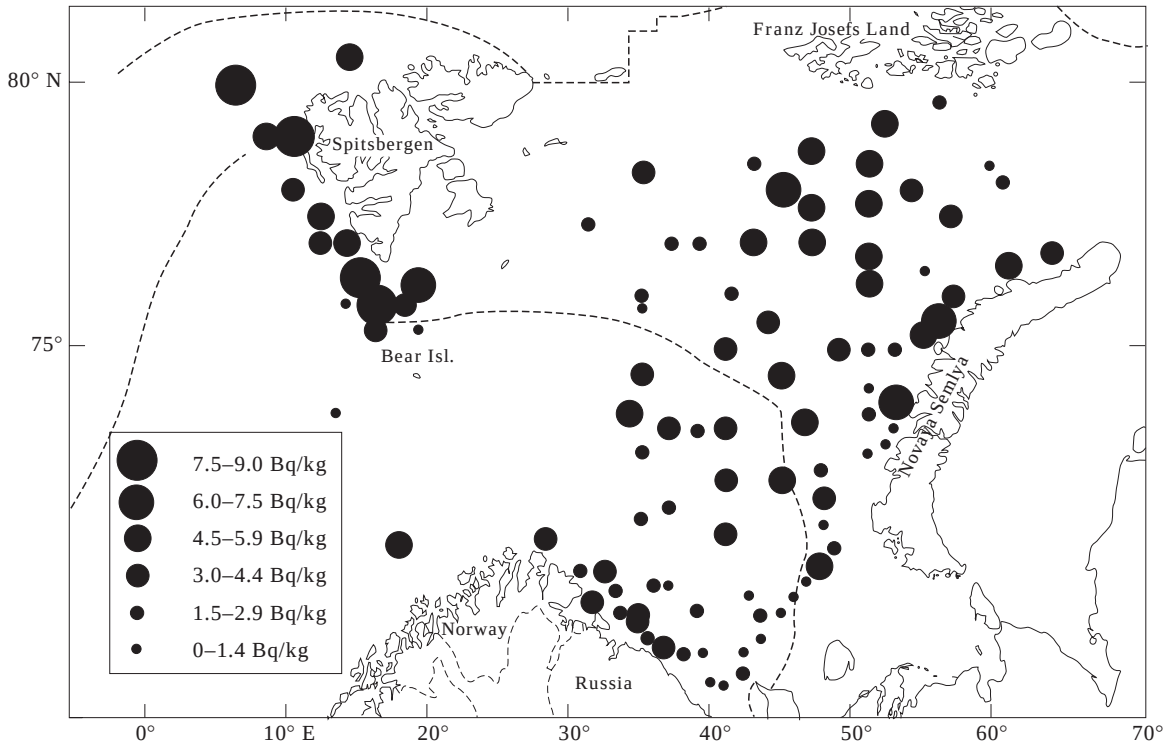

Figure 2. Cesium-137 values in Bq · kg^{-1} d.w. of the first cm of sediment samples from the Barents Sea.

Sea. Kautsky (1980) reported from measurements in 1969 only small variations of ^{137}Cs in sea water from the whole Barents Sea. However, fall-out on the ice-covered islands, i.e. the Spitsbergen islands, Franz Josefs Land, and Novaya Zemlya will be slowly transported towards the sea by glaciers and enter into the water through the melting of the ice. High ^{137}Cs values (>60 Bq · kg^{-1}) in sediments in some Spitsbergen fjords observed last year (1994) by Ivanov (pers. com., 1995) may indicate transport via glaciers to the fjords.

The transport of radiocesium, discharged from Sellafield into the Irish Sea, to the Barents Sea is well documented (Kautsky, 1980 and Kautsky, 1988). The transport routes of radiocesium observed by Kautsky (1988) follows the main current systems of the Atlantic current and the Norwegian coastal current, i.e. at the entrance of the Barents Sea one branch is towards the east and one is north towards Spitsbergen (Fig. 3). Kautsky (1988) found the highest values (40–50 Bq · m^{-3} of ^{134}Cs + ^{137}Cs) in the coastal current close to Norway as far east as 30°E in his observations in 1979 and values between 10 and 20 Bq · m^{-3} to the west of Spitsbergen.

From this distribution, it can be assumed that the main part of the radiocesium was transported eastward into the Barents Sea. The distribution pattern observed by Kautsky (1988) is in good agreement with the common transport pattern of fish larvae. Why do our observations of ^{137}Cs in the sediments not reflect the same pattern?

The content of radiocesium in the sediments reflects the grain size distribution in the actual sediment samples and, as pointed out by Meili (1994), cesium is often found in the clay fraction of sediments. Figure 4 shows a plot of the ^{137}Cs values against percentages of particles <63 µm in the individual samples. As can be seen, there is an overall increase in ^{137}Cs content in the sediments, with an increasing percentage of particles <63 µm in the samples. However, some out-lyers disturb this picture, of which one station with the lowest value of contamination and with >80% of the particles <63 µm represents the vicinity of the sunken nuclear submarine ''Komsomolets'' at 1670 m depth. The samples with relatively high contamination and a varying fairly low percentage of particles <63 µm represent the area around Spitsbergen.

To further elaborate the relationship between the ^{137}Cs content in the samples and the particle composition, a cluster analysis (STATISTICA, Clusteranalysis, K-means clustering) of the grain size data was performed. The program forms three clusters with the aim of: (1) minimizing variability within clusters; and (2) maximizing variability between clusters to get the most significant ANOVA results. The plot of means for each

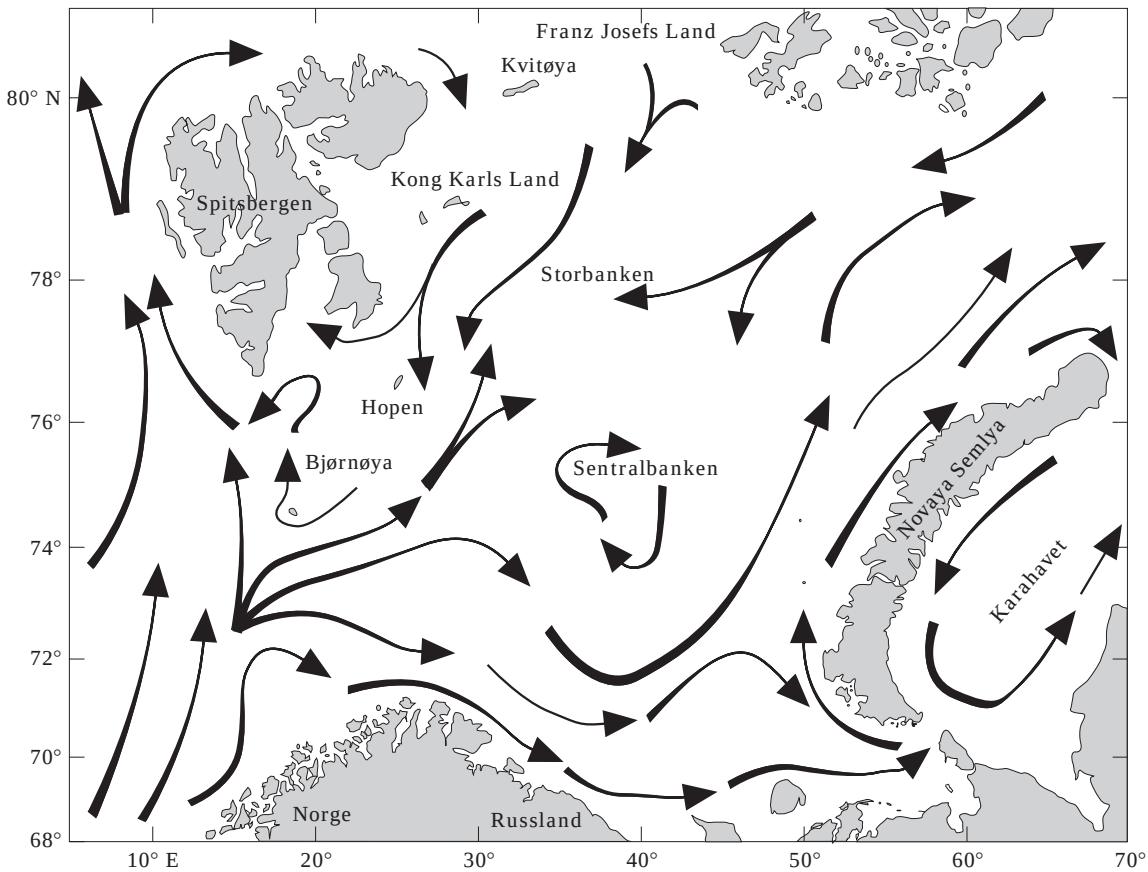

Figure 3. The main pattern of the surface water movements in the Barents Sea. Modified after Klungsøyer *et al.* (1995. Man's impact on the Barents Sea. Arctic, 48: 279–296).

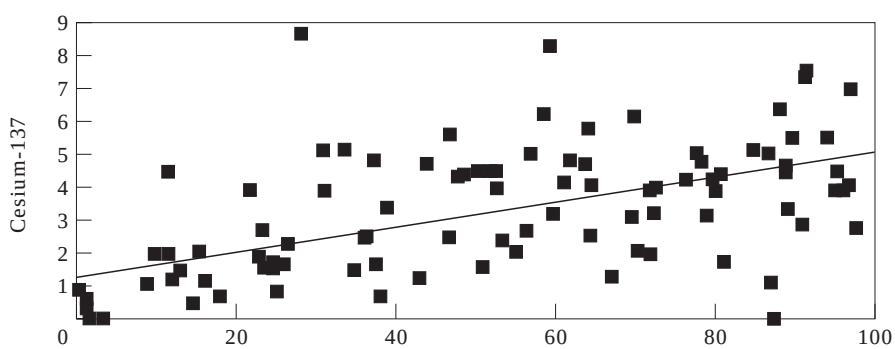

Figure 4. The observed ^{137}Cs values plotted against the percentage of particles in the sediment samples <63 µm.

of the three clusters is given in Figure 5. From the plot it can be seen that cluster no. 1 contains samples with particles mainly <63 µm, but also a relatively large fraction of particles >2000 µm. Cluster no. 2 contains samples with most of the particles in the fraction 125–250 µm. Cluster no. 3 contains samples with particles mainly <63 µm.

Figure 6 shows the ^{137}Cs values plotted according to how the particle composition of the corresponding samples is related to the three clusters. The sediment samples having most of their particles in the fraction <63 µm (cluster no. 3) have an average ^{137}Cs content of 4.0 Bq · kg^{-1} d.w. The major part of the samples belong to this type. The samples with the major part of the

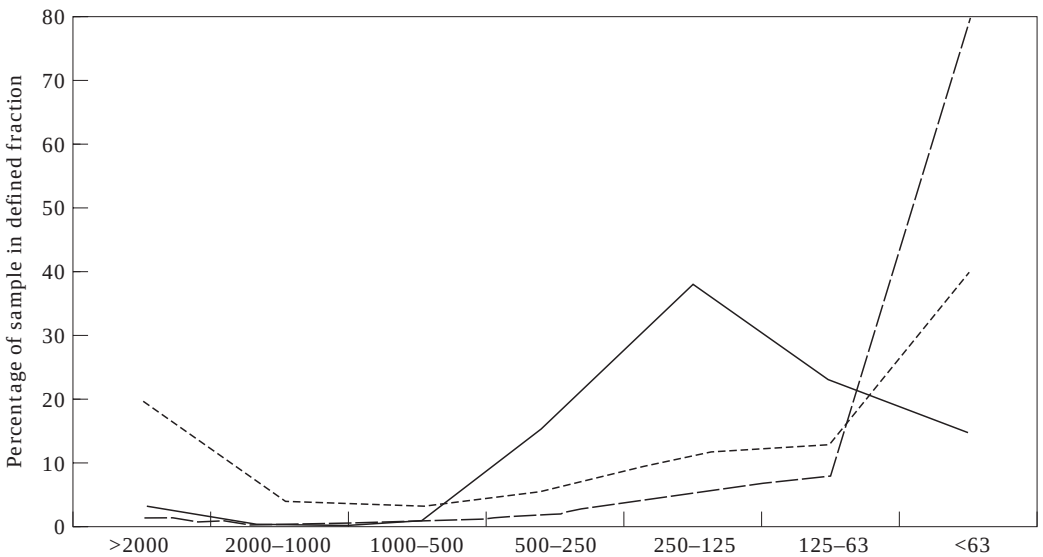

Figure 5. A cluster plot of the means for each of the three clusters according to the percentage of the samples in the defined particle size fractions. (— — —) Cluster 1, (——) cluster 2 and (– – –) cluster 3.

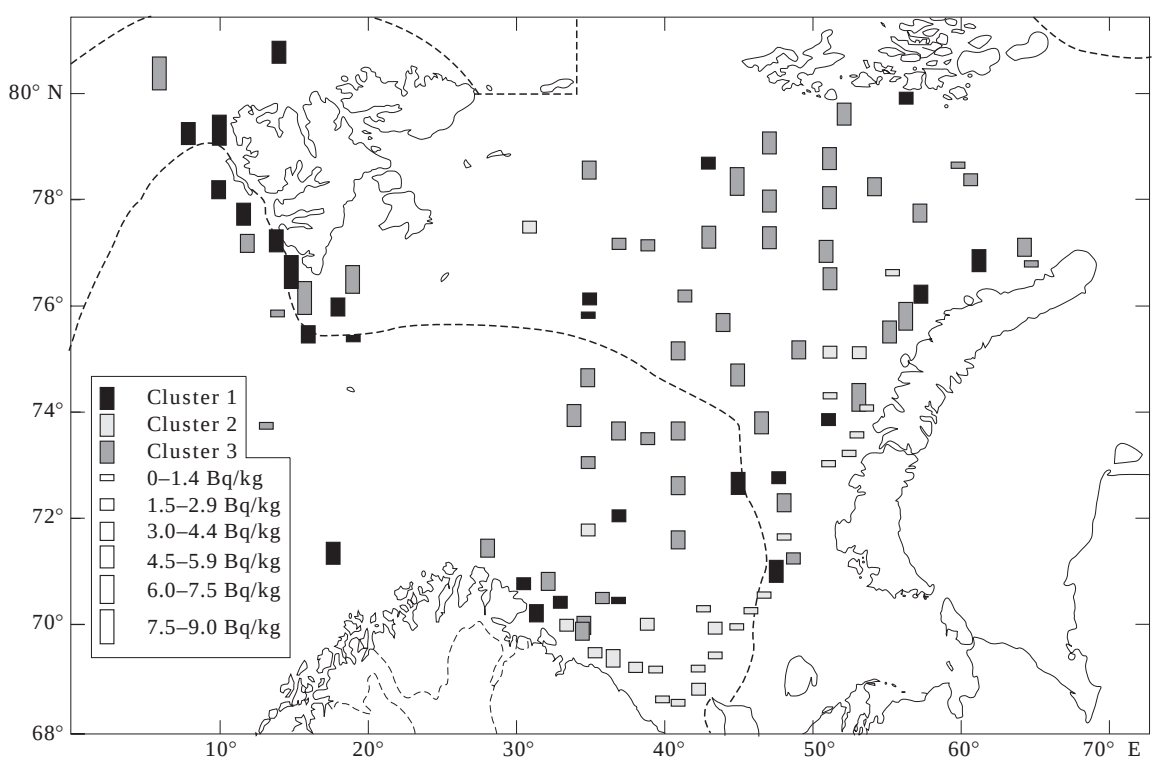

Figure 6. The ^{137}Cs values of the sediment samples plotted in relation to the three clusters.

particles having grain size 125–250 µm and the smallest amount of particles <63 µm (cluster no. 2) have an average ^{137}Cs content of 1.2 $Bq \cdot kg^{-1}$ d.w. These samples are mostly from the south-eastern part of the Barents Sea but also with some from the area close to Novaya Zemlya. The samples with the major part of the particles either in the fraction >2000 µm or <63 µm (cluster no. 1) have an average ^{137}Cs content of 3.6 $Bq \cdot kg^{-1}$ d.w. These samples represent most of the stations around Spitsbergen.

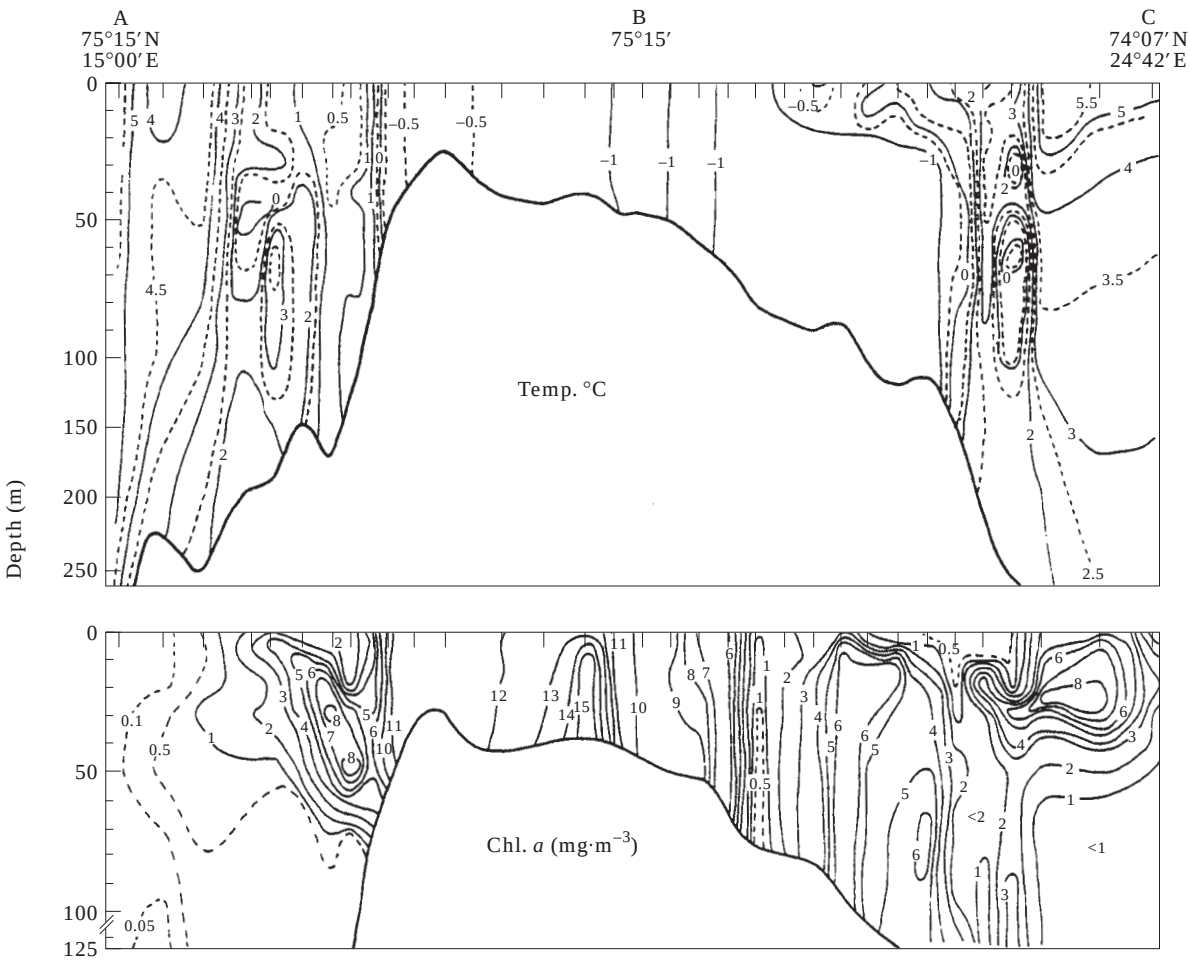

Figure 7. Temperature and chlorophyll *a* distribution in the hydrographic section A, B, C (Fig. 1) August 1984 (Rey, pers. com., 1995).

It seems that the samples collected from the area around Spitsbergen do not fit into the general view, as expressed by Meili (1994) that small sized particles, i.e. clay, will contain most of the cesium. As we find the highest values in this area where the sediments might be expected to contain less due to the grain size, and also the fact that this area should be less affected by European reprocessing water according to the normal water transport pattern, other explanations have to be found.

Dahlgaard (1994) summarizes concentration factors (CF) between phytoplankton and seawater for some elements giving a CF for cesium of 20. In comparison, Dahlgaard (1994) refers to plutonium and americium having CFs of 1×10^5 and 2×10^5 respectively while strontium has a CF of 3. This indicates that phytoplankton may play an important role in the contamination process of the sediments.

The positions of the sediment stations around Spitsbergen are located in a very geographically stable polar front area, and according to observations by Rey (pers. com., 1995), in this special regime the phytoplankton may actively act as scavenging elements. An example of his (Rey, pers. comm., 1995) observations, from 1984, are presented in Figure 7. The actual hydrographic section, A, B, C, is shown in Figure 1. As can be seen, there is an extensive primary production in the area, and high chlorophyll concentrations are found all the way to the bottom. The corresponding temperature profiles show clearly a non-stratified water mass which indicates an active transport of phytoplankton down to the bottom. The phytoplankton consists mainly of diatoms, and the process is, according to Rey (pers. com., 1995), not a normal sedimentation process, where only the ungrazed phytoplankton sinks out in a slow sedimentary process (normal for other parts of the Barents Sea), but an active and fast transport of phytoplankton down to the bottom.

If we assume that this active scavenging process is able to remove the cesium present in the water column

throughout the production period and actively deposit the cesium at the bottom, this can explain the higher degree of ^{137}Cs contamination in this area compared to the rest of the Barents Sea. The main sources of this contamination are most likely to be the water masses transported from the south along the Norwegian coast, i.e. discharges from European nuclear industry and from the Chernobyl contribution to the Baltic.

The branch of the Atlantic and coastal water moving eastward in the Barents Sea is spread and mixed with Arctic water, as is indicated in Figure 3. The Barents Sea is partly covered by ice during the winter. As the ice starts to melt in spring and the ice-edge slowly retreats northwards, the melt water flows on top of the more saline water creating favourable conditions for primary production (Rey and Loeng, 1985). This spring bloom is distributed in a fan-like manner along the ice edge (polar front) as the ice melts and recedes northwards.

The rich phytoplankton production is heavily grazed by zooplankton (Hassel *et al.*, 1991), and ungrazed phytoplankton sink to the bottom along with faecal pellets of zooplankton. As this production zone is moving northwards during the melting season, the sedimentation, and thereby the transfer of radiocesium to the sediments, will be more widely distributed. This distribution is in accordance with our findings of ^{137}Cs in the sediment of the north-eastern part of the Barents Sea.

The potential of the dumped radioactive waste, mostly in the Kara Sea, to contaminate the Barents Sea and the possible contribution from land-based sources in Russia, is due to westward transport from the Kara Sea to the Barents Sea. The main potential of a pronounced contamination is from the nuclear industrial complexes upstream on the rivers Ob and Yenesei in the north-eastern part of the Kara Sea. In addition, considerable potential sources of radioactive contamination of the Barents Sea are also found in the many nuclear submarines berthed in fjords of the Kola peninsula waiting for decommissioning. Three underwater nuclear tests (in 1955, 1957 and 1961, respectively), performed by the former Soviet Union in the eastern part of the Barents Sea close to the south coast of Novaya Zemlya, are also contributing to the contamination of the Barents Sea. According to Anon (1995) the present (1995) amount of ^{137}Cs from these tests would be about 150 TBq.

Føyn and Nikitin (1994) reported preliminary results from the third Norwegian–Russian expedition to the Kara Sea and fjords along the east coast of Novaya Zemlya, that contamination from the dumped solid radioactive material was only demonstrated in close vicinity to the dumped material.

Vinje and Kvambekk (1991) estimate that there is an average annual ice transport of 500 km^3 from the Kara Sea to the Barents Sea through the strait between Frans Josefs Land and Novaya Zemlya. In comparison they refer to the dominant ice flow in the Arctic, the Transpolar Ice Drift Stream pouring 4000 to 5000 km^3 of ice per year into the Greenland Sea. If some of the ice that enters the Barents Sea is frozen in the estuaries of the rivers Ob and Yenesei, there is a possibility that this ice may transport sediments frozen into the ice in the estuaries and thereby contribute to the radioactive contamination of the Barents Sea if the sediments of the two estuaries are contaminated. Aagaard (1993) mentions that large amounts of shelf sediments can be locked into the ice during freezing, and Weeks (1993) has discussed in more detail the mechanisms of incorporating bottom sediments in the ice.

A possible transport of contaminated sediments by the ice should be reflected in the ice melting zone where possible sediments contained in the ice will be released. Our data do not allow for any conclusion regarding a possible transport of radionuclides from the actual estuaries and the Kara Sea by ice to the Barents Sea. But it may be assumed that if such sediment transports occur, some signals of particular contamination of the sediments in the ice melting zone would be seen.

Conclusion

The sediments of the Barents Sea are far less contaminated by cesium-137 than the North Sea. The observed contamination is unevenly distributed, which is in accordance with the particle composition of the sediments, i.e. in sediments with a high percentage of fine particles (<63 μm) relatively high values of ^{137}Cs are found. However, in the area close to Spitsbergen the highest values of ^{137}Cs contamination are found. It is assumed that the particular primary production taking place in this area, due to the behaviour of the polar front, actively transports phytoplankton to the sediments and thereby scavenges the water for radiocesium throughout the production period.

To interpret a variable distribution of radionuclides in the sediments of a wide area as the Barents Sea, good knowledge of the hydrographic situation and also the primary production processes are needed.

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