# BALTIC SEA ENVIRONMENT PROCEEDINGS

No. 31

THREE YEARS OBSERVATIONS OF THE LEVELS OF SOME RADIONUCLIDES IN THE BALTIC SEA AFTER THE CHERNOBYL ACCIDENT

Seminar on Radionuclides in the Baltic Sea 29 May 1989 Rostock—Warnemünde, German Democratic Republic



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#### PREFACE

The Seminar on Radionuclides in the Baltic Sea was held on 29 May 1989 in Rostock-Warnemünde, German Democratic Republic, in conjunction with the fourth meeting of the Group of Experts on Monitoring of Radioactive Substances in the Baltic Sea (MORS), of the Baltic Marine Environment Protection Commission - Helsinki Commission.

This publication contains most of the proceedings presented at the seminar as well as conclusions prepared by the participants.

The authors of each scientific paper published in this seminar publication are responsible for their texts in their personal capacity as scientists. The participants of the seminar and the fourth meeting of the expert group MORS are responsible for the content of the summary and conclusions.

OBSERVATIONS OF THE LEVELS OF SOME YEARS THREE SEA AFTER THE CHERNOBYL BALTIC RADIONUCLIDES IN THE on Radionuclides in the Baltic ACCIDENT: Sea. Seminar 29 May 1989, Rostock-Warnemünde, German Democratic Republic

#### INTRODUCTION

Anneli Salo, Chairman of the Seminar and MORS 4

Radioactive substances are present in oceans both naturally and as a result of human activities.

Some of naturally occurring radionuclides have such long half-lifes that they have been in existence since the origin of the solar system. Other naturally occurring radionuclides are produced by the interaction of highenergy cosmic radiation with the atmosphere. The former category reaches the oceans mostly as a result of run-off from weathered rocks or by decay of the primordial substances in the water itself. The latter are primarily deposited on the surface of the ocean by precipitation.

Those human activities which have added to the inventories of ocean radionuclides are testing of nuclear weapons, operational discharges of effluents from nuclear reactors and fuel reprocessing plants, ocean dumping of low level radioactive waste and nuclear accidents. Of these activities only nuclear weapons testing has global impact on the ocean contamination by certain radionuclides, see Tables 1 and 2 (1,2,3,4,/). Other human activities have mainly had local impact on radionuclide concentrations in the oceans, except the releases from the Sellafield reprocessing plant, which have been detected also in the Baltic Sea and up to the Arctic Ocean after a peak release period in the 70's. The radionuclides originating from the Chernobyl accident are another exception as regards the contamination of the Baltic Sea. Table 1. Fallout in global oceans [1,2,3,4]

	Input PB	Input PBq	
	Northern Hemisphere	Southern Hemisphere	PBq
14 C	Not less than 4	0	170
90 Sr	250	120	440
<sup>1 3 7</sup> Cs	410	200	640
<sup>2 3 9</sup> Pu	3.3	1.6	
<sup>2 4 0</sup> Pu	2.2	1.1	16

Table 2. <sup>137</sup> Cs inventories for all world oceans [2]

	PBq		GBq km <sup>-2</sup>
	Water column	Sediment	Areal
N.Atlantic	119(1	2.1(2	3.4
S.Atlantic	28	3.6	0.4
Caribbean	11	0.6	2.4
Mediterranean	10	0.5	4.1
Black Sea	1.8	0.2	3.9
N.Pacific	210	4.8	2.7
Bering Sea	4.6	0.2	2.0
S.Pacific	102	4	1.1
Indian	86	3.7	1.1
North Sea	0.9	1.0	1.5
Baltic Sea	0.5	0.05	1.3
Norwegian Sea	6.8	0.1	4.9
Greenland Sea	4.7	0.1	3.9
Arctic	47	5.2	5.6
	640	30	

(1 +5 Sellafield

(<sup>2</sup> +4 dumping

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) /5/ estimated the total release of <sup>137</sup>Cs from Chernobyl accident to be 70 PBq. Of this they estimated only 4.7 PBq to have deposited on the oceans. This figure is challenged by the present studies within the MORS-programme as being too low. The release plume from Chernobyl moved over the Baltic Sea to Sweden and Finland. Part of the radionuclides deposited on the sea surface giving rise to initial differences in radionuclide concentrations of water in the various Baltic Sea areas. Similar uneven radionuclide distribution pattern on the surrounding land areas has been explained by observed rainshower patterns.

The present document discusses the status of radionuclide contamination in the Baltic Sea, in particular, the addition from the Chernobyl accident.

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- 5 United Nations Scientific Committee on the Effects of Atomic Radiation, 1988. Sources, Effects and Risks of Ionizing Radiation, Report to the General Assembly, with Annexes. United Nations Sales Publication No. E. 88. IX United Nations, New York

THREE YEARS OBSERVATIONS OF THE LEVELS OF SOME RADIONUCLIDES IN THE BALTIC SEA AFTER THE CHERNOBYL ACCIDENT; Seminar on Radionuclides in the Baltic Sea, 29 May 1989, Rostock-Warnemünde, German Democratic Republic

#### SUMMARY AND CONCLUSIONS

The summary was adopted by the fourth meeting of the Group of Experts on Monitoring of Radioactive Substances in the Baltic Sea (MORS) on 2 June 1989. (The list of participants of the meeting is included in this report on page 151).

Based on the oral presentations at the Seminar and the discussions of the preliminary papers, the following summary can be made:

Water: Since the Chernobyl accident on 26 April 1986, a number of cruises have been made in the Baltic Sea and many water samples have been taken and analyzed. The results from the analyses of these samples show that - from a radiological point of view - the most important artificial radionuclides in the Baltic are Cs-137 and Cs-134. In addition, Sr-90 and transuranics have been studied.

Chernobyl derived radionuclides have been observed over all areas of the Baltic Sea. In 1986 the highest concentrations were measured in the Bothnian Sea, the Gulf of Finland and at the Mecklenburg Bight. In 1987 and 1988 dilution and removal processes reduced these initial high concentrations. Concentrations increased in water from certain areas during 1987 and 1988 due to advective transport. Run off had its highest influence during 1986.

There are still some questions concerning the behaviour of radiocaesium and its physical/chemical form immediately after the accident.

At an informal meeting of experts in Risoe, Denmark 30-31 January 1989, available data was used to calculate the inventories of radionuclides. A preliminary estimate showed that the inventory of Cs-137 in the Baltic water mass was 324 TBq during 1983; 4620 TBq by September 1986; and 3700 TBq during August 1987. The Sr-90 inventory was 416 TBq in 1983; 460 TBq in October 1986; and 391 TBq in August 1987.These calculations have still to be refined to get more accurate results.

The inventory of Sr-90 in the water column was altered very little by fallout from Chernobyl. The contribution of Cs-137 from Chernobyl in 1986 has increased the total water mass inventory by a factor of 15, and will influence the total concentration Cs-137 in different compartments of the Baltic Sea for a long time.

The Cs-137 in the water column was depleted by 25 per cent between 1986 and 1987, based on preliminary calculations made in Risoe. This loss of Cs-137 is difficult to explain, but one possible explanation could be the high sedimentation of radiocaesium both in the Gulfs of Bothnia and Finland.

<u>Sediments</u>: A significant fraction of the Cs-137 is found in the Baltic Sea sediments. Based on pre-Chernobyl data, an inverse correlation was demonstrated between water depth and accumulated caesium in the sediment. This factor partly explains the low deposition of Chernobyl caesium in the sediments of the Baltic Proper. Another important factor is the permanent discontinuity layer preventing surface water from contact with the sediments in this area, as well as e.g. the lower sedimentation rate in this area. Preliminary calculations of the Chernobyl-drived inventory for Cs-137 in the sediments was 520 TBq in 1986 and 856 TBq in August 1987. These figures are based on measurements of Cs-134 and an initial activity ratio of 0,52. Direct calculations from the measured Cs-137 content in the sediment gives an inventory of 1466 TBq in August 1987 and is the sum of Cs-137 from all sources. This inventory may be underestimated due to loss of activity during sampling.

The Chernobyl impact on transuranic inventory in the Baltic Sea has been rather low. Most of the Pu-239/240 inventory is now found in the sediments. The redox conditions in the sediment may play an important role in regulating the concentration of the transuranic elements in the sediment. Sr-90 levels in sedimentory deposits is very low.

Biota: Presently the only important radionuclides from Chernobyl in the biota from a dosimetric point of view are Cs-137 and Cs-134. These radionuclides can be found in biota all through the Baltic.

The quantity of Cs-137 and Cs-134 in plankton may be of interest in calculating the removal of Cs from the water column.

In fish the concentrations differed with the trophic level. The activity of radiocaesium in Baltic fish is low, compared to fish from inland lakes. In some predatory fish radiocaesium was still increasing in 1988, but levels in most other fish have started to decrease. This trend can also be seen in the bioindicator Fucus vesiculosus.

The highest concentrations in fish caught in the Baltic Sea were found in the freshwater predatory fish pike, perch and pike-perch.Among the true marine fishes, cod had the highest content of caesium. Also Ag-110m and Sr-90 have been measured in cod. The amount of radionuclides in the fish and their variation in time, can be rather well explained by the behaviour of the water masses in the Baltic; the penetration of the radionuclides to deeper water layer and seasonal migration of Baltic herring.

#### Radiation doses due to fish consumption

Individual doses from fish consumptions have been estimated. The dose from Sr-90 is some orders of magnitude lower than the corresponding dose from Cs-137 and Cs-134.

Assuming an annual consumption of 12 kg of Baltic Sea fish, an individual effective dose equivalent of 0,004 mSv was found for the Federal Republic of Germany for 1988, based on measurement in December 1988. Data from the German Democratic Republic give the same result for the internal dose. Researchers from Finland based their calculation on an average consumption of 7,7 kg of Baltic herring and 5,8 kg of other sea fish per capita and year. This would give the following average internal doses from Cs-137 and Cs-134:

1986	0,005 mSv
1987	0,008 mSv
1988	0,009 mSv.

Compared to the general background radiation doses to man - which is of the order 2 mSv per year - the contribution from Cs-137 and Cs-134 from Chernobyl is seen to be less than one per cent. The dose received from swimming, boating, sunbathing on beaches, and handling fishing gear would be very much less.

#### Matters related to Quality Assurance / Quality Control

Several discussions, reports and new data related to analytical quality assurance were presented to the participants attending the meeting.

- 1. The observer from the IAEA, International Laboratory for Marine Radioactivity, described some preliminary data for man-made and natural radionuclide concentrations measured in an intercomparison sample of Baltic Sea sediment. Scientists from 75 institutes, including 14 participants to the MORS program, submitted analytical results for different radionuclides. Although some outliners were identified, the MORS analytical data base was in general of good quality and in agreement with the global set of results.
- 2. The observer from ICES (International Council for the Exploration of the Sea) described current ICES guidelines for monitoring of non-radioactive contaminants in sediments and recommended procedures for sampling sediments. Although the programme is not concerned with radioactive contaminants, the methods applied for sampling and processing samples for non-radioactive and radioactive contaminants should be comparable. The guidelines therefore, are extremely relevant to the MORS effort.
- 3. Translation of a ten-year-old report on Nordic Intercalibration exercise was presented by the participant from Denmark showing analytical results for radionuclides in sedimentary material extracted from five different sediment samplers; the aim being to see if the different samplers showed different results. This experiment was conducted several years ago and sediment samples were distributed to different laboratories which reported significant differences in radionuclide concentrations. These results showed there was a definite need to continue analytical intercalibrations of this nature to help resolve these differences. Similar "sampler" intercalibration studies with the same objective were recently conducted with participation by several HELCOM members states. The Federal Republic of Germany samples have been collected and distributed to 17 investigators but results are only now being collated and were not available for discussion at this meeting.

The Finnish results from sediments collected after Chernobyl with 2 types of gravity corers and 1 box corer consistently showed that the sediment from the STUK corer contained higher inventories of Cs-137 than material extracted from the box corer and smaller diameter gravity corer. It is difficult to explain the differences encountered in the concentrations measured at this time.

#### Future work

From the oral presentations and from the discussion one could find some areas where there seemed to be a more general agreement that further work should be carried out. Among the more important were the following:

- There should be continued sampling and measurement of the concentration of Cs-137 in the different compartments of the Baltic Sea.
- The inventories of Cs-137, Cs-134 and Sr-90 as well as the transuranics should be calculated both in the water masses and in the sediments.

- Special attention should be paid to the methodological problems encounted in this work.
- Work should be carried out in order to arrive at better models to predict the concentration of Cs-137 in the Baltic Sea as well as the radioation doses subsequent of normal and accidental releases. The fallout from Chernobyl gives a good possibility to validate models for for the Baltic Sea.
- There is a need for more intercalibration exercises for both sampling comparison as well as comparison of methods used to prepare samples and measure radionuclides. Sediment sampling seems to require immediate attention.
- There should be a discussion on how to present the result of the dose calculations or concentration of radionuclides in the environment. The result should be compared to our natural radiation environment. Radiation doses should be compared to the impact from other toxic or harmful substances in the Baltic Sea.
- Give examples on how the data base built up in the MORS group could be used for assessment. This would help in arriving at conclusions on the future collection of data, and how this information could be presented for use inside HELCOM and to fullfill the obligation placed upon MORS.

### RADIATION SITUATION IN THE BALTIC SEA IN 1986 IN SEA WATER AND SEDIMENTS

Survey draft on the materials of the HELCOM prepared by experts of the USSR \*

\*) Remark by the Secretariat:

In spring 1986, after the accident of the Chernobyl Nuclear Power Plant in April, the Executive Secretary of the Helsinki Commission, Prof. Harald Velner, requested the seven Baltic Sea States - Contracting Parties to the Convention on the Protection of the Baltic Sea - to intensify their research activity on radioactive substances in the Baltic Sea area. In 1987, the Helsinki Commission requested the USSR to compile the information received from the Baltic Sea States and to provide the compilation for the use of the Commission. Assuming that there was a common interest to the compilation prepared by experts in the USSR, this document was made available as a preprint copy suitable for citing in open literature in 1988 and a list of some relevant documents published by the Baltic Sea States was attached.

#### ANNOTATION

The survey contains summarized data on radioactive contamination of the Baltic Sea for the period from May to November, 1986. The survey is composed of the materials submitted to the HELCOM.

The data on contamination of the surface water layers of the Baltic Sea with radioactive cesium are presented in map-charts for three observation periods: May-June, July-August, October-November, 1986. A nonuniformity of contamination has been noticed. Areas with increased contamination have been detected. The results are considered concerning the depth distribution of radioactive cesium. The difference is shown of the rate of penetration into the deep water layers in different parts of the sea. Data are presented on presence of cesium-134, as well as cesium-137, in bottom sediments and hydrobionts of the Baltic Sea. The results of radioactive cesium determination in river waters are presented. It is shown that the change of the radiation situation that took place in the Baltic Sea in 1986, is fully determined by radioactive cesium. The data are compared of atmospheric fallout of radioactive cesium and its content in the water of the Baltic Sea.

The survey of 1987 summarized the data on radioactive contamination of environmental objects in the Baltic Sea region in April-July, 1986<sup>\*)</sup>. It was shown on the basis of analysis of the results available then, that due to air transport and further atmospheric fallout, the radioactive substances that entered the atmosphere as the result of Chernobyl NPP accident, reached the Baltic Sea region and caused additional contamination against the background of the global one. Most part of the radioactive aerosols from the Chernobyl NPP accident fell out in the region of the Baltic Sea practically during a short period of time: from the end of April to the end of May, 1986, but the density of the fallout was different in different parts of the Baltic region. Investigation of individual areas of the Baltic Sea clearly showed nonuniformity, "spottiness" of contamination, reflecting different densities of atmospheric radioactive fallout. Among the detected radionuclides the radioactive isotopes of cesium: cesium-134 and cesium-137, were the main dose-forming ones. Their activity ratio (cesium-134/cesium-137) was 0.55 in the initial period<sup>\*\*)</sup>.

Observations carried out in April-July 1986, covered a considerable part of the Baltic Sea water body, but separate parts, in particular, the Gulf of Bothnia and the northwestern part of the Baltic Sea Proper, remained unexplored. Besides, the results presented in that period, in most cases were preliminary ones.

The information on which this survey draft is based, embraces the period from the end of April to November, 1986. The list of the documents submitted to the HELCOM for survey is given in the list of the sources used | 1-11 |.

\*)Radioactive contamination of environmental objects in the Baltic Sea region in April-July, 1986. HELCOM 8/4c/6.20.02.87 \*\*)In this period iodine-131 was also among the main dose-forming isotopes. The obtained materials contain the results of investigation of practically the whole aquatory of the Baltic Sea; they cover a long observation period, more than half a year. In these materials the data submitted earlier are defined more exactly.

Most experimental material contains the results of determination of radioactive cesium as the most informative in the given case of contamination. A great amount of data concerns cesium-134 and cesium-137 determination in the sea water. As the observations of the radioactive cesium content in the Baltic Sea waters were carried out at different times, it seemed expedient to choose three periods of time and to combine the data thereof in map-charts. In Figs. 1, 2 and 3 there are the results of cesium-137 determination in the surface waters of the Baltic Sea in the periods: May-June (Fig. 1) | 1, 3, 6-11 |, July-August (Fig.2) | 1, 6-8, 10, 11 | and October-November (Fig.3) 3-8, 10, 11 . Side by side with cesium-137 determination, cesium-134 was determined in all samples. The activity ratio cesium-134/cesium-137 was 0.55 in the initial period of observations, which corresponded to the activity ratio of these nuclides in the damaged block of the Chernobyl NPP at the time of the accident. In all the observations this value was the tracer of the given type of contamination and enabled to fix it reliably. The value of the cesium-134/cesium-137 activity ratio decreased with time due to preferential decay of cesium-134.

Different periods of observation and in different areas of the sea hinder the comparison, of course, however the general picture of the radiation situation in the Baltic Sea in 1986 appears clearly enough. First of all, the nonuniformity, "spottiness" of the Baltic Sea's radioactive contamination is confirmed. This nonuniformity was the consequence of atmospheric precipitations of different intensity in the time of passage of air masses contaminated with radionuclides from the Chernobyl NPP accident, and as a result of it, a very nonuniform density of radioactive fallout. The density of the atmospheric fallout, in its turn, reflected in the distribution of the radioactive contamination, cesium-137, in particular, in the surface layer of the sea. A

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comparison of all the data obtained by the Baltic countries on the distribution of radioactive cesium in the surface waters of the Baltic Sea in 1986 (Fig. 1-3) shows that the greatest "spots" of contamination took place in the central and eastern parts of the Gulf of Finland and in the southern part of the Gulf of Bothnia. In June 1986 the highest activity concentrations of cesium-137 among the observed in the Baltic Sea waters, over 3000  $Bg/m^3$ , were detected to the north of the central part of the Gulf of Finland and in the southwestern part of the Bothnian Sea. In  $1100 \text{Bg/m}^3$  of the same period near NPP Olkiluoto a value cesium-137 was fixed. Judging from the data of May-June (Fig. 1) in the southwestern part of the Baltic Sea Proper and in the region of the straits there was also a "spot" of increased contamination. The average value of cesium-137 content in the surface water of this region in May was 240 Bq/m<sup>3</sup>, reaching 1089 Bg/m<sup>3</sup> in one of the points. In June the average value of cesium-137 content decreased to 177  $Bq/m^3$ , and in August (Fig. 2) the content of cesium-137 in the surface water of this region was 69 Bq/m3 on the average, in October-November, 74 Bq/m<sup>3</sup> (Figs. 1-3; the region of the straits and the southwestern part of the sea).

In the central and eastern parts of the Gulf of Finland the "spot" of radioactive contamination was steadily traced in June and August (Figs. 1 and 2). On the whole it changed little by the level of cesium-137 content in that period, but the high activity concentration values, as  $3100 \text{ Bg/m}^3$ , were not encountered yet in August. In general, the "spot" moved westwards: in the point of the western part of the Gulf of Finland where in June 97  $Bq/m^3$  of cesium 137 were observed, in August it was 530 Bq/m $^3$  (Figs. 1 and 2). In October-November there were only 2 measurements of cesium-137 in the Gulf of Finland: in the central part (180  $Bq/m^3$ ) and in the Koporie bay (190  $Bq/m^3$ ). These two results may but only partly characterize the situation in the Gulf of Finland, nevertheless they give grounds to assume a considerable decrease of contamination of the surface waters in the Gulf of Finland by October-November, 1986.

Most data for the Gulf of Bothnia were obtained in October-November, 1986. Judging by the fact that even in that period, i.e. half a year after its rise, cesium-137 contamination in the Bothnian Sea, especially in its southern part, reached Ba/m<sup>3</sup> 700-800 (Fig. 3); there was a spacious "spot" of contamination there. The same is testified by the value of cesium-137 content in the littoral water of Forsmark NPP in June. A characteristic feature of the radioactive cesium's behaviour dynamics in the water of the Bothnian Sea near the Forsmark NPP is an increase of its content in June-July, 1986, apparently due to washing the contamination off from the coastal territory, and further on, a decrease of its content | 10 |. In the northwestern part of the Baltic Sea Proper relatively high concentrations of cesium-137 (600-800  $Bq/m^3$ ) were observed in October-November, but absence of the data for the period May-June doesn't enable to trace the way of the contamination's appearance immediately from the atmospheric source due to fallouts or due to spreading southwards of the contaminated water from the Bothnian Sea.

Separate patches of an increased content of radioactive cesium also took place in the central part of the Baltic Sea Proper, up to 500 Bq/m<sup>3</sup> of cesium-137 (Figs. 1 and 2), but those were only individual points and in immediate proximity to them values less by factor of 10 were observed.

Comparison of the results obtained in different periods shows that the processes of redistribution in water masses cause dispersion of highest concentrations of radioactive cesium in the Baltic surface waters of the Sea. In the course of the observations side by side with horizontal movement, an intensive penetration of contamination into the deep layers of the sea was revealed. Depth sections were done in many points of the sea and the content of radioactive cesium was determined in different layers. As one would expect, the rate of the contamination's vertical spread was different in various parts of the sea. The available information should be considered by separate regions of the sea.

In the waters of the Danish straits cesium-137 of Chernobyl origin was observed in the bottom layers already in May, 1986, but its quantities differed greatly relative to the content in the surface layer. If we take cesium-137 content in the surface layer as a unity, the share of cesium-137 in the bottom layer in May was from 0 to  $1.9 \mid 1 \mid$ , in August, from 0.5 to  $2.4 \mid 1 \mid$ , in October-November, from 0.9 to  $1.7 \mid 3 \mid$ . Evidently, it was only in October-November, i.e. half a year after the ingress, that a relatively uniform distribution of cesium-137 in the water mass was reached in that region. For all that a characteristic feature of the vertical distribution in a number of points in that region was higher content of cesium-137 in the bottom layer in comparison with the surface one, which gives evidence of transport of cesium-137 with deep-water flow from neighbouring more contaminated regions of the sea.

In the waters of the southwestern part of the Baltic Sea a uniform distribution of radioactive cesium was observed within the limits of the upper 10-20 m in May, 1986; in some cases, like in the straits, the contamination in the deep-water layers was greater than in the surface ones | 3, 7-9 |.

In the waters of the central part of the Baltic Sea Proper in May-June the content of cesium-137 at a depth of 10-20 m was 0.1-0.2 from the surface layer content taken as a unit; at the same time there were points with equal content of cesium-134 | 3, 7, 8 |. In July in the waters of the eastern part of the Baltic Sea Proper the content of cesium-137 of Chernobyl origin in the bottom layers was about 1/3 of its value in the surface layer | 11 |. Several determinations were carried out in the region of the Gottland deep in May-November, 1986. Table 1 summarizes the available data on the activity concentration of cesium in the Gottland deep waters.

Sampling date	Sampling horizon,	Content, Bq/m <sup>3</sup>		Reference
	m	cesium-134	cesium-137	
May 9	0	33	74	
	4.5	8,6	28	/6/
	100	17	48	
July 23	0	13	41	/11/
	200	6	22	
August 17	0	35	81	/6/
	230	13	49	
October-				
November	1	45,3	113	/7,8/
	50	7,4	27,4	

Table 1. Activity concentration of cesium-134 and cesium-137 in the waters of the Gottland deep of the Baltic Sea in 1986

As seen from the table, the obtained data differ considerably. On the one hand, already on May 9 the content of cesium-134 at a depth of 100 m was half of its content in the surface layer. On the other hand, in October-November at the same depth cesium-134 content was less than 0.06 from that in the surface layer taken as a unit. The content of cesium-134 at a depth of 200-240 m in July-August, according to one data , was 0.3-0.5 in relative units from the surface layer content, and according to the other ones, in October-November it was less 0.06 in the same relative units.

The difference in the results may be explained by many reasons. The sampling in May coincided with the growth of plankton, which was intensely absorbing radioactivity. The dead particles of plankton were sinking lower, causing the penetration of radioactive cesium into deeper layers of water. That was why cesium was found in deep water. At the same time there is a possibility of errors due to methodology. When sampling in deep layers, the sampler was passing through a contaminated surface layer and the contamination was introduced in the sample.

In the Gulf of Finland in June, 1986, in the bottom layer of a point 34 m deep, there was about 1/3 of the surface amount of radioactive cesium. In August in the same region a uniform distribution of contamination at a 30 m layer was already observed. In regions with greater depth there wasn't observed uniform distribution in August either.

In the Bothnian Sea a lowering down of the radioactive cesium's content was appreciable at a depth of 40 m in October-November, 1986. Within the limits of the upper 0-20 m the content of radioactive cesium was uniform. In the Gulf of Bothnia radioactive cesium was distributed practically uniformly by all the layers.

On the whole, in spite of extensive information on the depth distribution of radioactive cesium, the obtained data are often hard to compare. Doubtless is the very fact of an intensive penetration into the deep water layers of the radioactive contamination that has fallen out on the water surface. The rate of the contamination's spread into the deep water layers is different in different parts of the sea. The arising distribution is unstable because there "spots" of radionuclides are of increased contamination that gradually disperse in the sea. Some depth distribution of radioactive cesium need data on the refinement. Nevertheless, the obtained material shows that the radioactive contamination fallen out on the surface of the Baltic Sea, in half a year reached the bottom water layers practically on the whole water body of the sea. In many shallow-water areas it took only 1-2 months, sometimes less. Finding radioactive cesium of Chernobyl origin in bottom sediments is a confirmation of the contamination's penetration throughout the whole water depth of the sea in such a short time.

The results of studying the bottom sediments of the Baltic Sea in 1986 are presented in a number of works | 2, 6, 7, 9, 10 |. In

the northeastern part of the Baltic Sea Proper at a depth of 166 m in mid-June there was noted cesium-134 in the 1st cm of the bottom sediment; however, it wasn't found in the same place in October | 6 |. In August and October cesium-134 was determined in the bottom sediments of the Bothnian Sea and the Gulf of Finland 6 [. In the Grelfswald Bay in July 1986 radioactive cesium from the Chernobyl NPP accident was determined in the upper 3 cm of the bottom sediment [ 7 ]. In June a mixture of cesium-134 and cesium-137 was determined in the bottom sediments at depths of 63-109 m in the southern part of the sea | 9 | . In September in the Baltic Sea Proper in the layer 0-2cm of bottom sediments cesium radionuclides were determined in the following amounts: cesium-134, 7 Bq/kg of dry residue, cesium-137, 14 Bq/kg of dry residue | 10 |. Cesium-137 content was determined in the layer 0-3 cm of the bottom sediments in different parts of the sea 2 , but it isn't clear from the presented data which traction of cesium-137 is related to cesium-134 with the ratio 0.55.

the fact is established of the radioactive cesium Thus, of Chernobyl origin depositing into the bottom sediments of the Baltic Sea. To determine the mechanism of penetration is a research problem. Important information is provided by systems of "traps" used mainly in the vicinity of NPP's | 4, 6, 10 |. In the course of penetration of radioactive cesium from the water mass into the bottom sediments, a certain part is played by the suspended matter and its quantity (Water turbidity). According to reference | 4 |, in the Gulf of Bothnia about 5% of radioactive cesium in water is bound with particles , while in the Bothnian Sea, only 1%. Analogous studies in the water of the Koporie bay of the Gulf of Finland showed that 2% of radioactive cesium is bound with suspended matter | 10 |. Evidently, observations of the process of radioactive cesium's sedimentation on the bottom under the conditions of the arisen increased contamination of the Baltic Sea become the most essential in the complex of further studies.

Cesium-137 that drifted to the Baltic Sea region as a result of air transport from the site of the Chernobyl accident, added to

the global one and like the latter entered all the links of the ecological chain. In a short time cesium-137 in combination with cesium-134 was registered practically in all the biological objects of the Baltic Sea | 1, 6, 9, 10 |. The arising radioactive contamination was reacted to by the plankton and algae, in particular, Fucus.

Characteristic for these fixators of contamination is their fast response to an increase of water contamination: a sharp growth of radionuclide content in biomass is observed, and then, a decrease in line with the decrease of the water medium contamination. In fish taken from the Baltic Sea, radionuclides were also found, in particular, cesium-134 and cesium-137. The content of radioactive cesium in fish is largely determined by the accumulation effect. There is also a species dependence.

The radiation situation in the Baltic Sea is substantially influenced by river discharges. Under the conditions of an increased contamination of the Baltic Sea, the contamination level of the river waters acquires a particular significance. The main bulk of the river discharge takes place in the eastern part of the sea and mainly in the gulfs. In reference | 11 | there are the data on the radioactive cesium content in the waters of the largest rivers discharging into the Baltic Sea from the territory of the USSR. The main part of the information concern July, 1986. In that period the activity concentration in the water of the Soviet Baltic rivers varied within the limits 16-76 Bq/m<sup>3</sup>. The data on cesium-137 content in the waters of the Neva and the Narva rivers were obtained in May, the cesium-137 activity 240-290  $Bq/m^3$  in that period. A repeat concentration being in December, 1986, showed a decrease of the determination activity concentration of cesium-137 in the water of the Neva river down to 10 Bq/m<sup>3</sup>.

In Finland 5 large rivers discharging into the Baltic Sea are under constant monitoring. In 1986 the sampling was done in May, August and October. Greatest values of the activity concentration of cesium-137 and cesium-134 took place in May, especially in the waters of two rivers: the Kymijoki river that discharges into the Gulf of Finland, and the Kokemäenjoki river discharging into the Bothnian Sea. The activity concentration of cesium-137 in those rivers reached 1900-2500 Bq/m<sup>3</sup> in May, then decreased to 510-930 Bq/m<sup>3</sup> in October<sup>\*)</sup>.

As it has been said, the main attention in studying the radiation situation in the Baltic Sea in 1986 was paid to radioactive cesium. Nevertheless, in some cases other nuclides were also determined. It was found that the fallout of strontium-90 into the Baltic Sea region was insignificant, therefore the increase of its content in the waters of the sea didn't exceed 10-30% from the global level. At the same time there is information that there were in the atmospheric fallout individual particles with high contents of strontium-90<sup>\*\*)</sup>. The determination of plutonium-239,240 carried out in November, 1986, practically on the whole water body of the sea, showed the activity concentration of plutonium-239, 240 to be from 0.003 to 0.036  $Bq/m^3$ . The upper limit is higher than the one of the previous years \*\*\*), but only points out of 15 in 3 that the volume activity of plutonium-239,240 was over 0.02  $Bq/m^3$ . The determination of other transuranic elements was also carried out.

No increase was noted of tritium content in the Baltic Sea waters in 1986. Although iodine-131, ruthenium-103 and 106, tellurium-132 did occur, the increased radiation background created by them wasn't a longtime one because of relatively short half-lives of the said radionuclides.

\*) Radioactivity of surface water in Finland after the Chernobyl accident in 1986. STUK-A60. June 1987. Helsinki, Finland \*\*) Radioactivity of wet and dry deposition and soil in Finland after the Chernobyl accident in 1986. STUK-A57, June 1987 \*\*\*) Study of radioactive materials in the Baltic Sea. IAEA-TECDOC-362, Vienna, 1986

Thus, the change of the radiation situation in the Baltic Sea was practically completely determined by the fallout of radioactive cesium, the atmospheric fallout of which couldn't be measured immediately on the surface of the sea. The determination of radioactive cesium in the Baltic Sea waters indicated only the consequence of the atmospheric fallout. That is why the attempt to reconstruct the picture of the radioactive fallout by the radioactive cesium distribution in the water masses, undertaken in work | 4 |, is undoubtedly noteworthy. The calculation made use of the results of numerous determinations of radioactive cesium in the water masses of the sea, the activity ratio of characterizing cesium-134/cesium-137 the contamination's Chernobyl origin, the hydrological characteristics, the data on the temperature and salinity of water. As the result a picture was obtained of the distribution of the cesium-137 fallout on the surface of the Baltic Sea in the period of the end of April-May, 1986. It is presented in Fig. 4. This picture has been completed with the calculated values of cesium-137 fallout in the eastern part of the Baltic Sea Proper and in the Gulf of Finland, obtained on the basis of reference | 11 |.

The obtained evaluations should be considered as preliminary ones; they may and must be specified, completed and possibly, revised. The data used for calculation were obtained at different times, the observation period covering half a year since the moment of the contamination's ingress. In this time, as has been said above, spatial changes took place. The radioactive "spots" arisen as the result of atmospheric fallouts partly washed out and displaced. But on the whole the obtained picture seems rational. For comparison in Fig. 4 there are given the data on the radioactive cesium fallout on the territory of the Baltic countries, taken from works | 1, 9 | \*-\*\*\*\*).

In conclusion it should be noted that in the period under consideration the Baltic Sea States obtained a great deal of valuable information enabling to characterize the radiation situation that arose in the Baltic Sea in 1986.

\*) Environmental gamma radiation measurements in Finland and the influence of the meteorological conditions after the Chernobyl accident in 1986. STUK-A65, June 1987, Helsinki, Finland

\*\*) I. Vintersved et al. IEEE Transactions on Nuclear Science. Vol. NS-34, N l, .p 590-594, February 1987. (Early measurements of the Chernobyl fallout in Sweden)

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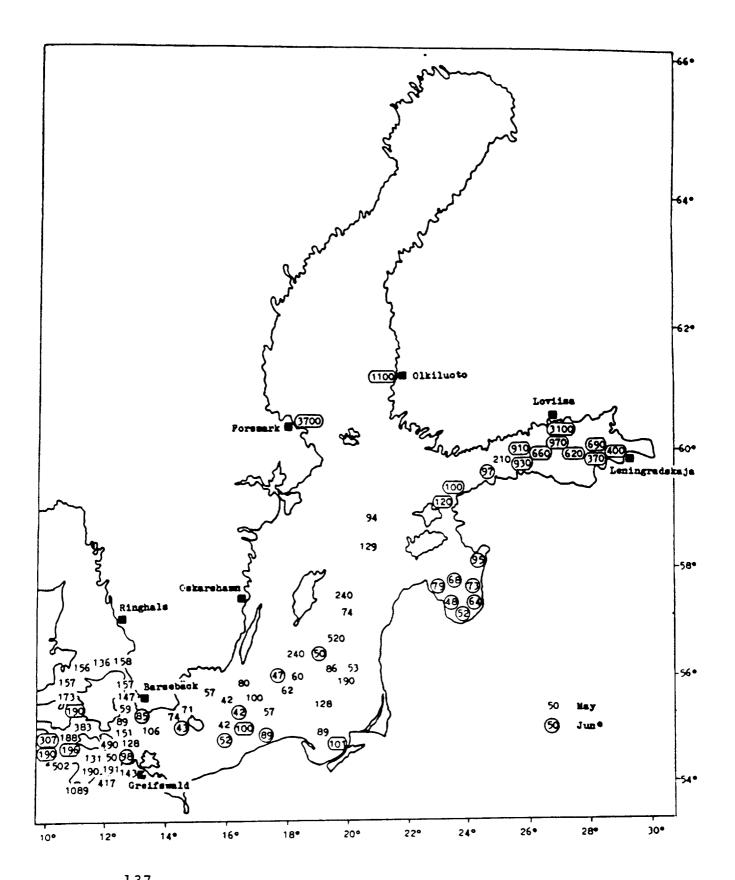


Fig. 1. <sup>137</sup> Cs activity concentration, Bq/m<sup>3</sup>, in the surface water of the Baltic Sea in May-June 1986.

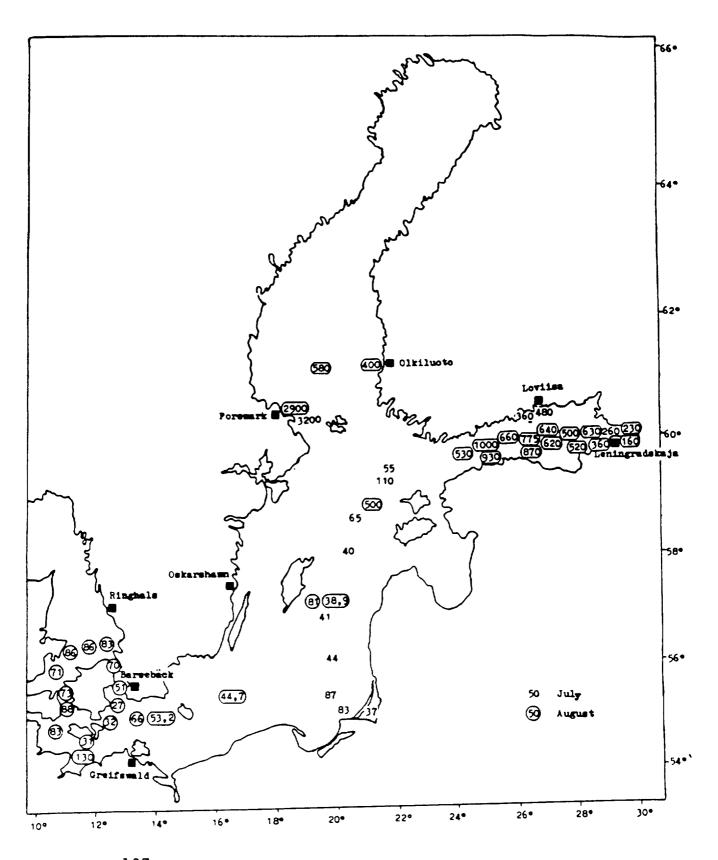


Fig. 2. 137 Cs activity concentration, Bq/m³, in the surface water of the Baltic Sea in July-August 1986.

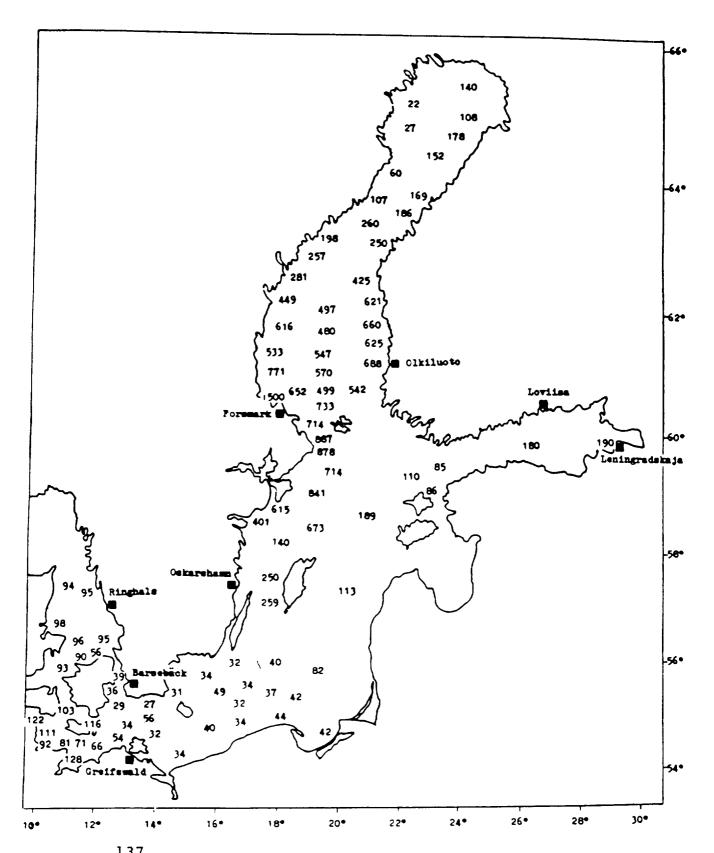


Fig. 3. 137 Cs activity concentration, Bq/m<sup>3</sup>, in the surface water of the Baltic Sea in October-November 1986.

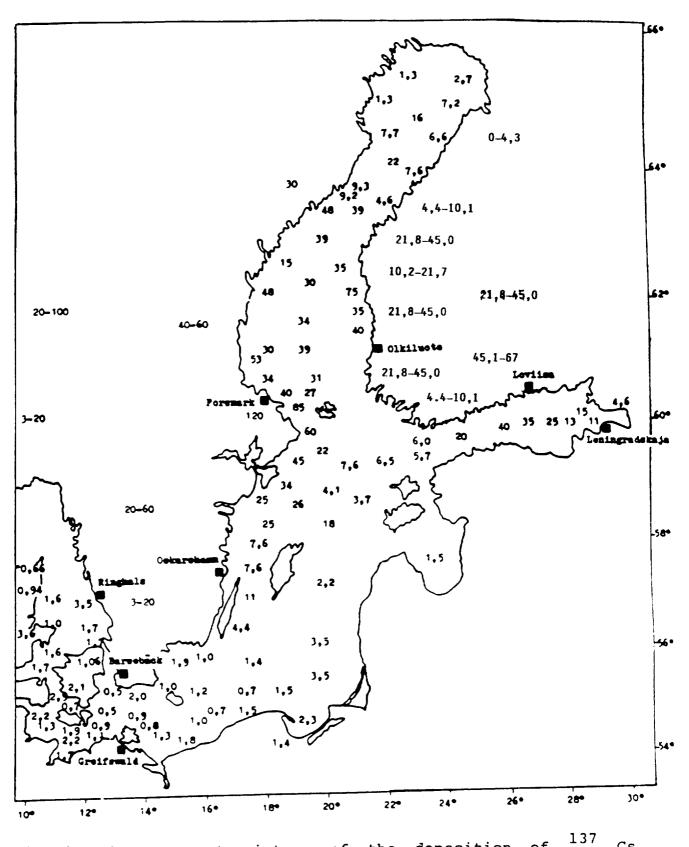


Fig. 4. The apparent picture of the deposition of  $^{137}$  Cs, kBq/m<sup>2</sup>, at the surface of the Baltic Sea.

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#### THE DISTRIBUTION OF THE CHERNOBYL FALLOUT OVER THE BALTIC SEA AND ITS CHANGE DURING 1987 AND 1988 IN SEAWATER

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#### Introduction

Artificial radioactivity in the North Sea and the Baltic Sea has been studied for many years by the German Hydrographic Institute. The main sources of radioactivity in these two seas have been global fallout by nuclear weapon tests mainly in the sixties, and liquid discharges into coastal waters from the European reprocessing plants, primarily at Sellafield (Irish Sea) and La Hague (English Channel). This contamination has been transported into the Baltic Sea from time to time through the Danish Straits due to the prevailing current pattern of the North Sea. From around 1975 up to 1985 the activity concentration of <sup>137</sup>Cs in the Baltic Sea correlated with the salinity according to the changing discharges from Sellafield (IAEA, 1984). The <sup>137</sup>Cs activity concentration decreased from the Kattegatt to the Bay of Bothnia according to decreasing salinity. <sup>90</sup>Sr did not correlate in this way with the salinity.

The serious accident at the nuclear power reactor in Chernobyl in April 1986 changed this situation competely. Large quantities of radioactivity were released into the environment and deposited over all areas of Europe. Part of these releases also reached the marine environment of the North and Baltic Seas. Due to its special geographical nature as a semi-enclosed shelf sea, the fallout into the Baltic Sea will remain for the years to come, whereas the initial Chernobyl input into the North Sea was transported from the North Sea area and diluted along the Norwegian coast (NIES, 1989).

#### Chernobyl Fallout over the Baltic Sea

The accident at Chernobyl released a great number of artificial nuclides into the environment, such as  $^{137}Cs$ ,  $^{134}Cs$ ,  $^{90}Sr$ ,  $^{95}Zr$ ,  $^{95}Nb$ ,  $^{99}Mo$ ,  $^{103}Ru$ ,  $^{106}Ru$ ,  $^{110m}Ag$ ,  $^{129m}Te$ ,  $^{131}I$ ,  $^{132}Te$ ,  $^{140}Ba$ ,  $^{141}Ce$ ,  $^{144}Ce$ ,  $^{239}Np$ , and  $^{242}Cm$ . Most of these nuclides have short half-lifes of only a few days and therefore have had no impact on the marine environment. However, the most important long-lived nuclides of this fallout were  $^{137}Cs$  and  $^{134}Cs$ , which provide us with the possibility to identify the Chernobyl contribution by its characteristic fallout activity ratio  $^{134}Cs/^{137}Cs$  of about 0.5 in April 1986. The two years half-life product  $^{134}Cs$  was not present in the Baltic Sea before the accident at Chernobyl.

The atmospheric input of the Chernobyl fallout led to a patchy distribution in the water of the Baltic Sea, which was the result of the different intensity of the atmospheric precipitation. First samples of seawater from the Baltic Sea were taken shortly after the the accident in the Baltic Proper between 29 April to 14 May 1986. The highest values of <sup>137</sup>Cs between 100 and 240

Bq m<sup>-3</sup> could be measured initially after the input at the sea surface, before vertical mixing of the contamination started dilution (STUK, 1987a; DHI, 1987). A few days later the surface concentration in the southern Baltic resulted in values between 40 and 100 Bq m<sup>-3</sup>, while in the Gulf of Kiel and Lübeck Bight values between 240 and 1090 Bq m<sup>-3</sup> <sup>137</sup>Cs were determined, which is indicative for higher areal deposition in this area due to washout of the atmosphere by rainfall.

An investigation for the same area was repeated from 4 to 11 June. Meanwhile the values had decreased to activity concentrations of about 40 to 50 Bq m<sup>-3</sup> in the Baltic Proper east of Bornholm. In the Gulf of Kiel values between 160 and 200 Bq m<sup>-3</sup> for <sup>137</sup>Cs have been measured (DHI, 1987). In fig. 1 the temporal development of the activity concentration of <sup>137</sup>Cs and <sup>90</sup>Sr is presented (NIES and WEDEKIND, 1987).

The strong halocline in the southern part of the Baltic Sea limited the complete vertical mixing of the Chernobyl contamination. Beneath the halocline, which lies in a depth of 40 to 60 m, no Chernobyl derived nuclides have been ascertained. However, a sample of water close to the sediment in 76 m depth showed a significant contribution of <sup>134</sup>Cs, which is a result of the rapid particulate transport through the water column (KEMPE AND NIES, 1987).

A survey of the entire Baltic Sea was carried out in international collaboration in October 1986 with the German RV GAUSS. Water samples were collected at 94 positions at different water depths. The samples were analysed for 137Cs, 134Cs, and <sup>90</sup>Sr. A selected number of samples were also analysed for Plutonium, Americium and Curium isotopes. However, the Chernobyl impact can be recognized unequivocally by the 134Cs activity concentration. Figs. 2 and 3 represent the horizontal and vertical distribution of 137Cs in seawater during October/November 1986 in different areas of the open Baltic Sea. The concentration of the isotope about half the values of those described in figs. 2 and <sup>134</sup>Cs is 3 according to the fairly constant activity ratio of these two isotopes in the Chernobyl fallout. The investigation with RV GAUSS represents the values from the open sea. In coastal waters of Sweden and Finland higher levels were measured for longer periods due to the limited mixing in these shallow waters (STUK, 1987a).

It is obvious that the levels of contamination are quite different in different regions. The highest values were found in the southern part of the Bothnian Sea. The surface water west of the Aaland Isles had an activity concentration of about 900 Bq m<sup>-3</sup> for <sup>137</sup>Cs and 450 Bq m<sup>-3</sup> for <sup>134</sup>Cs. The values decreased northward in the Gulf of Bothnia and southward to the Baltic Proper from that position. The surface concentration in the Bothnian Sea lay between 500 and 800 Bq m<sup>-3</sup>, in the western part of the Bothnian Bay from 100 to 180 Bq m<sup>-3</sup> and in the eastern part from 20 to 40 Bq m<sup>-3</sup>. In southward direction the activity concentration decreased to 250 Bq m<sup>-3</sup>  $^{137}$ Cs in the area of Öland Bq m<sup>-3</sup> and in the eastern and Gotland. The samples of surface water from the southern Baltic Proper between the Danish Isles Moen, Bornholm and Bay of Bq m⁻³ <sup>137</sup>Cs and 10 to 20 Gdansk contained around 30 to 40 Bg m<sup>-3</sup> <sup>134</sup>Cs. The distribution of <sup>137</sup>Cs and <sup>134</sup>Cs in the surface water of the entire Baltic Sea is presented in figs. 4 and fig.

5, respectively. These figures are computed by using the positions of the mesurements and interpolating the concentrations of the areas between these data points.

The structure of the Chernobyl fallout distribution in the Baltic Proper is not very well resolved, because the range of the concentration is too large. However, the structure of the high contaminated area in the Bothnian Sea reveals some very interesting features. The picture does not represent the initial fallout distribution in May 1986. It shows a distribution in which both the small range "spottiness" half a year after the equilibrated and contaminated water input has been has been transported in the different the subregions according to 3 to 5 prevailing currents. The highest values in figures coincide well with the relative levels measured on land in Sweden (Sveriges Geologiska, 1986; SMHI, 1986) and Finland (STUK, 1987b). Higher levels are still to be found in the Gävle up to 120 kBq m<sup>-2</sup> district, which received areal deposition of (Grimås et al., 1986). In the eastern part of the Bothnian Sea a tongue-shaped pattern into northward direction can be identified in figs. 4 and 5. This obviously reflects the fresh-water run-off from the Finnish river Kokemäenjoki, which drains the high contaminated area in Central Finland. The river itself carried also a higher Cesium concentration than the other Finnish rivers (STUK, 1987c). This pattern follows the general water circulation of the Bothnian Sea.

In a southward direction contaminated water was already transported southward from the Aaland Sea into the Baltic Proper. The increasing concentration field in eastern direction of the Gulf of Finland is not necessarily realistic, because the number of data points do not resolve this region sufficiently.

The contribution of the nuclide 90 Sr in the Chernobyl fallout was rather low. If one uses the 90 Sr-values from previous years, one obtains a Chernobyl activity ratio 90 Sr/137Cs of 0.010 to 0.013 in the water samples of the northern Baltic Sea. Fig. 6 shows the distribution of 90 Sr in the surface water six months after the accident. The distribution is very similar to those in figs. 4 and 5. The same phenomena both of water movement southward from the Aaland Sea and of river run-off in the eastern part of the Bothnian Sea can be recognized in much smaller concentration as already demonstrated for the isotopes 134Cs and 137Cs. The lowest values of 90 Sr are determined also in the northwestern positions of the Bay of Bothnia.

Regarding the depth penetration of the Chernobyl fallout, one can establish that the fallout has completely reached the bottom water in the Bay of Bothnia (fig. 3). Even higher concentrations than in surface water can be detected in samples close to the sediment. This is due to the weak density gradient between surface and bottom water. However, the concentrations of  $^{137}$ Cs and  $^{134}$ Cs in the Bothnian Sea decrease with increasing depth. Significant amounts of  $^{134}$ Cs could be measured below the permanent halocline.

In the Baltic Proper the strong halocline impeded the penetration of Chernobyl derived Cesium into layers below the halocline (fig. 2), which lies at a depth of 40 to 60 m, whereas the fallout has reached the bottom in the shallow western Baltic. In August 1987 the investigation of the entire Baltic Sea was repeated with SWRV ATAIR, in order to trace the changes of the contamination. As can be seen in figs. 7 to 10 the activity concentrations in the Aaland Sea and the Bothnian Sea dropped within the nine months, but the highest concentrations were still to be found in the same areas as in 1986. The maximum surface concentration of  $^{137}$ Cs and  $^{134}$ Cs in the Aaland Sea was about 400 Bq m<sup>-3</sup> and 140 Bq m<sup>-3</sup>, respectively. Due to surface water transport from the northern Baltic Proper into the southern and eastern Baltic Proper, the values increased in the areas of Bornholm and the Gdansk Bay to about 100 Bq m<sup>-3</sup> for  $^{137}$ Cs. Now the fallout was also detectable below the halocline.

The concentration in bottom waters of the Bay of Bothnia were in general higher than the surface water, which reveals the penetration of contaminated water from the Bothnian Sea into the deep layers of the Bay of Bothnia. The cyclonic movement of water in the Bay of Bothnia led furthermore to elevated concentration in the western part of this sea area.

The minimum concentration in the western Baltic was detected in April 1987 (fig. 1). Since then slightly increasing concentrations have been ascertained. The results of the investigation in August 1987 demonstrate the general movement of the contaminated water, following the general circulation of the Baltic Sea.

The results obtained from samples in 1988 showed no big difference to those from 1987. A few samples could be analysed from a cruise of RV GAUSS in May/June 1988 through the entire Baltic Sea. These data are given in Table 1. The highest activity concentration was determined in the southern Bothnian Sea.

Position	n	<sup>137</sup> Cs Bq m <sup>-3</sup>	134Cs Bq m <sup>-3</sup>
$ \begin{bmatrix} 55 \circ 00 & N & 14 \\ 54 \circ 52 & N & 19 \\ 57 \circ 07 & 17 \\ 60 \circ 05 & 19 \\ 61 \circ 05 & 19 \\ 62 \circ 05 & 19 \\ 62 \circ 05 & 19 \\ 62 \circ 05 & 24 \\ 59 \circ 02 & X & 21 \\ 59 \circ 02 & X & 21 \\ \end{bmatrix} $	° 13'E Kattegatt °05'E west of Bornholm °08'E Gdansk Bay °10'E west of Gotland °09'E Aaland Sea °35'E Bothnian Sea °59'E Bothnian Sea °21'E Bay of Bothnia °50'E Gulf of Finland °55'E Baltic Proper °15'E east of Gotland	$78\\113\\92\\335\\32162\\129562\\11537\\1537\\115$	232 47 897 34 406 73 406
Table 1 $^{137}Cs$ and $^{136}20$ May to 2	4Cs (Bq m <sup>-3</sup> ) during a c June 1988	ruise with	RV Gauss,

The analysis of water samples of a cruise with SWRV ATAIR in April 1988 to the western Baltic revealed the slow increase of water activity in this part of the Baltic and the Kattegatt (Fig. 11). This is due to the outflow of low salinity surface water, which carries a higher activity concentration from the northern Baltic Proper. The increasing mixing of Chernobyl derived Cesium below the halocline can be observed in the samples taken close to the bottom in the area of the Island Bornholm.

The temporal development of the activity concentration of <sup>137</sup>Cs and <sup>90</sup>Sr during 1988 and March 1989 at the three positions Schleimündung, Fehmarn Belt and Lübeck Bight in the western Baltic is presented in fig. 12. The development of the concentration is not unequivocal. The lower values in December 1988 at these three positions are connected to higher salinity in the samples, whereas the position "Schleimündung" seems to be the most sensitive one for salinity changes.

## Conclusion

The investigations initiated after the accident of the nuclear power reactor at Chernobyl revealed a considerable increase of artificial radioactivity in the Baltic Sea. The fallout was very unevenly distributed. The highest deposition occurred in the southern part of the Bothnian Sea.

The most important nuclides, which can be determined for a longer period, are  $^{137}$ Cs and  $^{134}$ Cs, which were released from Chernobyl in an activity ratio  $^{134}$ Cs/ $^{137}$ Cs of about 0.5. By means of this ratio the Chernobyl derived Cesium can be identified, because  $^{134}$ Cs was not detectable in the Baltic Sea before the accident. The contribution of the nuclide  $^{90}$ Sr in the fallout was rather low, but measurable.

Since the time of deposition the study of the highly soluble Cesium isotopes in seawater gave the opportunity to comprehend the water movement within the different regions of the Baltic Sea. Additionally, the vertical mixing of pollutants through the permanent halocline could be investigated, because the input took place from the air. The change of the concentration of Chernobyl derived Cesiumisotopes indicate a water mass transport as is presented in fig. 13. This figure does not present quantitative residual currents, but it describes qualitatively the water movement, deduced from the data obtained since 1986. This figure is comprehensively consistent with oceanographic conceptions and models of the water transport in the Baltic Sea (e.g. Kielmann, 1984). Small-scale eddies can, of course, not be 1981; SMHI, resolved by these considerations. The data of Radiocesium obtained by different countries since 1986 could provide an exellent basis for validating numeric hydrodynamic models of the Baltic Sea.

Radiocesium does not behave completely conservatively in seawater, which implies some adsorption to fine grain sediments in the Baltic Sea. This will reduce the inventory within the water column.

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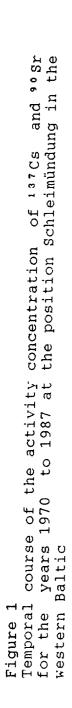
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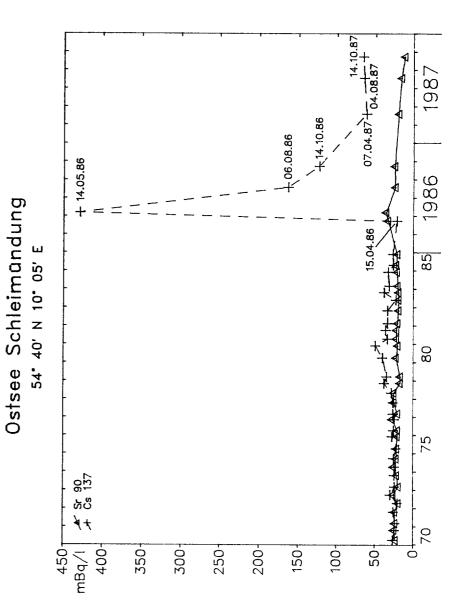
Figures

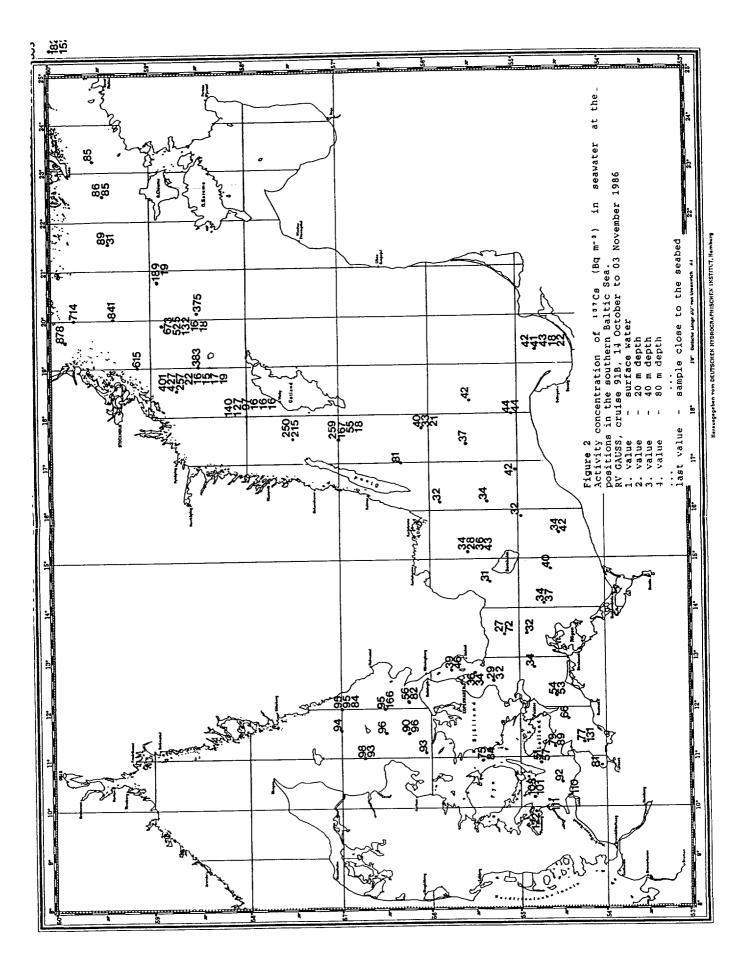
H. Nies The Distribution of the Chernobyl Fallout over the Baltic Sea and its change during 1987 and 1988 in Seawater Figure 1 Temporal course of the activity concentration of 137Cs and 90 Sr for the years 1970 to 1987 at the position Schleimündung in the western Baltic Figure 2 Activity concentration of 137Cs (Bq m<sup>-3</sup>) in seawater at the positions in the southern Baltic Sea. RV GAUSS, cruise 91B, 14 October to 03 November 1986 1. value surface water --2. value \_ 20 m depth 3. value -40 m depth 4. value 80 m depth last value - sample close to the seabed Figure 3 Activity concentration of 137Cs (Bq m<sup>-3</sup>) in seawater at the positions in the Gulf of Bothnia. RV GAUSS, cruise 91B, 14 October to 03 November 1986 1. value surface water ----2. value ---20 m depth 3. value ---40 m depth 4. value 80 m depth --last value - sample close to the seabed Figure 4 Activity concentration of 137Cs (Bq m-3) in surface seawater of the entire Baltic Sea. RV GAUSS, cruise 91B, 14 October to 03 November 1986 Figure 5 Activity concentration of 134Cs (Bq m-3) in surface seawater of the entire Baltic Sea. RV GAUSS, cruise 91B, 14 October to 03 November 1986 Figure 6 Activity concentration of ""Sr (Bq m-") in surface seawater of the entire Baltic Sea. RV GAUSS, cruise 91B, 14 October to 03 November 1986 Figure 7 Activity concentration of 137Cs (Bq m<sup>-3</sup>) in seawater at the positions in the southern Baltic Sea. SWRV ATAIR, cruise 2, 4 August to 1 September 1987 1. value surface water ----2. value ----20 m depth 3. value 40 m depth 80 m depth 4. value ----. . . . . . last value sample close to the seabed

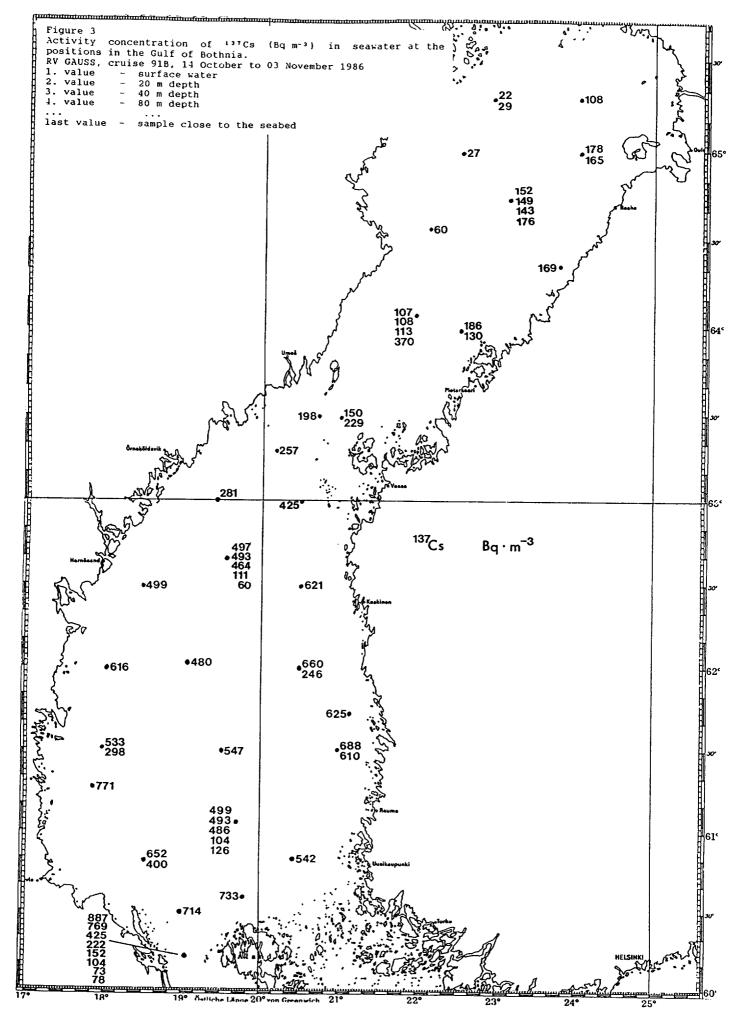
Figure 8 Activity concentration of <sup>137</sup>Cs (Bq m<sup>-3</sup>) in seawater at the positions in the Gulf of Bothnia. SWRV ATAIR, cruise 2, 4 August to 1 September 1987 - surface water 1. value - 20 m depth 2. value - 40 m depth 3. value - 80 m depth 4. value . . . . . . last value - sample close to the seabed Figure 9 Activity concentration of <sup>137</sup>Cs (Bq m<sup>-3</sup>) in surface seawater of the entire Baltic Sea. SWRV ATAIR, cruise 2, 4 August to 1 September 1987 Figure 10 Activity concentration of 134Cs (Bq m<sup>-3</sup>) in surface seawater of the entire Baltic Sea. SWRV ATAIR, cruise 2, 4 August to 1 September 1987 Figure 11 Activity concentration of 137Cs (Bq m<sup>-3</sup>) in seawater of the western and southern Baltic Sea. SWRV ATAIR, cruise 4, 5 April to 11 April 1988 1. value - surface water last value - sample close to the seabed Figure 12 Temporal development of the activity concentration of 137Cs and 90 Sr (Bq m<sup>-3</sup>) at three positions of the western Baltic in 1988. Figure 13 Qualitative water mass transport in the Baltic Sea as deduced

from radioactivity measurements since 1986.









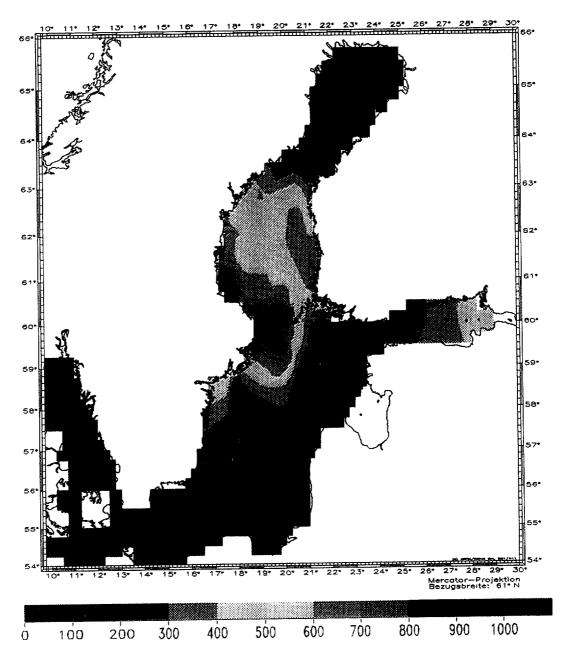
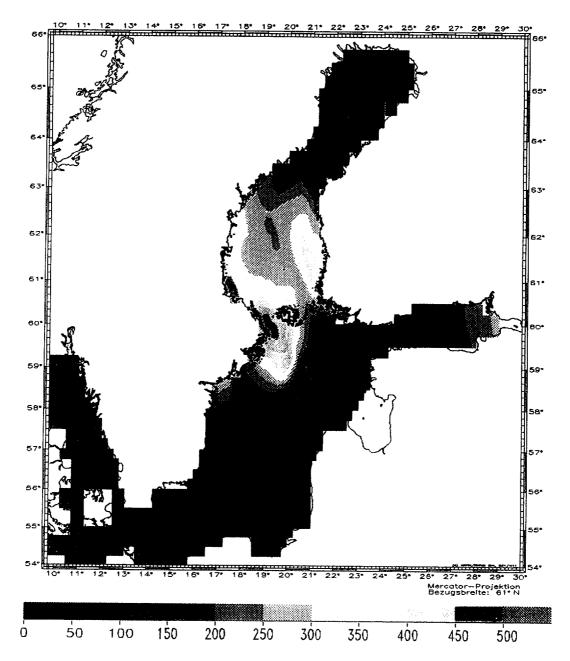


Figure 4

Activity concentration of <sup>137</sup>Cs (Bq m<sup>-3</sup>) in surface seawater of the entire Baltic Sea. RV GAUSS, cruise 91B, 14 October to 03 November 1986



## Figure 5

Activity concentration of <sup>134</sup>Cs (Bq m<sup>-3</sup>) in surface seawater of the entire Baltic Sea. RV GAUSS, cruise 91B, 14 October to 03 November 1986

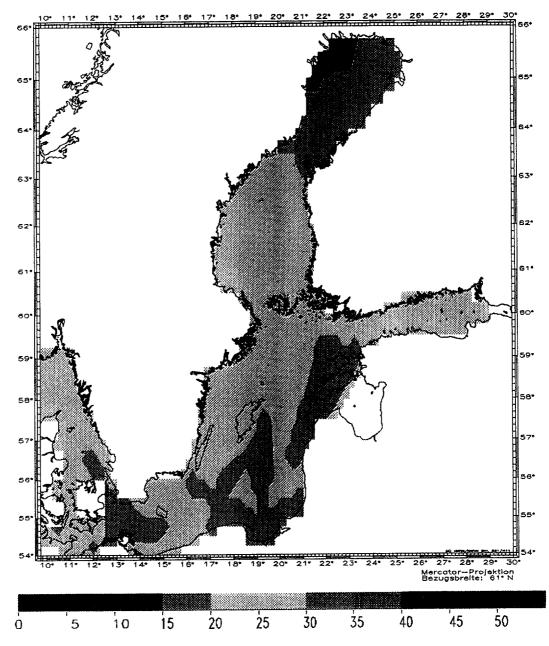
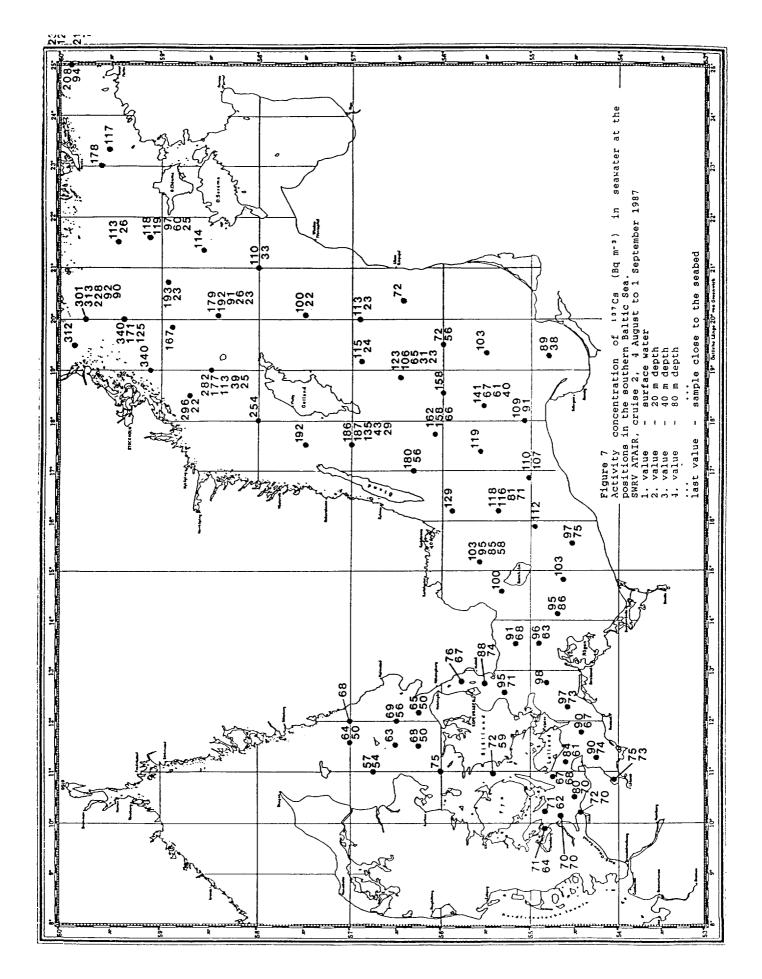
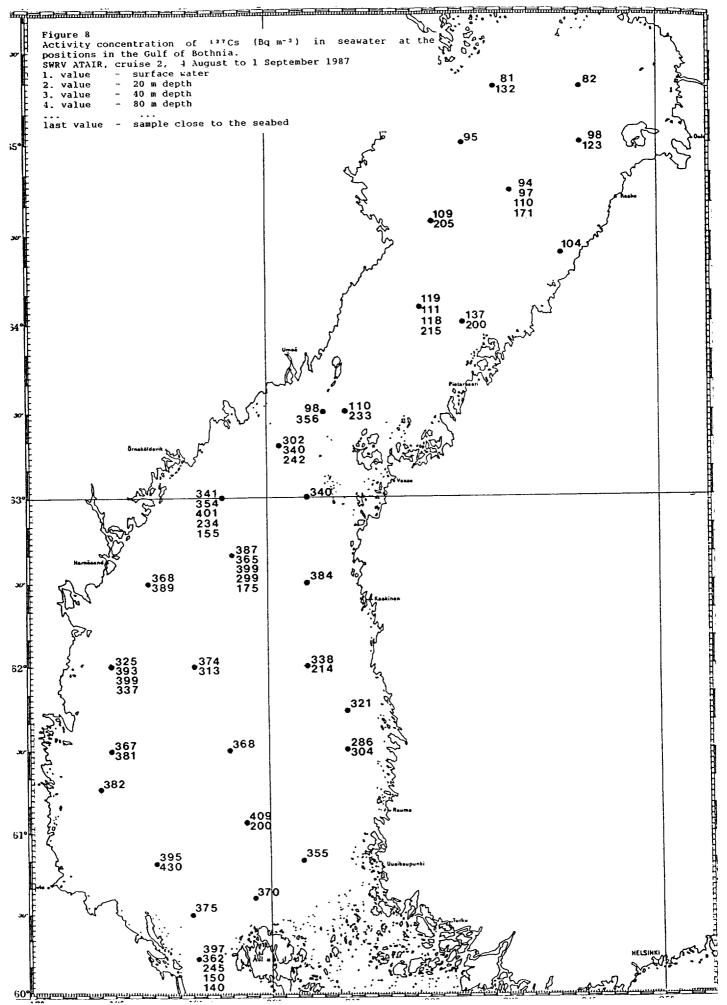


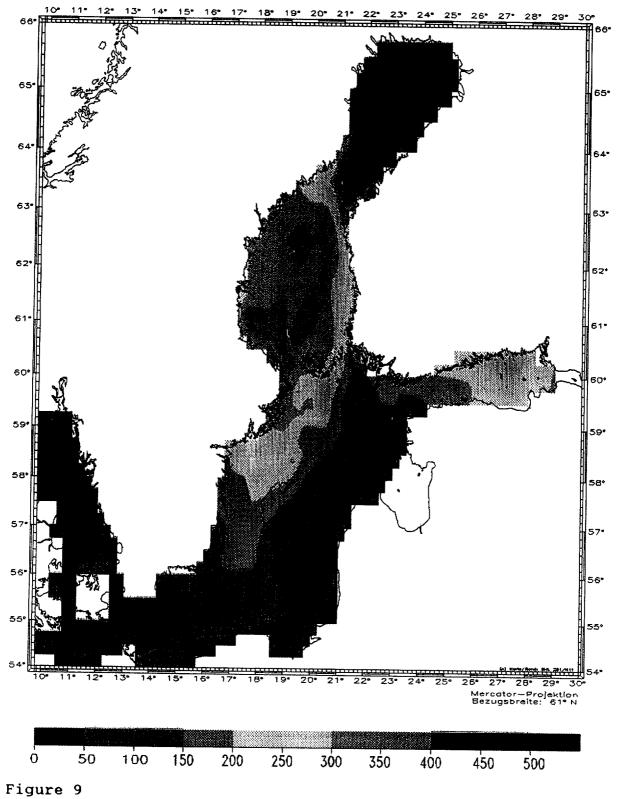
Figure 6

Activity concentration of "Sr (Bq m-3) in surface seawater of the entire Baltic Sea.

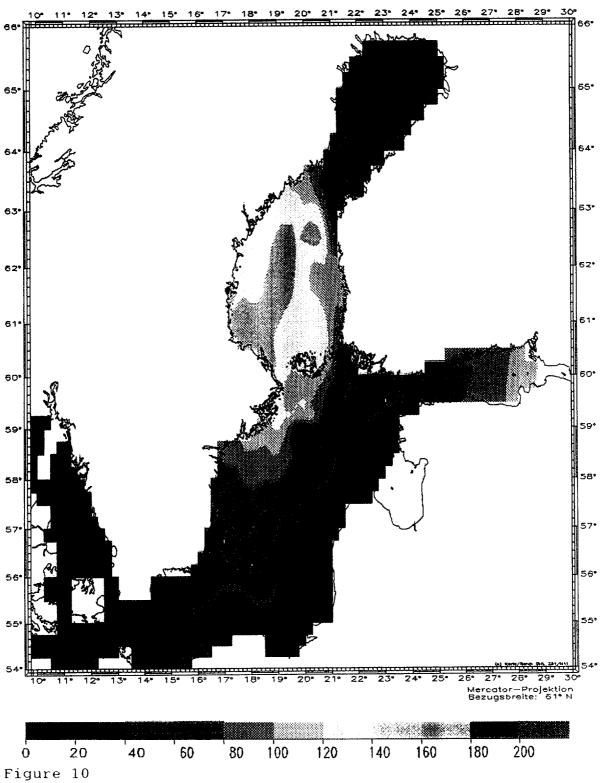
RV GAUSS, cruise 91B, 14 October to 03 November 1986





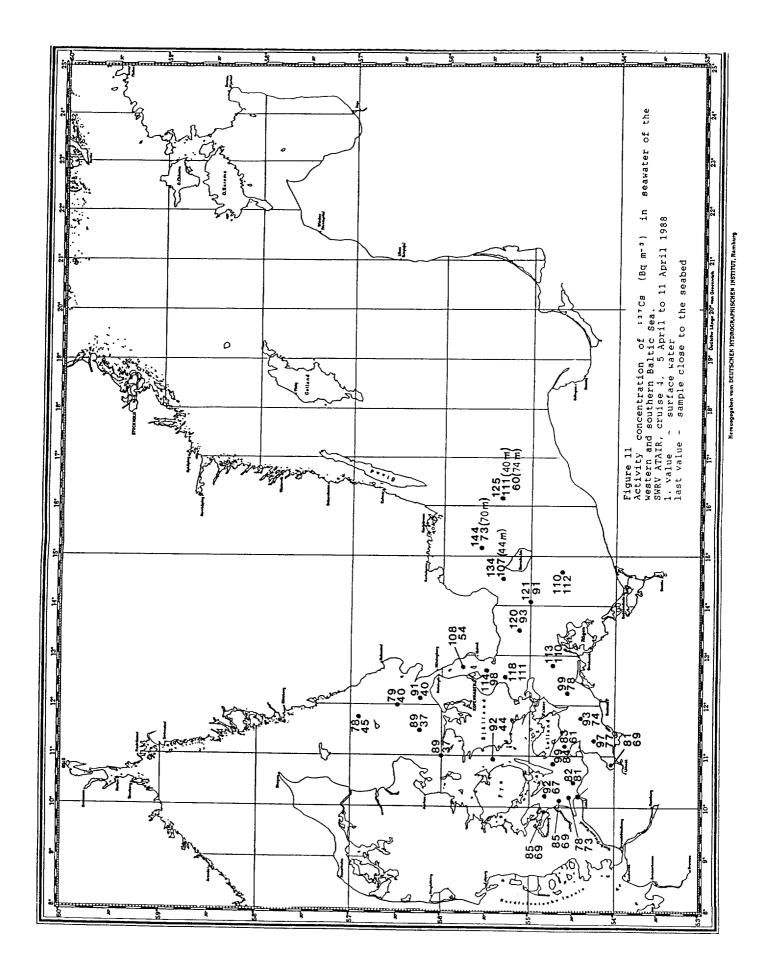


Activity concentration of <sup>137</sup>Cs (Bq m<sup>-3</sup>) in surface seawater of the entire Baltic Sea. SWRV ATAIR, cruise 2, 4 August to 1 September 1987



Activity concentration of 134Cs (Bq m<sup>-3</sup>) in surface seawater of the entire Baltic Sea.

SWRV ATAIR, cruise 2, 4 August to 1 September 1987



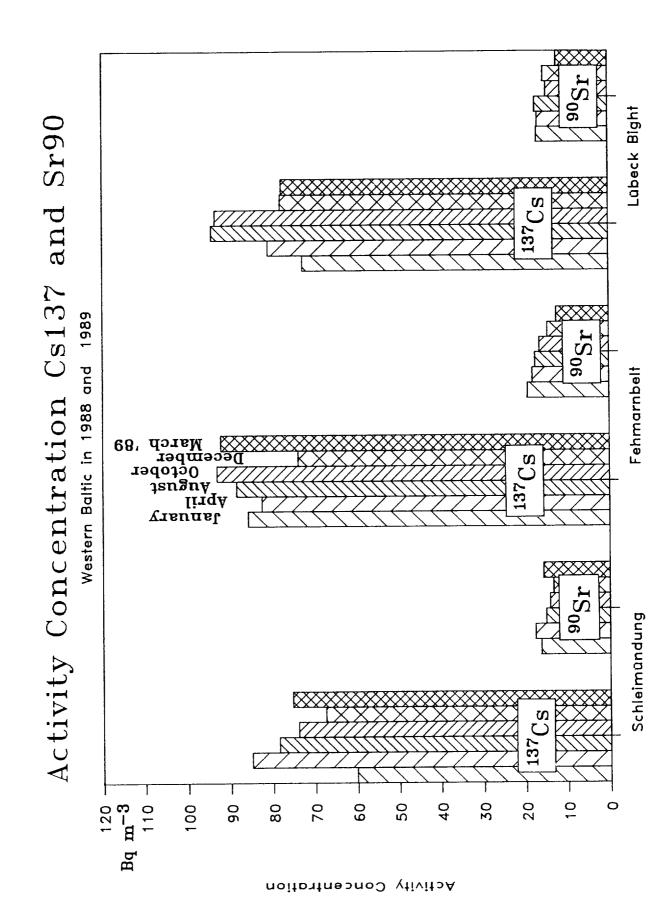
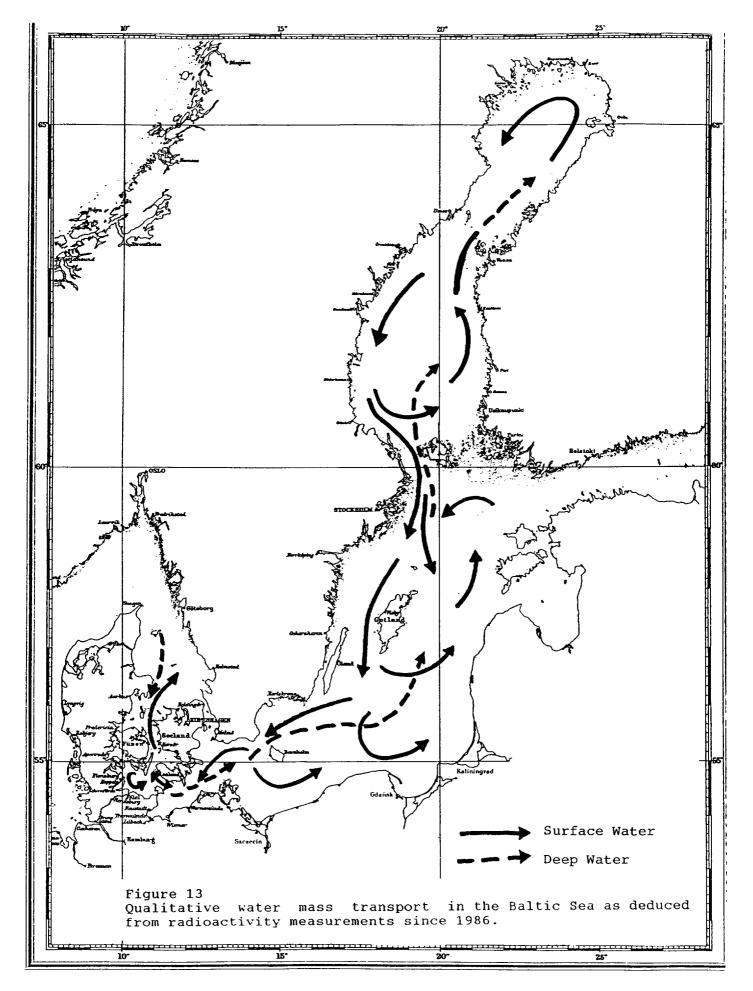


Figure 12 Temporal development of the activity concentration of  $^{137}$ Cs and  $^{90}$ Sr (Bq m<sup>-3</sup>) at three positions of the western Baltic in 1988.



THE RESULTS OF DETERMINATION OF 90Sr, 134Cs AND 137CS IN THE WATER OF THE BALTIC SEA IN 1988.

Gritchenko Z.G., Ivanova L.M., Orlova T.E., Tishkova N.A., Toporkov V.P., Tochilov I.I.

V.G.Khlopin Radium Institute, Leningrad, USSR.

Sample index	Coordina tes sampling	tes of the ing site	Sampling date	Overall water	Sampling depth.	Salinity,	90 <sub>Sr</sub>
	Lat. N	Long. F	_	depth, m		°/°	Bq/m <sup>3</sup>
		Baltic 1	Sea Proper				
BY-28	59°021	21°05"	12.07	105	0	7,42	20,0+3,5
BY-28	\$9°02*	21°051	12.07	105	100	8,44	17,0+3,0
USSR-6	58°00°	20°451	12.07	96	0	7,98	18,0+3,0
USSR-6	58°00°	200451	12.07	96	06	9,20	16,0+3,0
BY-15	57°201	20°03	13 <b>.</b> 07	230	0	7,74	20,0+3,5
BY-15	57°201	20°03	13.07	230	200	11,71	19,0+3,0
USSR-46	56°06¶	19014.	17.07	120	0	8,26	19,0+3,0
USSR-46	56°06°	19014	17.07	120	100	10,22	21,0+3,5
USSR-50	55°30'	18°53'	18.07	85	0	8,40	
USSR-50	55°30°	18°531	18.07	85	80	8,62	20,0+3,5
USSR-51	55°00'	14°001	26.07	45	0	8,46	20,0+3,5
USSR-51	55°00°	14°00'	26.07	45	40	10,53	18,0+3,0
USSR-52	54°43'	120471	26 <b>.07</b>	22	0	9,43	17,0±3,0
USSR-52	54°43'	120471	26.07	22	20	18,72	18,0+3,0
USSR-53	54°281	120131	26.07	19	0	11,25	21,0+3,5
USSR-53	540281	120131	26.07	01	17	۲ ۵0	00 O T 2

Sample index	Coordina tes sampling	tes of the ing site	Sampling date	Overall water	Sampling depth,	Salinity,	90 <sub>Sr</sub>
	Lat. N	Long. E		depth,m	Ħ	°/°°	Bq/m <sup>3</sup>
			Gulf of ]	Finland			
USSR-3	59° 56 '	30°08'	06.08	4	0	0,08	27,0+4,5
USSR-7	59° 59 '	29° 52'	06.08	ſ	0	0,08	23,0+4,0
USSR-10	60°051	29°201	10.08	28	0	3,90	19,043,0
USSR-10	60°05	29°201	10.08	28	25	4,54	22,0+3,5
USSR-12	60°021	29°031	06.08	30	0	4,22	22,0+3,5
USSR-12	60°021	29°031	06.08	30	20	4,33	24,0+4,0
USSR-13	60°021	28°45'	06.08	35	0	4,25	23,0+4,0
USSR-13	60°021	28°45'	06.08	35	30	5,08	23,0+4,0
USSR-14	60°12'	28°41	07.09	30	0	3,09	23,0+4,0
USSR-15	59°55'	28°341	06.08	28	0	4,41	23,0+4,0
USSR-15	59°55°	28°34"	06.08	28	25	4,44	23,0+4,0
USSR-18	60°321	28°231	04.09	20	0	2,53	21,0±3,5
ussr-19	60°151	27°59'	06.07	34	0	4,40	22,0+3,5
USSR-19	60°151	27° 59°	06.07	34	30	7,41	20,0+3,5
USSR-5	59° 57 '	27°001	07.07	65	0	4,97	23,0+4,0
USSR-5	• 7 7 • 5 7 •	27°001	07.07	65	50	7,95	22,0+3,5

x

Continue table 1.

Sample index	Coordinates of sampling sit	tes of the ng site	Sampling date	Overall water	Sampling depth,	Salinity,	4806
-	Lat. N	Long. "E		dep th, m	Ħ	~~/ <sup>~</sup>	Bq/m <sup>)</sup>
			Gulf of	Finland			
USSR-8	59°431	250571	07.07	80	0	3,96	21,043,5
USSR-24	59°40'	24°401	70.60	80	0	4,98	20,0+3,5
USSR-24	59°40°	24°40'	70.00	8	75	8,58	23,0+4,0
USSR-11	59°261	23°091	11.07	96	0	6,07	21,0+3,5
USSR-11	59°261	23°091	11.07	96	90	9,70	19,0+3,0
USSR-27	Bay of C	Coporie	28.08	11	0	4,35	21,0+3,5
USSR-27	of	Coporie	28.08	11	10	4,46	20,043,5
USSR-28	Bay of C	Coporie	28.08	20	0	4,43	19,0±3,0
ussr-32	Bay of C	Coporie	28,08	13	0	4,34	21,0+3,5
			Courish	Gulf			
USSR-K1	Central	part	25.07	ц	0	2,67	18,0+3,0

Continue table 1.

Sample	Coordinat	tes of the	Sampling	Overall	Sampling	Concent	Concentration, Bq/m <sup>2</sup>	13402
index	sampling	ng site	date	water	depth,	134 <sub>6 a</sub>	137 <sub>na</sub>	
	Lat. N	Long. E		depth,m	e	2		137 <sub>CE</sub>
			Baltic	Sea Proper	ਮ			
BY-28	120°02	210051	12.07	105	0	47,047,0	175,0+21,0	0,27
BY-28	120.65	21°05'	12.07	105	100	32,046,0	123,0±15,0	0,26
USSR-6	58°00°	200451	12.07	96	0	33,0+6,0	141,0+17,0	0,23
USSR-6	55°001	20°45'	12.07	96	90	21,0±5,0	98,0+12,0	0,21
BY-15	57°201	20°03	13.07	230	0	40,047,0	198,0+23,0	0,20
BY-15	57°201	20°031	13.07	230	200	13,0±3,0	53,047,0	
USSR-46	190.05	190141	17,07	120	0	27,0+5,0	127,0±16,0	0,21
USSR-46	- 90 - 96	19014	17.07	120	100	29,044,0	123,0±14,0	0,24
USSR-50	55°30°	18°53'	18.07	85	0	26,0±4,0	103,0±12,0	
USSR-50	55°30°	18°53'	18.07	85	8	41,0+6,0	132,0±16,0	0,31
USSR-51	55°00'	14°00'	26.07	45	0	35,0±5,0	142,0±17,0	0,24
USSR-51	55°00'	14°00'	26.07	45	40	23,0+4,0	110,0±13,0	0,21
USSR-52	54°43'	12047.	26.07	22	0	26,0+4,0	125,0±15,0	0,21
USSR-52	540431	120471	26 <b>.</b> 07	22	20	27,0+6,0	141,0+18,0	0,19
USSR-53	54°281	120131	26 <b>.</b> 07	<u>с</u>	0	18,0+3,0	92,0±11,0	0,19
USSR-53	54°28'	120131	26°07	19	17	25,0+4,0	114,0+14,0	0,22

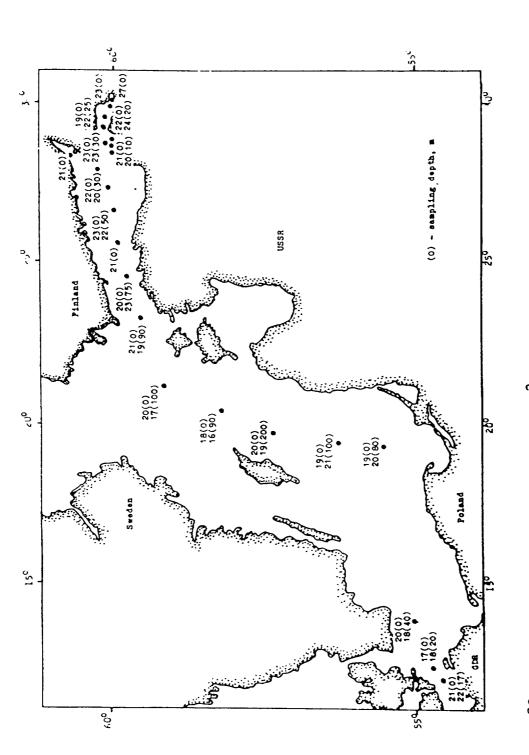
137 134-

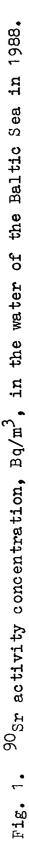
Sample	Coordinates	tes of the	Sampling	Overall	Sampling	Concentration,	ation, Bq/m <sup>3</sup>	13402
index	sampling	ng site	date	water	dep <b>th,</b>	134 c	127	
	Lat. N	Long. E		depth,m	ш	n D	Cs Cs	<sup>137</sup> <sub>Cs</sub>
			÷	Gulf of Fin	Finland			
ussr-3	59° 56 °	30°08'	06.08	4	0	6,0+2,0	23,0+5,0	0,27
ussr-7	59°59'	29°52'	06.08	ſ	0	4,8+1,5	18,0+4,0	0,27
USSR-10	60°05	29°201	10.08	28	0	30,0+4,0	105,0+12,0	0,29
USSR-10	60°05	29°201	10.08	28	25	35,0±5,0	138,0+16,0	0,25
USSR-12	60°021	29°03'	06.08	30	0	38,0±5,0	134,0±15,0	0,29
USSR-12	600021	150°031	06.08	30	20	33,0±5,0	150,0+17,0	0,22
USSR-13	60°02'	200451	06.03	35	0	42,047,0	160,0+20,0	0,26
USSR-13	60°02'	280451	06.08	35	30	43,0+6,0	165,0+19,0	0,26
USSR-14	60°121	280411	60.70	30	0	29,0+6,0	102,0±13,0	0,28
USSR-15	59°55'	28°341	06.08	28	0	36,0±6,0	147,0418,0	0,24
USSR-15	59°55'	28°341	06.08	28	25	੍ਰ	155,0+18,0	0,23
USSR-18	60°321	28°231	60.40	20	0	20,043,0	75,0±9,0	0,27
USSR-19	60°15'	27°59'	06°07	34	0	41,0±6,0	152,0±17,0	0,27
USSR-19	60°151	27°59'	06.07	34	30	42,047,0	167,0±20,0	0,25
USSR-5	59°57'	27°001	07 <b>.</b> 07	65	0	47,0+7,0	173,0±20,0	0,27
USSR-5	59°57°	27°00 °	07.07	65	50	31,0±6,0	115,0+14,0	0,27

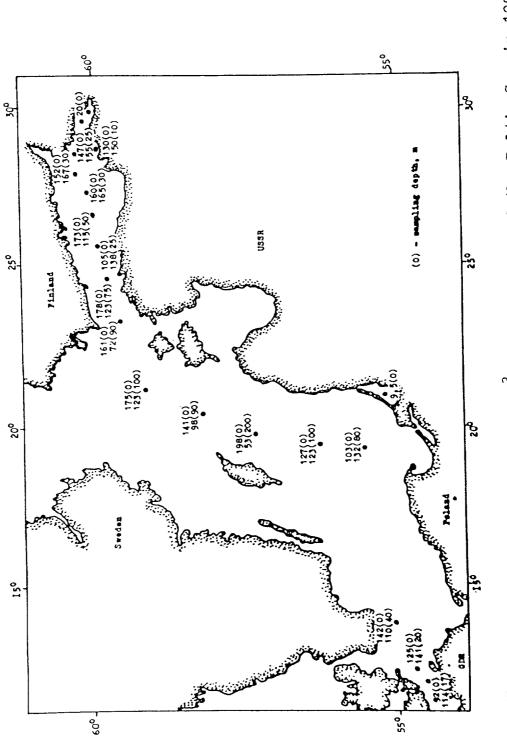
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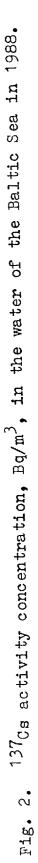
e         Coordinates of the Sampling         Overall         Sampling         Concentration, $Bq/m^3$ sampling site         date         water         depth,         134Gs         137Gs           Lat, N         Long. E         depth,         m         134Gs         137Gs           Lat, N         Long. E         depth,         m         134Gs         137Gs           Lat, N         Long. E         depth,         m         134Gs         137Gs           Gulf of Finland         Gulf of Finland         m         134Gs         137Gs           59°40'         24°40'         09.07         80         0         40,046.0         178.0219.0           59°26'         23°09'         11.07         96         0         23,044.0         161.0219.0           7         59°26'         23°09'         11.07         96         90         72.029.0           7         Bay of Coporie         28.08         11         10         23,044.0         133.0416.0           8         Sourieh         13         0         35,025.0         133.0416.0         1640.0           8         Sourie         28.08         11         10         23,044.0         133.0416.0	Con	Continue table	e 2 <b>.</b>						
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	ple	Coordina	Ъ О Г	Sampling	Overall	Sampling	Concentr		134 <sub>Cs</sub>
Lat, NLong. Edepth.mmLat, NLong. Edulf of Finland59°43'25°57'07.0780059°40'24°40'09.07807535.0 $\pm$ 6.059°40'24°40'09.07807535.0 $\pm$ 7.0159°40'24°40'09.07807535.0 $\pm$ 7.0159°26'23°09'11.0796020.0 $\pm$ 4.0759°26'23°09'11.0796020.0 $\pm$ 4.0759°26'23°09'11.0796020.0 $\pm$ 4.0759°26'23°09'11.0796020.0 $\pm$ 4.0759°26'23°09'11.0796020.0 $\pm$ 4.0759°26'23°09'111023.0 $\pm$ 4.0103.0 $\pm$ 12.0759°26'23°09'111023.0 $\pm$ 4.0103.0 $\pm$ 16.0759°26'23°09'111023.0 $\pm$ 4.0103.0 $\pm$ 16.08811023.0 $\pm$ 4.0103.0 $\pm$ 16.089.5035.0 $\pm$ 6.0132.0 $\pm$ 16.0897035.0 $\pm$ 6.0132.0 $\pm$ 16.0811023.0 $\pm$ 4.0103.0.2.16.0811023.0 $\pm$ 4.0103.0.2.16.0811023.0 $\pm$ 4.0133.0 $\pm$ 16.0811023.0 $\pm$ 4.0133.0 $\pm$ 16.0911023.0 $\pm$ 4.0133.0 $\pm$ 16.0111 <th>index</th> <th>i Lqmpli</th> <th>ng site</th> <th>da te</th> <th>water</th> <th>depth,</th> <th>134<sub>Ga</sub></th> <th>137<sub>Cs</sub></th> <th>137.</th>	index	i Lqmpli	ng site	da te	water	depth,	134 <sub>Ga</sub>	137 <sub>Cs</sub>	137.
Gulf of Finland $59^{\circ}43^{\circ}$ $25^{\circ}57^{\circ}$ $07.07$ $80$ $0$ $37.0\pm6.0$ $156.0\pm18.0$ $4$ $59^{\circ}40^{\circ}$ $24^{\circ}40^{\circ}$ $09.07$ $80$ $0$ $40.0\pm6.0$ $178.0\pm21.0$ $4$ $59^{\circ}40^{\circ}$ $24^{\circ}40^{\circ}$ $09.07$ $80$ $75$ $35.0\pm5.0$ $178.0\pm21.0$ $1$ $59^{\circ}26^{\circ}$ $24^{\circ}40^{\circ}$ $09.07$ $80$ $75$ $35.0\pm5.0$ $178.0\pm21.0$ $1$ $59^{\circ}26^{\circ}$ $24^{\circ}40^{\circ}$ $09.07$ $80$ $75$ $35.0\pm7.0$ $123.0\pm14.0$ $7$ $59^{\circ}26^{\circ}$ $23^{\circ}09^{\circ}$ $11.07$ $96$ $0$ $43.0\pm7.0$ $161.0\pm19.0$ $7$ $59^{\circ}26^{\circ}$ $23^{\circ}09^{\circ}$ $11.07$ $96$ $90$ $20.0\pm4.0$ $72.0\pm9.0$ $7$ $Bay$ $0f$ $00$ $11.07$ $96$ $90$ $20.0\pm4.0$ $151.0\pm19.0$ $7$ $Bay$ $0f$ $00$ $11.07$ $96$ $90$ $20.0\pm4.0$ $151.0\pm19.0$ $7$ $Bay$ $0f$ $00$ $11.07$ $0$ $39.0\pm7.0$ $151.0\pm19.0$ $8$ $Bay$ $0f$ $00$ $0$ $33.0\pm5.0$ $133.0\pm16.0$ $8$ $Bay$ $0f$ $00$ $0$ $36.0\pm6.0$ $133.0\pm16.0$ $8$ $13$ $0$ $20.06\pm0.0$ $0$ $35.0\pm5.0$ $133.0\pm16.0$ $8$ $13$ $0$ $20.06\pm0.0$ $132.0\pm16.0$ $132.0\pm16.0$ $8$ $13$ $0$ $2.000$ $2$ $0$ $2.2\pm0.8$		1			depth,m	Ħ	1		8 2
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59°40'       24°40'       09.07       80       75       35,0±5,0       123,0±14,0         59°26'       23°09'       11.07       96       0       43,0±7,0       161,0±19,0         59°26'       23°09'       11.07       96       90       20,0±4,0       72,0±9,0         59°26'       23°09'       11.07       96       90       20,0±4,0       72,0±9,0         Bay of Coporie       28.08       11       10       23,0±4,0       103,0±12,0         Bay of Coporie       28.08       11       10       23,0±4,0       103,0±16,0         Bay of Coporie       28.08       13       0       35,0±6,0       132,0±16,0         Bay of Coporie       28.08       13       0       36,0±6,0       132,0±16,0         Central part       25.07       5       0       2,2±0,8       9,5±2,5	R-24	59°40'	24°40'	70.60	80	0	40,0+6,0	78,0+21,	0,23
59°26'       23°09'       11.07       96       0       43,0±7,0       161,0±19,0         59°26'       23°09'       11.07       96       90       20,0±4,0       72,0±9,0         Bay of Coporie       28.08       11       0       39,0±7,0       151,0±19,0         Bay of Coporie       28.08       11       10       23,0±4,0       103,0±12,0         Bay of Coporie       28.08       11       10       23,0±4,0       103,0±12,0         Bay of Coporie       28.08       13       0       33,0±5,0       133,0±16,0         Bay of Coporie       28.08       13       0       36,0±6,0       132,0±16,0         Conrish Gulf       Conrish Gulf       0       2,2±0,8       9,5±2,5	R-24	100.05	24°40'	09°07	80	75	35,0±5,0	123,0±14,0	0,29
59°26'       23°09'       11.07       96       90       20,04,0       72,049,0         Bay of Coporie       28.08       11       0       39,047,0       151,0419,0         Bay of Coporie       28.08       11       10       23,044,0       103,0412,0         Bay of Coporie       28.08       11       10       23,044,0       103,0412,0         Bay of Coporie       28.08       13       0       33,045,0       133,0416,0         Bay of Coporie       28.08       13       0       36,046,0       132,0416,0         Bay of Coporie       28.08       13       0       36,046,0       132,0416,0         Courish Gulf       Courish Gulf       0       2,240,8       9,542,5	R-11	59°261	23°091	11.07	96	0	43,047,0	161,0+19,0	0,27
<ul> <li>Bay of Coporie 28.08 11 0 39,0±7,0 151,0±19,0</li> <li>Bay of Coporie 28.08 11 10 23,0±4,0 103,0±12,0</li> <li>Bay of Coporie 28.08 20 0 33,0±5,0 133,0±16,0</li> <li>Bay of Coporie 28.08 13 0 36,0±6,0 132,0±16,0</li> <li>Courish Gulf</li> <li>Central part 25.07 5 0 2,2±0,8 9,5±2,5</li> </ul>	R <b>-</b> 11	59°26	190052	11.07	96	06	20,044,0	72,0±9,0	0,28
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Bay of Coporie 28.08 13 0 36,046,0 Courish Gulf Central part 25.07 5 0 2,240,8	R <b>-</b> 28	o fo	)oporie	28.08	20	0	33,0±5,0	133,0+16,0	0,25
Central part 25.07 5 0 2,2±0,8 9,5±2,	R <b>-</b> 32	of	Joporie	28.08	13	0	36,0 <u>+</u> 6,0	132,0±16,0	0,27
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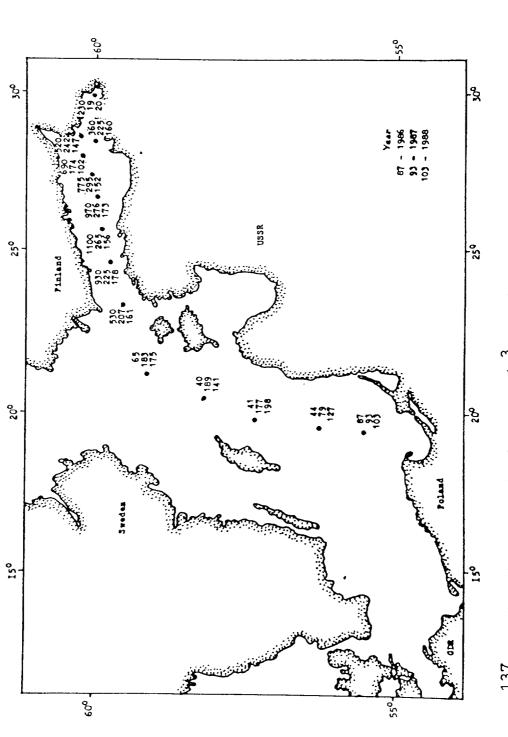
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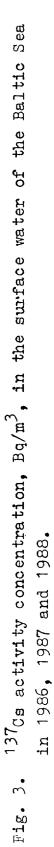












Strontium-90, cesium-134 and cesium-137 in water reservoirs of the Soviet Union's Baltic region ( 1986-1998 ).

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Study of the Baltic Sea basin's radioaktive contamination, carried out by the V.G.Khlopin Radium Institut, include, as a rule, determination of the radionuclides content in the water of rivers flowing into the Baltic Sea from the territory of the Soviet Union / 1-7 /. The obtained data show that before the Chernobyl NPP accident the Baltic river's radioactive contamination due to nuclear weapond tests was determined by long-lived radionuclides of global spread: strontium-90 and cesium-137. The activity ratio of cesium-137/strontium-90 that get to the earth surface as a result of global fallout, was 1.7 by that time. The time variation in the cesium-137/strontium-90 activity ratio in river waters is illustrated by Fig. 1. The concentration of cesium-137 in river waters in the 1980's was less 1 Bq/m<sup>3</sup>, and that of strontium-90, about 15 Bq/m<sup>3</sup>. A similar distribution of cesium-137 and strontium-90 was observed in the rivers flowing into the Baltic Sea from the territory of Finland / 8 /.

The said fractionation testifies to a stronger fixing of cesium-137 by soils in comparison with strontium-90. Thus, for a long period till the end of April 1986, Baltic rivers brought to the Baltic Sea relatively less cesium-137 contaminated water, which encouraged lowering of this radionuclide's activity in the Baltic Sea water.

The accident at the Chernobyl NPP changed the radiation situation in the Baltic Sea basin. The radioactive contamination, due to air transport, spread far beyond the Chernobyl NPP area. in particular northwards, having covered the Baltic Sea water area and the territories of the Baltic countries / 9 /. Determinations of radionuclides in atmospheric precipitations showed that the main long-lived dose- forming nuclides in the Baltic Sea region are cesium-134 and cesium-137, whose activity ratio ( cesium-134/cesium-137 ) in the initial period after the accident was 0.55. According to observations in the region of Leningrad atmospheric fallout of cesium-137 from the end of April to the end of May 1986 was 4610 Bq/m<sup>2</sup>, i.e. the level of cesium-137 in the region of Leningrad, accumulated as a result of global fallout, increased 3 times / 10 /. In this situation a necessity arose to carry out a broader survey of not only usually observed Baltic rivers, but also other water reservoirs directly or indirectly connected with the Baltic Sea. The

primary task of such a survey was, unconditionally, dosinetric estimation of the contamination level of one ore another water recervoir. This concerned, first of all, reservoir - sources of water-supply.

In the period May-July 1986 the Radium Institute surveyed over 30 water reservoirs in the territory of the Leningrad region, the Estonian, Latvian, Lithnanian Soviet Socialist Republics, the Kaliningrad region and the northern part of the Byelorussian SSR. Water reservoirs with a relatively high content of radioactive cesium remained under observation in 1987 and 1988. Radionuclides were determined in water samples by radiochemical methods.

The results obtained for the period 1986-1988 are surrarized in Table 1 and 2.

Consideration of the obtained results shows that in May-June 1986 both radionuclides of cesium were noted in all the mater reservoirs under survey. The values of the cesium-134/ cesium-137 activity ratio varied within the limits 0.37-0.68, being 0.5 on the average. In connection with the urgent character of work in 1986, some samples with a low content of radioactive cesium were measured with an insufficient statistic, hence in some cases there are only upper limits of the cesium-134 and cesium-137 contents in the tables.

In Fig. 2 there are presented concentrations of cesium-137 in waters of a number of great Baltic rivers in 1986, and the level that had existed in 1983-1984. In the same Fig. 2 the permissible concentration of cesium-137 in drinking water for a limited part of population, accepted in the USSR / 11 /, is cited for comparison. It follows from the juxtaposition that the cesium-137 content in the waters of the rivers under consideration, increased 100-200 times as compared to 1983-1984, but remained by 3-4 orders lower than the permissible concentration. The content of strontium-90 in the river's waters increased slightly; a comparison with the results of the previous years showed that the increase was within the limits of 30% of the 1983-1984 level.

All the results of cesium-137 determination in river and lake waters in the period 1986-1988 are presented in the map-chart of the USSR Baltic territory (Fig. 3). The obtained picture of cesium-137 distribution shows that contamination of

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rivers and lakes in the western part of the Baltic region is less than in the eastern one, which is the consequence of nonuniformity, "spottiness" of atmospheric radioactive fallouts in 1986 after the Chernobyl accident. Another essential feature of the arisen contamination of river and lake waters was fast lowering of the contamination level. One may expect further decrease of the cesium radionuclides' content in the water of Baltic rivers because intensive washing-out from soils isn't characteristic of cesium. An attempt was made to describe mathematically the character of variation of the cesium-137 content in the river's water depending on time, on the basis of available experimental data. In view of different character of the water-gathering areas and the contamination levels, empirical equations were selected separately for two groups of rivers: the Neva, the Narova, the Luga and the Daugava, the Neman. The best correspondence to experimental data was at selecting a hyperbolic curve equation for the first group of rivers , and an exponential curve, for the second one. Of course, the available experimental data being limited, one cannot clair. that the happening processes are exactly described by these equations, but they may be used for the practical purpose of determining the discharge of cesium radionuclides with river waters into the Baltic Sea.

In Fig. 4 and 5 there are presented experimental data on the cesium-137 content in river waters in 1986-1988 and calculated curves of this nuclide's content variation.

In Table 3 there are data on the water flow rate in the Neva river in 1986-1988 / 12 / and obtained by calculation monthly averaged values of the cesium-137 content in the Neva's water. In the period January-March 1986 the concentration of cesium-137 in the water of the Neva river is assumed equal 1  $Bq/m^3$ , proceeding from the data of 1983-1984. Applying the empirically obtained equation for the first group of rivers, the discharge of cesium-137 into the Baltic Sea with the Neva river waters in 1986-1988 is calculated. The obtained result has showed that about 11 TBq of cesium-137 got with the Neva waters into the Gulf of Finland in the period 1986-1988.

Using the data on the monthly average water flow rate and, in a number of cases, the annual average data on the water flow rate (Table 1 / 12-14 / ), it is possible to estimate in the same manner the discharge of cesium-137 with the waters of the Luga. the Narova, the Daugava and the Neman. The results of

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the calculation showed that in the period 1986-1988 the discharge of cesium-137 was: with the waters of the river Luga -0.6 TBq, the Narova - 1.4 TBq, the Daugava - 0.3 TBq, the Neman - 0.3 TBq. The summary discharge of cesium-137 in 1986-1988 with the waters of these rivers including the Neva, was, by estimation, 14 TBq.

The annual water discharge of the Neva river is 80 km<sup>3</sup> on the average, of the Luga - 3 km<sup>3</sup>, the Narova - 11 km<sup>3</sup>, the Daugava - 16 km<sup>3</sup>, the Neman - 17 km<sup>3</sup>, which add up to 127 km<sup>3</sup> a year, i.e. about 30% of the total river discharge into the Baltic Sea. Thus, the calculated discharge of cesium-137 with the waters of the said rivers of the Soviet littoral, estimated as 14 TBq for the period 1986-1988, may characterize this nuclide's coming with waters making 30% of the total river discharge into the Baltic Sea. References

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River	Sampling	C onc	Concentration, Bq/m <sup>3</sup>	/m <sup>3</sup>	134 <sub>CB</sub>		Average flow
(sampling grea)	da ve	90 <sub>Sr</sub>	134 <sub>CB</sub>	137 <sub>CB</sub>	137 <sub>CB</sub>	auring samp- ling period, m <sup>3</sup> /sec	tor severat yenrs, m <sup>3</sup> /sec
Neva (the mouth)	08.05.86	26.0+4.0	96.0+8.0	240.0+7.0	0.40	3010	2510
	17.12.86	26.0+4.5	9.5+1.0	22.0+2.0	0.43	1740	
	29.10.87	26.0+4.5	3.040.9	9.7+1.9	0.31	3230	
	23.06.88	20.0+3.5	6.5+2.9	31.0+3.0	0.21	3300	
Luga	26.05.86	<b>a</b> )	150.0+20.0	250.0+20.0	0.60	225	91.6
(50 km from the mouth)	17.07.87	22.5+3.4	20.0+5.0	50.6+6.0	0.40		-69
	03.11.87	a) -	20.0+5.8	45.046.2	0.44		_
	28,06,88	17.0+3.0	5.0	14.0+2.8	1		
	05.08.88	15.0+2.5	6.0+2.0	23.0+2.3	0.26		
Narva (the mouth)	22.05.86	ਦ	130.0+10.0	290.0+10.0	0.45		347
	17.07.87	18.0+3.0	7.9+1.3	22.1+2.4	0.36		
	03.11.87	ีย)	3.9+0.9	12.4+1.6	0.31		
	05.08.88	16.5+3.0	6.5+1.8	25.0+2.0	0.26		
Narva (the source)	22.05.86	(u-	130.0+10.0	290.0+10.0	0.45		322
Pirita (the mouth)	01.08.86	15.0+2.5	47.0+6.0	76.0+7.0	0.62		6.46
	27.07.87	11.5±1.7	16.9+3.5	50.4.46.0	0.34		
	03.08.88	11.0+2.0	7.5±1.5	30.0±1.7	0.25		

River ( aamuling area)	Sampling date	Conc	centration, Bq/m <sup>3</sup>	Em/E	134 <sub>CB</sub>	Plow during samp-	Average flow for several	
		90 <sup>Sr</sup>	134 <sub>CS</sub>	137 <sub>Cs</sub>	<sup>137</sup> CB	ling period, years, m <sup>3</sup> /sec m <sup>3</sup> /sec	years, m <sup>3</sup> /sec	
Pjarnu (the mouth)	01.08.86	16.0+3.0	11.0+5.0	30.0+5.0	0.37		47.9	
	27.07.87	12.0+1.8	2.6+0.6	7.2+1.0	0.36			
	03.08.88	10.0+1.8	1	2.4±0.6	1			
Geuja	23.06.86	( a -	с, 20	34.0±7.0	1		67.8	
(60 km from the mouth)								
Daugava (the mouth)	31.07.86	22.0+4.0	Z 10	24.0+6.0	I		500	
1	26.07.87	21.5+3.2	1.5±0.4	A.1+0.8	0.37			-
	02.08.88	25.5+4.5	ł	21.4	1			70.
Daugava	27.06.86	25.0+4.5	33.0±10.0	66.0+10.0	0 • 50		449	_
(266 km from the mouth)								
Venta (the mouth)	23.07.86	11.0+2.0	<i>と</i> 10	23.0+5.0	t		64.7	
	20.07.87	12.5+2.0	2.4±0.6	6.9+1.0	0.35			
	18.07.88	13.0+2.3	4.7+1.0	14.2+1.7	0.33			
Barta (the mouth)	23.07.86	8.2+1.5	< 10	16.046.0	1		19.3	
	20.07.87	15.5+2.5	4.7+0.9	13.1+1.6	0.36			
	18.07.88	14.5+2.5	( มี เ	(°)-	I			
Dane (the mouth)	20.07.87	17.0+2.5	1.740.5	5.0±1.0	0.34		2.52	
	26.07.88	8.0+1.5	1	<0.7	ł			
Neman (the mouth)	25.07.86	10.5±2.0	×18	27.0+7.0	ı	299	540	
	23.07.87	14.0+2.0	0.9+0.4	2.5+0.6	0.36			
	26.07.88	14.0+2.5	ţ	<0.7	1			

Table 1. continued.

90 <sub>Sr</sub> $90_{Sr}$ $134_{Gs}$ $137_{Gs}$ $137$	River (complace conc.)	Sampling 32+5	Conc	Concentration, Bq/m <sup>3</sup>	q/m <sup>3</sup>	134.CB	Flow during grann-	Average flow for several
z5.06.86 $21.0\pm 3.5$ $20$ $33.0\pm 10.0$ $-$ km from the mouth) $26.06.86$ $22.5\pm 4.0$ $415$ $90.0\pm 10.0$ $-$ km from the mouth) $24.07.86$ $33.0\pm 6.0$ $39.0\pm 6.0$ $39.0\pm 6.0$ $ -$ 1(the mouth) $24.07.86$ $33.0\pm 6.0$ $39.0\pm 6.0$ $   -$ a(the mouth) $24.07.86$ $33.0\pm 6.0$ $39.0\pm 6.0$ $     -$ a(the mouth) $29.05.86$ $15.0\pm 2.5$ $68.0\pm 17.0$ $15.0\pm 2.0$ $0.44$ $ 20.05.86$ $15.0\pm 2.5$ $68.0\pm 17.0$ $150.0\pm 20.0$ $0.44$ $ 21.06.88$ $18.0\pm 3.0$ $22.7\pm 5.5$ $113.0\pm 5.0$ $0.44$ $ 21.06.88$ $18.0\pm 3.0$ $120.0\pm 20.0$ $0.44$ $       0$ (the mouth) $20.05.86$ $17.0\pm 3.0$ $120.0\pm 20.0$ $0.44$ $        0$ (the mouth) $20.05.86$ $17.0\pm 3.0$ $02.0\pm 0.0$ $0.24.0$ $0.24.0$ $0.53$ $ 0$ (the mouth) $20.05.86$ $17.0\pm 3.0$ $02.0\pm 0.0$ $0.24.0$ $0.53$ $ 0$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ $0.44$ $14$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ $0$ <tr< th=""><th>(ap.18 Suitduns)</th><th></th><th>90<sub>Sr</sub></th><th>134<sub>Cs</sub></th><th>137<sub>Cs</sub></th><th>137<sub>CB</sub></th><th>ling period, m<sup>3</sup>/sec</th><th></th></tr<>	(ap.18 Suitduns)		90 <sub>Sr</sub>	134 <sub>Cs</sub>	137 <sub>Cs</sub>	137 <sub>CB</sub>	ling period, m <sup>3</sup> /sec	
km from the mouth)a26.06.86 $22.5\pm4.0$ $415$ $90.0\pm10.0$ athe mouth) $24.07.86$ $33.0\pm6.0$ $39.0\pm6.0$ $75.0\pm7.0$ $0.52$ 1(the mouth) $24.07.86$ $33.0\pm6.0$ $39.0\pm6.0$ $30.0\pm6.0$ $30.0\pm6.0$ $30.0\pm6.$	Neman	25.06.86	21.0+3.5		33.0+10.0	1		
a $26.06.86$ $22.5\pm4.0$ $4.15$ $90.0\pm10.0$ $-$ Im from the mouth) $24.07.86$ $33.0\pm6.0$ $39.0\pm6.0$ $39.0\pm6.0$ $75.0\pm7.0$ $0.52$ Im the mouth) $24.07.86$ $33.0\pm6.0$ $39.0\pm6.0$ $39.0\pm6.0$ $75.0\pm7.0$ $0.52$ Image: the mouth) $24.07.86$ $33.0\pm6.0$ $39.0\pm6.0$ $39.0\pm6.0$ $0.52.5\pm7.0$ $0.52$ Image: the mouth) $24.07.86$ $15.0\pm2.5$ $68.0\pm17.0$ $75.0\pm7.0$ $0.45$ $5$ Image: the mouth) $29.05.86$ $15.0\pm2.5$ $68.0\pm17.0$ $150.0\pm20.0$ $0.44$ $14$ Image: the mouth) $20.05.86$ $18.0\pm3.0$ $32.7\pm5.5$ $113.0\pm5.0$ $0.44$ $14$ Image: the mouth) $20.05.86$ $18.5\pm3.5$ $-25.0\pm2.8$ $-26.0\pm2.8$ $-26.0\pm0.8$ $14.7.0\pm3.0$ $0.0\pm10.0$ $0.44$ $14$ Image: the mouth) $20.05.86$ $17.0\pm3.0$ $62.0\pm9.0$ $117.0\pm9.0$ $0.53$ $1$ Image: the mouth) $20.05.86$ $17.0\pm3.0$ $62.0\pm9.0$ $117.0\pm9.0$ $0.53$ $1$ Image: the mouth) $20.05.86$ $17.0\pm3.0$ $21.0\pm0.0$ $0.53$ $1$ Image: the mouth) $20.05.86$ $17.0\pm3.0$ $21.0\pm0.0$ $0.53$ $1$ Image: the mouth) $20.05.86$ $19.0\pm3.5$ $-2.1.4$ $-2.1.4$ $-2.1.4$ Image: the mouth) $20.05.86$ $19.0\pm3.5$ $-2.1.4$ $-2.1.4$ $-2.1.4$ $-2.1.4$ Image: the mouth) $20.05.86$ $19.0\pm3.5$ $-2.1.4$ $-2.1.4$	(280 km from the mouth	(1						
<pre>km from the mouth) 1 (the mouth) 24.07.86 33.0<math>\pm</math>6.0 39.0<math>\pm</math>6.0 75.0<math>\pm</math>7.0 0.52 22.07.87 19.0<math>\pm</math>3.5 <math>\pm</math> 3.0 <math>\pm</math> 0 0.52 22.07.87 19.0<math>\pm</math>3.5 <math>\pm</math> 3.0 <math>\pm</math> 0 0.52 22.07.88 16.0<math>\pm</math>3.5 <math>\pm</math> 0 0.6 26.07.88 16.0<math>\pm</math>3.5 <math>\pm</math> 0 0.6 26.07.88 15.0<math>\pm</math>7.5 5 113.0<math>\pm</math>20.0 0.45 5 0 (the mouth) 29.05.86 15.0<math>\pm</math>2.5 68.0<math>\pm</math>17.0 150.0<math>\pm</math>20.0 0.44 14 23.06.88 18.5<math>\pm</math>3.5 68.0<math>\pm</math>17.0 150.0<math>\pm</math>20.0 0.44 14 23.06.88 18.5<math>\pm</math>3.5 68.0<math>\pm</math>10.0 270.0<math>\pm</math>10.0 0.53 1 (the mouth) 20.05.86 17.0<math>\pm</math>3.0 120.0<math>\pm</math>10.0 270.0<math>\pm</math>10.0 0.53 1 (the mouth) 20.05.86 17.0<math>\pm</math>3.0 120.0<math>\pm</math>10.0 270.0<math>\pm</math>10.0 0.53 1 (the mouth) 20.05.86 17.0<math>\pm</math>3.5 5.0<math>\pm</math>113.0<math>\pm</math>5.0 0.53 1 (the mouth) 20.05.86 17.0<math>\pm</math>3.0 120.0<math>\pm</math>10.0 270.0<math>\pm</math>10.0 0.53 1 (the mouth) 20.05.86 17.0<math>\pm</math>3.5 11.0 0.210.0 0.38 4 (the mouth) 20.05.86 18.0<math>\pm</math>3.5 1.0 0.0210.0 0.53 1 (the mouth) 20.05.86 19.0<math>\pm</math>3.5 - 2.1.4 (the mouth) 20.05.86 19.0<math>\pm</math>3.5 - 3.1.41.0 - 23.06.88 19.0<math>\pm</math>3.5 1.0 0.53 0.140.0 - 1.0 0.53 0.140.0 - 1.0 0.53 0.140.0 - 23.06.88 19.0\pm3.5 - 2.1.4 2.1.4 2.2.05.86 18.0\pm3.0 2.0.2.90.0 0.38 4 2.2.04 2.2.05.86 18.0\pm3.0 2.0.2.90.0 0.53 1 2.2.05.86 19.0\pm3.0 2.000.90.0 0.53 1 2.2.05.86 - 2.005.86 19.0\pm3.5 - 2.1.4 2.1.4 2.2.05.88 19.0\pm3.0 2.0.50.88 19.0\pm3.0 2.000.90.0 0.53 1 2.2.04 2.2.05.86 18.0\pm3.0 2.000.90 0.000.91 2.2.06.88 19.0\pm3.5 - 2.1.4 2.1.4 2.2.05.86 - 2.2.05.86 - 2.1.4 2.1.4 2.2.05.86 - 2.1.4 2.1.4 2.1.4 2.2.05.86 - 2.1.4 2.1</pre>	Vilije	26.06.86	22.5+4.0		90.0+10.0	ı	73.4	109
1 (the mouth) $24.07.86$ $33.0\pm6.0$ $39.0\pm6.0$ $75.0\pm7.0$ $0.52$ $22.07.87$ $19.0\pm3.5$ $-a$ ) $-a$ ) $-a$ ) $-a$ ) $-a$ ) $26.07.88$ $16.0\pm3.0$ $-c$ $68.0\pm17.0$ $150.0\pm20.0$ $0.45$ $5$ $26.07.88$ $15.0\pm2.5$ $68.0\pm17.0$ $150.0\pm20.0$ $0.45$ $5$ $a$ (the mouth) $29.05.86$ $15.0\pm2.5$ $68.0\pm17.0$ $150.0\pm20.0$ $0.44$ $14$ $21.06.88$ $18.0\pm3.0$ $32.7\pm5.5$ $113.0\pm5.0$ $0.28$ $a$ (the mouth) $20.05.86$ $17.0\pm3.0$ $120.0\pm10.0$ $270.0\pm10.0$ $0.44$ $14$ $(the mouth)$ $20.05.86$ $17.0\pm3.0$ $52.0\pm9.0$ $117.0\pm9.0$ $0.53$ $1$ $(the mouth)$ $20.05.86$ $18.0\pm3.0$ $120.0\pm10.0$ $270.0\pm10.0$ $0.38$ $4$ $(the mouth)$ $20.05.86$ $18.0\pm3.0$ $210.0$ $0.31$ $1.0\pm0.6$ $-$ $23.06.88$ $18.0\pm3.0$ $218.0$ $ 21.4$ $-$ 21.4 $-23.06.88 17.0\pm3.0 218.0  21.4 -1.0\pm0.6 -1.0\pm0.6 -1.0\pm0.6 -2.005.86 18.0\pm3.0 218.0  2.14 -2.1.4$ $-2.0.05.86 19.0\pm3.0  140.0\pm0.0 -1.0\pm40.6 -2.0.05.86 19.0\pm3.0  33.0\pm9.0 66.0\pm8.0 0.38 4 44.000000000000000000000000000000000000$	(245 km from the mouth	( t						
22.07.8719.0 $\pm 3.5$ $a$	Pregol (the mouth)		33.0+6.0	39.046.0	75.047.0	0.52	21.8	85.3
26.07.8816.0 $\pm$ 3.0- $<$ 0.6-a (the mouth)29.05.8615.0 $\pm$ 2.568.0 $\pm$ 17.0150.0 $\pm$ 20.00.455ov (the mouth)29.05.8618.0 $\pm$ 3.032.7 $\pm$ 5.5113.0 $\pm$ 5.00.2814ov (the mouth)20.05.8618.5 $\pm$ 3.5-a)180.0 $\pm$ 20.00.441423.06.8818.5 $\pm$ 3.5-a)180.0 $\pm$ 20.00.441423.06.8818.5 $\pm$ 3.5-25.0 $\pm$ 2.8(the mouth)20.05.8617.0 $\pm$ 3.5-25.0 $\pm$ 2.8-23.06.8819.0 $\pm$ 3.5-25.0 $\pm$ 10.00.441423.06.8819.0 $\pm$ 3.5-2.1.4(the mouth)20.05.8617.0 $\pm$ 3.5-2.1.423.06.8818.5 $\pm$ 3.5-2.1.41(the mouth)20.05.8617.0 $\pm$ 3.05.0 $\pm$ 10.00.531(the mouth)20.05.8618.0 $\pm$ 3.02.1.423.06.8817.0 $\pm$ 3.05.0 $\pm$ 10.00.531m from the mouth)22.06.8817.0 $\pm$ 3.05.0 $\pm$ 0.00.38423.06.8819.0 $\pm$ 3.0-2.1.424.023.06.8819.0 $\pm$ 3.0125.05.8618.0 $\pm$ 3.033.0 $\pm$ 9.00.534a from the mouth)22.06.8819.0 $\pm$ 3.0140.020.05.8626.05.		22.07.87	19.0+3.5	ເມ]	-a)	1		
a (the mouth) 29.05.86 $15.0\pm2.5$ $68.0\pm17.0$ $150.0\pm20.0$ 0.45 5 21.06.88 $18.0\pm3.0$ $32.7\pm5.5$ $113.0\pm5.0$ 0.28 21.06.88 $18.5\pm3.5$ $-a$ $180.0\pm20.0$ $410.0\pm20.0$ 0.44 $14$ 23.06.88 $18.5\pm3.5$ $- 25.0\pm2.8$ $-a$ $14$ $-25.0\pm2.8$ $-$ (the mouth) 20.05.86 $17.0\pm3.0$ $120.0\pm10.0$ $270.0\pm10.0$ 0.44 $-$ (the mouth) 20.05.86 $17.0\pm3.0$ $120.0\pm10.0$ $270.0\pm10.0$ 0.44 $-$ (the mouth) 20.05.86 $17.0\pm3.0$ $120.0\pm10.0$ $270.0\pm10.0$ $ -$ (the mouth) 20.05.86 $17.0\pm3.0$ $         -$		26.07.88	16.0+3.0	1	< 0.6	ı		-7
21.06.88 $18.0+3.0$ $32.7\pm5.5$ $113.0\pm5.0$ $0.28$ ov (the mouth) $20.05.86$ $-a$ $180.0\pm20.0$ $410.0\pm20.0$ $0.44$ $14$ $23.06.88$ $18.5\pm3.5$ $ 25.0\pm2.8$ $  25.0\pm2.8$ $-$ (the mouth) $20.05.86$ $17.0\pm3.0$ $120.0\pm10.0$ $270.0\pm10.0$ $0.44$ $14$ $23.06.88$ $19.0\pm3.5$ $ 26.0\pm0.8$ $   -$ (the mouth) $20.05.86$ $17.0\pm3.0$ $62.019.0$ $117.0\pm9.0$ $0.53$ $ 23.06.88$ $19.0\pm3.5$ $      23.06.88$ $18.0\pm3.0$ $62.019.0$ $117.0\pm9.0$ $0.53$ $  23.06.88$ $17.0\pm3.0$ $        23.06.88$ $17.0\pm3.0$ $         17.0mm$ $20.05.86$ $18.0\pm3.0$ $33.0\pm9.0$ $86.0\pm6.0$ $  -$ <td>Vuoksa (the mouth)</td> <td>29.05.86</td> <td>15.0+2.5</td> <td>68.0+17.0</td> <td>150.0420.0</td> <td>0.45</td> <td>532</td> <td>252</td>	Vuoksa (the mouth)	29.05.86	15.0+2.5	68.0+17.0	150.0420.0	0.45	532	252
ov (the mouth) 20.05.86 $-a$ 180.0±20.0 410.0±20.0 0.44 14 23.06.88 18.5±3.5 - 25.0±2.8 - (the mouth) 20.05.86 17.0±3.0 120.0±10.0 270.0±10.0 0.44 23.06.88 19.0±3.5 - 6.0±0.8 - (the mouth) 20.05.86 17.0±3.0 62.019.0 117.0±9.0 0.53 1 23.06.88 18.5±3.5 - $-2.1.4$ - (the mouth) 20.05.86 18.0±3.0 51.0±10.0 - 19 $\pm 0.6$ - m from the mouth) 22.06.88 19.0±3.5 - 140.0±20.0 26.0±0.8 - m from the mouth) 22.06.88 19.0±3.5 - 140.0±20.0 20.0±20.0 0.64 m from the mouth) 22.06.88 19.0±3.5 12.5±3.7 45.5±3.7 0.27		21.06.88	18.0+3.0	32.7+5.5	113.0+5.0	0.28		
$ \begin{array}{llllllllllllllllllllllllllllllllllll$	Volkhov (the mouth)	20.05.86	a)	180.0+20.0	410.0+20.0	0.44	1480	660
(the mouth) $20.05.86$ $17.0\pm3.0$ $120.0\pm10.0$ $270.0\pm10.0$ $0.44$ $23.06.88$ $19.0\pm3.5$ - $6.0\pm0.8$ $10.0$ $44$ $23.06.88$ $17.0\pm3.0$ $62.0\underline{1}9.0$ $117.0\pm9.0$ $0.53$ $11$ $23.06.88$ $18.5\pm3.5$ - $114$ $114$ $114$ $23.06.88$ $18.0\pm3.0$ $-12.0$ $-12.0$ $0.53$ $-117.0\pm9.0$ $23.06.88$ $18.0\pm3.0$ $-12.0$ $-12.0$ $-12.0$ $-12.0$ $23.06.88$ $17.0\pm3.0$ $-12.0$ $-19.0.6$ $-11.4$ $-11.4$ $23.06.88$ $17.0\pm3.0$ $-12.0$ $-19.0.6$ $-11.4$ $-11.4$ $23.06.88$ $17.0\pm3.0$ $-12.0$ $-19.0.6$ $-11.4$ $-11.4$ $23.06.88$ $19.0\pm3.0$ $-11.00$ $-11.4$ $-11.4$ $-11.4$ $23.06.88$ $19.0\pm3.0$ $-11.00$ $-11.4$ $-11.4$ $-11.4$ $20.05.86$ $18.0\pm3.0$ $-11.00$ $-11.4$ $-11.4$ $-11.4$ $20.05.86$ $-19.0\pm3.0$ $-11.4$ $-11.4$ $-11.4$ $-11.4$ $26.05.86$ $-1140.0\pm20.0$ $220.0\pm20.0$ $0.64$ $-140.0\pm20.0$ $0.64$ $140.0\pm20.0$ $220.0\pm20.0$ $220.0\pm20.0$ $0.64$ $-140.0\pm20.0$ $0.64$		23.06.88	18.5+3.5	I	25.0+2.8	ı		
(the mouth)23.06.88 $19.0\pm3.5$ - $6.0\pm0.8$ $7.0\pm3.0$ $0.53$ $1$ (the mouth)20.05.86 $18.0\pm3.0$ $62.0\pm9.0$ $117.0\pm9.0$ $0.53$ $1$ 23.06.88 $18.0\pm3.0$ $218.0\pm3.0$ $218.0$ $53.0\pm10.0$ $ 1$ 23.06.88 $17.0\pm3.0$ $218.0$ $53.0\pm10.0$ $ 1$ 23.06.88 $17.0\pm3.0$ $ 1.9\pm0.6$ $ 1.9\pm0.6$ $-$ 23.06.88 $17.0\pm3.0$ $33.0\pm9.0$ $86.0\pm8.0$ $0.38$ $4$ m from the mouth)22.06.88 $19.0\pm3.5$ $ 3.4\pm1.0$ $-$ 26.05.86 $ 140.0\pm20.0$ $220.0\pm20.0$ $0.64$ m from the mouth)28.06.88 $19.5\pm3.5$ $12.5\pm3.7$ $45.5\pm3.7$ $0.27$	Sjas (the mouth)	20.05.86	17.0+3.0	120.0+10.0	270.0+10.0	0.44	91.8	52.3
a (the mouth) $20.05.86$ $17.0\pm3.0$ $62.019.0$ $117.0\pm9.0$ $0.53$ 1 23.06.88 $12.5\pm3.5$ - $-1.4$ - $-1.4$ - $-1.4$ - $-21.4$ - $-21.4$ - $-23.06.88$ $17.0\pm3.0$ $-19.0$ $53.0\pm10.0$ - $1$ $-19\pm0.6$ - $-1$ $-10\pm0.4100$ - $-10\pm0.6$ - $-140.0\pm20.0$ $-26.05.86$ $19.0\pm3.5$ - $-140.0\pm20.0$ $220.0\pm20.0$ $0.64$ - $-140.0\pm20.0$ $220.0\pm20.0$ $0.64$		23.06.88	19.0+3.5	ı	6.0+0.8	1		
23.06.88 $1e.5\pm3.5$ - $\angle 1.4$ -(the mouth)20.05.86 $18.0\pm3.0$ $\angle 18.0$ $53.0\pm10.0$ -23.06.88 $17.0\pm3.0$ $\angle 1.9\pm0.6$ -123.05.86 $18.0\pm3.0$ $33.0\pm9.0$ $86.0\pm8.0$ 0.38km from the mouth) $22.06.88$ $19.0\pm3.6$ - $3.4\pm1.0$ -26.05.86 $19.0\pm3.5$ - $3.4\pm1.0$ -km from the mouth) $28.06.88$ $19.0\pm3.5$ - $3.4\pm7.0$ -26.05.86 $19.0\pm3.5$ - $140.0\pm20.0$ $220.0\pm20.0$ $0.64$		20.05.86	17.0+3.0	62.019.0	117.0+9.0	0.53	133	60.4
(the mouth) 20.05.86 $18.0\pm3.0 < 18.0$ 53.0 $\pm10.0$ - 1 23.06.88 $17.0\pm3.0 < 18.0$ 53.0 $\pm10.0$ - 1.9\pm0.6 - 23.05.86 $18.0\pm3.0$ 33.0 $\pm9.0$ 86.0 $\pm8.0$ 0.38 4 km from the mouth) 22.06.88 $19.0\pm3.5$ - 3.4 $\pm1.0$ - 26.05.86 - 140.0\pm20.0 220.0\pm20.0 0.64 km from the mouth) 28.06.88 $19.5\pm3.5$ $12.5\pm3.7$ $45.5\pm3.7$ 0.27		23.06.88	18.5+3.5	1	21.4	i		
23.06.88 17.0±3.0 - 1.9±0.6 - 20.05.86 18.0±3.0 33.0±9.0 86.0±8.0 0.38 4 20.00 the mouth) 22.06.88 19.0±3.5 - 3.4±1.0 - 26.05.86 - 140.0±20.0 220.0±20.0 0.64 the mouth) 28.06.88 19.5±3.5 12.5±3.7 45.5±3.7 0.27	Ojat (the mouth)	20.05.86	18.0+3.0	<18.0	53.0±10.0	1	128	52.2
20.05.86 18.0 <u>+</u> 3.0 33.0 <u>+</u> 9.0 86.0 <u>+</u> 8.0 0.38 4 km from the mouth) 22.06.88 19.0 <u>+</u> 3.5 - 3.4 <u>+</u> 1.0 - 26.05.86 - 140.0 <u>+</u> 20.0 220.0 <u>+</u> 20.0 0.64 km from the mouth) 28.06.88 19.5+3.5 12.5+3.7 45.5+3.7 0.27		23.06.88	17.043.0	•	1.9+0.6	1		
km from the mouth)       22.06.88       19.0±3.5       -       3.4±1.0       -         26.05.86       -       140.0±20.0       220.0±20.0       0.64         km from the mouth)       28.06.88       19.5+3.5       12.5+3.7       45.5+3.7       0.27	Svir	20.05.86	18.0+3.0	33.049.0	86.0±8.0	0.38	400	500
<pre>km from the mouth) 28.06.88 19.5+3.5 12.5+3.7 45.5+3.7 0.27</pre>	from the		19.0+3.5	ł	3.4±1.0	ł		
the mouth) 28.06.88 19.5+3.5 12.5+3.7 45.5+3.7	Saba	26.05.86	i <b>I</b>	140.0+20.0	220.0±20.0	0.64	14.8	8.75
	(12 km from the mouth)	) 28.06.88	19.5+3.5	12.5±3.7	45.5±3.7	0.27		

Table 1. continued.

Table 1. continued.	nued.						
River	Sampling	Concen	entration, Bq/m <sup>3</sup>	cm/f	134 <sub>CB</sub>	F].ow dinting somo-	Average flow for several
(sampling area)	date	90 <sup>S r</sup>	134 <sub>C</sub> s	137 <sub>Cs</sub>	137 <sub>CB</sub>	ling period, m <sup>3</sup> /sec	1
Pljussa	26.05.86		82.0+15.0	120.0+10.0		67.2	36.8
(15 km from the mouth )28.06.88 Cherekha 27.06.86	)28.06.88 27.06.86	12.0 <u>+</u> 2.0 20.0 <u>+</u> 3.5	1.8+0.6 ≺20	6.8 <u>+</u> 1.0 54.0 <u>+</u> 7.0	9 V • 0 1	1.77	15.8
(14 km from the mouth)							

a) - not analysed
- below detection limit

Table 2. Concentrations of strontium-90, cesium-134 and cosium-137 in water of the soviet lakes of the Baltic Sea region in 1986-1988.

Lake	Sampling		Concentration, Bq/m <sup>3</sup>	/m <sup>3</sup>	134 <sub>Ca</sub>
	date	90 <sub>Sr</sub>	134 <sub>C</sub> a	137 <sub>CB</sub>	137 <sub>C 8</sub>
Ladozhskoe	08.05.86	26.044.5	170.0+30.0	340.0+30.0	0.50
	17.12.86	28.5+5.0	15.0+3.8	47.5+4.8	0.32
	29.10.87	26.54.5	5.3+1.3	16.0+2.4	0.33
	23.06.88	20.5+3.5	I	7.9+2.6	ī
Chudskoe	22.05.86	(ដ [	56.0+10.0	130.0+10.0	0.13
	16.07.87	15.0+2.5	6 • 0 · 1 · L • V	13.5+1.6	0.35
Razliv	05.05.86	28.0+5.0	440.0+30.0	730.0+40.0	0.60
	29.06.87	੍ਰ ਰ	11.5+4.0	34.3+6.0	0.34
	04.11.87	16.0+3.0	3.2+0.7	10.3±1.3	0.31
	25.05.88	12.0+2.0	1	11.5+1.3	t
Krasnoselskoe	29.05.86	(ะ เ	130.0+30.0	310.0+30.0	0.42
	21.06.88	20.5+3.5	8.3+2.4	32.5+2.9	0.26
Savozero	19.05.86	19.0±3.0	28.0+9.0	76.0+8.0	0.37
	22.06.88	17.0+3.0	ı	< 2.2	ı
Spas-Katorskoe	26.05.86	(ธ.	270.0+20.0	570.0+20.0	0.47
	28.06.88	16.5+3.0	11.8+3.5	44.0+4.0	0.27

Lake	Sampling		Concentration, $Rq/m^3$	, <sup>m</sup> 3	134 <sub>C B</sub>	
	ດສ te	90 <sup>Sr</sup>	134 <sub>Cs</sub>	137 <sub>08</sub>	137 <sub>G.9</sub>	
Cirmas	27,06,86	(a_	لا 23	27.0+8.0	I	
Paunkj <b>ula</b> sskoe	23.06.86	ີ ອີ	31.048.0	75.0+8.0	0•4:1	
Virtej <b>arv</b>	23.06.86	<b>1</b> 0)	Z 20	24.0+8.0	ł	
	16.07.87	11.5+2.0	1.0-1-0.4	2.640.6	0.38	
Babites	24.06.86	( 1 1	22.0+7.0	36.0+7.0	0.60	
Naroch	26.06.86	58.5+10.0	39.0+8.0	62.048.0	0.63	-74
Trakaj	25.06.86	38.0+7.0	Z 20	45.0+8.0	ł	-

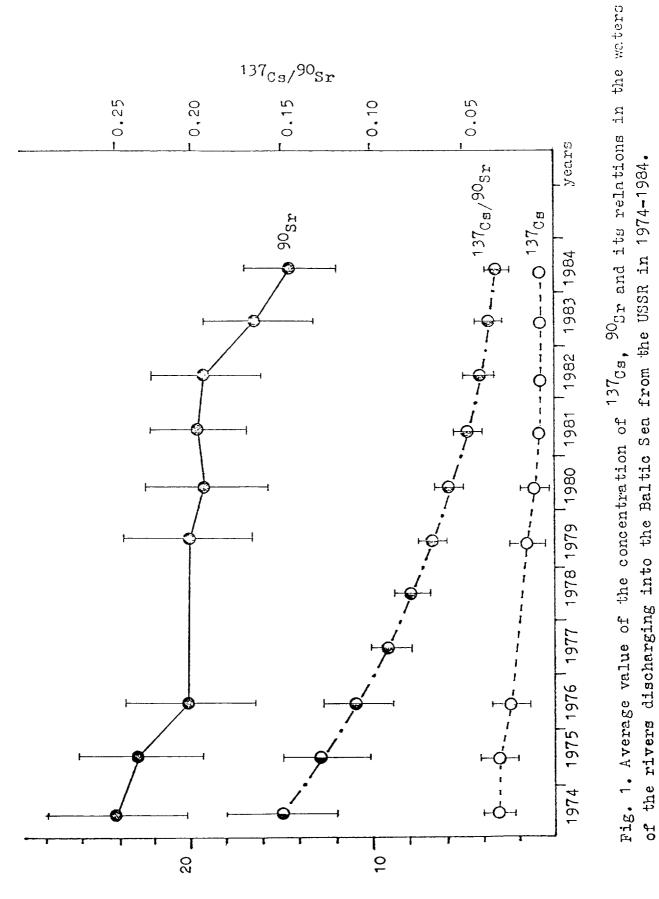
Table 2. continued.

a) - not analysed

Table 3. Evaluation of the flow of cesium-137 in the water of the Neva river in 1986-1988. ( on the basis of calculated concentrations of cesium-137 ).

ry Lary	1986			1987			1928	
ry lary	13 Bq	<sup>7</sup> Cg <sup>137</sup> Cs /m <sup>3</sup> TBq/ month	a Flow, m <sup>3</sup> /sec	<sup>137</sup> Cs Bq/m <sup>3</sup>	137 <sub>02</sub> TBq/ month	Flow, m <sup>3</sup> /sec	137 <sub>Cs</sub> Bq/m <sup>3</sup>	137 <sub>05</sub> TBq/ month
lary.	0	1 0.0036		36.5	0.15	1980	19.5	0.10
<b>_</b> .	0	1 0.0035		34.0	0.13	2130	18.9	0.10
		1 0.0046		32.0	0.14	2250	18.3	0.11
		1036 <sup>*)</sup> 0.5	2570	29.7	0.20	3190	17.7	0.14
May 3010			2910	28.0	0.22	3350	17.2	0.15
June 3050	<b>-</b>	1 1.00	3060	26.5	0.21	3300	16.7	0.14
July 2950		90.00	3100	25.2	0.21	3170	16.2	0.14
August 2820		0 0.53	3160	23.9	0.20	3080	15.8	0.13
September 2790		8 0.42	3240	22.9	0.19	3000	15.4	0.12
October 2710		0 0.37	3230	21.9	0.19	3010	15.0	0.12
November 2670		5 0.31	2830	21.0	0.15	2620	14.6	0.10
December 1740		0.19	1890	20.2	0.10	1520	14.3	0.06
Sum:		7.8			2.1			1.4

\*) - April 29-30, 1986.



Concentrations of 90Sr and 137Cs, Bq/m9

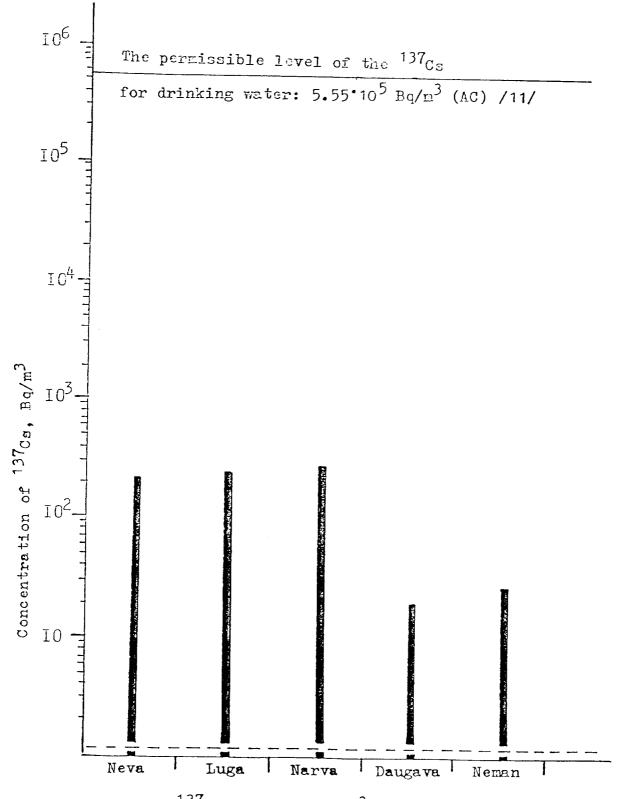


Fig. 2. Values of  $^{137}$ Cs content (Bq/m<sup>3</sup>) in the water of the rivers in May-July 1986 relative to the background level and the AC (acceptable concentration).

--- Level of the content of <sup>137</sup>Cs in the mater of the rivers in 1983-198:

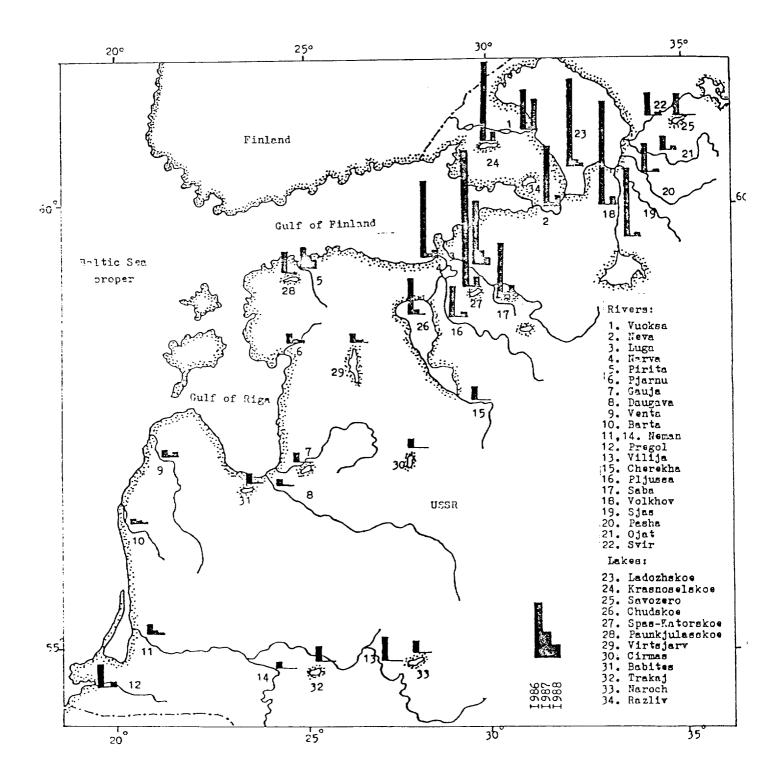
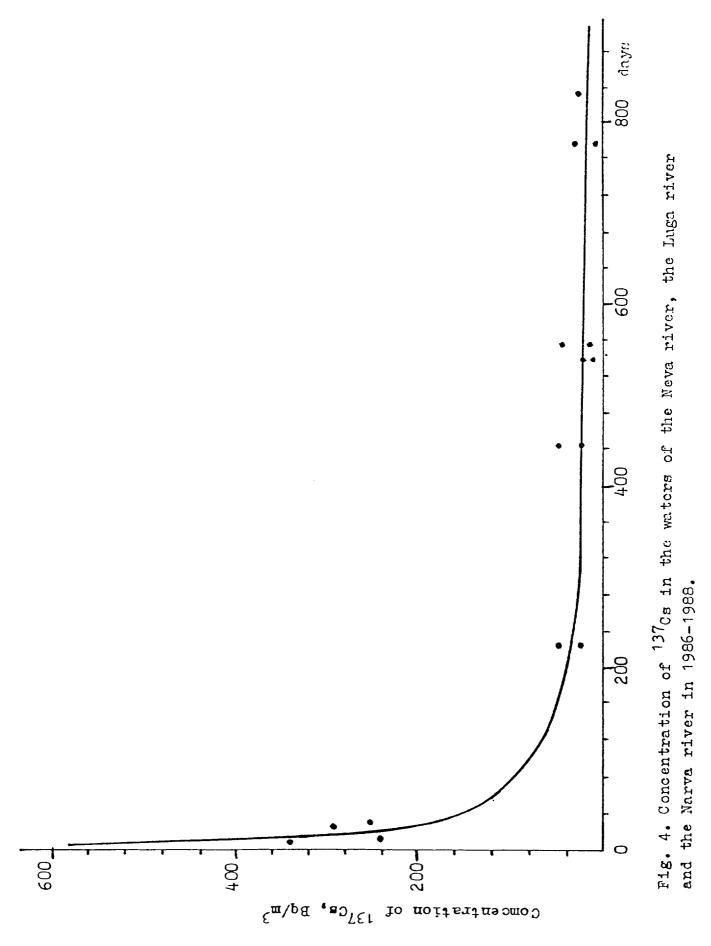
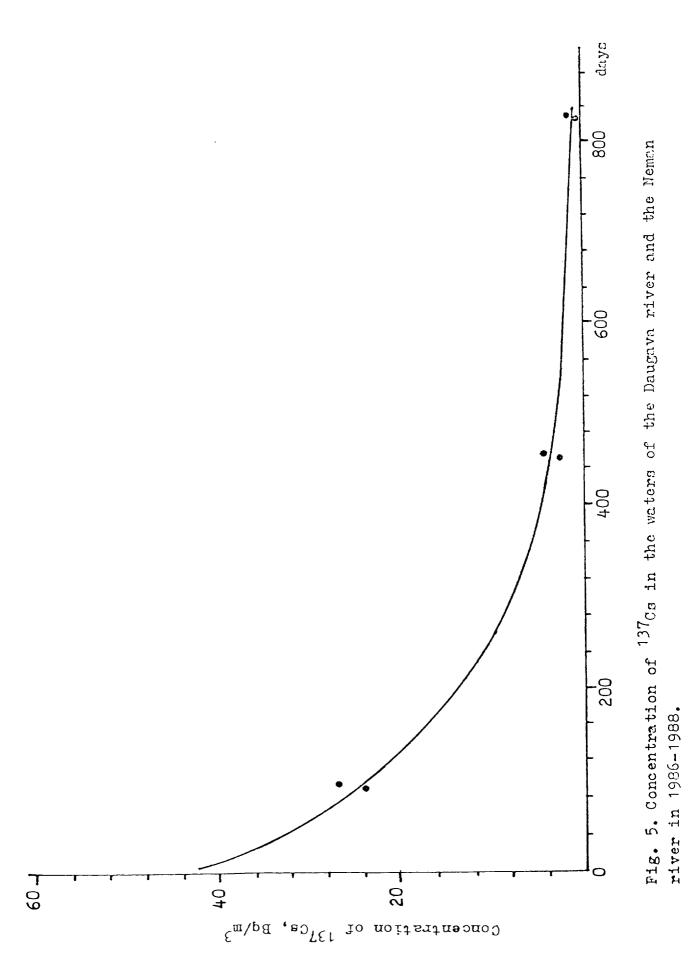


Fig. 3. The distribution of  $^{137}$ Cs in the waters of the lakes and rivers of the Baltic Sea region of the USSR in 1986-1988.





REVIEW OF THE FINNISH STUDIES ON RADIOACTIVE SUBSTANCES IN THE BALTIC SEA IN 1986-1988

Erkki Ilus, Tarja K. Ikäheimonen, Ritva Saxén, Kirsti-Liisa Sjöblom, Seppo Klemola and Hannele Aaltonen

Finnish Centre for Radiation and Nuclear Safety

### 1 INTRODUCTION

Studies on radioactive substances in the Baltic Sea have been carried out in Finland since the early 1960s. At that time the focus of the studies was on radionuclides originating from atmospheric nuclear arms testing conducted in the northern hemisphere. Until 1986 the concentrations of long-lived radionuclides had steadily decreased in the marine environment, but the serious accident in the nuclear power station at Chernobyl on April 26, 1986 provided a new impulse for radioecological studies in the Baltic Sea. The results of the Finnish studies carried out in the Baltic after the Chernobyl accident are published in six Supplements of the Annual Reports 1986, 1987 and 1988 of the Finnish Centre for Radiation and Nuclear Safety.<sup>1-6</sup> This article reviews the results of these studies. Special attention is paid to the horizontal distribution of fallout nuclides and to their sinking and sedimenting rate in the Baltic Sea.

#### 2 HORIZONTAL DISTRIBUTION

The uneven distribution was a specific feature of the Chernobyl fallout as compared with the fallout caused by nuclear arms tests in the 1960s. Another characteristic was the high proportion of the cesium isotopes  $^{137}$ Cs and  $^{134}$ Cs in the deposition. This

review is mainly concerned with cesium concentrations, but complete data on all radionuclides detected in the samples are given in the original reports.<sup>1-6</sup>

The areal differences in the amounts of deposition were significant at least in the land areas of Finland. The differences correlated well with the amounts of rainfall during the first days of the fallout situation. Considerable differences were also found in the radionuclide concentrations of sea water samples taken later in summer and autumn from different parts of the Baltic Sea (Fig. 2). Nevertheless, it is difficult to estimate the total amount of fallout directly deposited on the Baltic Sea surface. The impact of rainfall was probably less significant at sea than on land, because spring-time rains fall more often on land mass than on the sea.

After the fallout situation, several factors affected the concentrations of radionuclides in the surface layers of sea water, a.o. currents in the sea, mixing of water masses, river inflow from the surrounding land areas, sinking in the water column and deposition into bottom sediments.

The effect of river discharges and currents are well demonstrated in the  $^{137}$ Cs concentrations of <u>Fucus vesiculosus</u> collected along the Finnish coast in 1987.<sup>2</sup> The lowest values were generally found in coastal areas with low deposition values (Fig. 4). The highest concentrations, which were found in the eastern Gulf of Finland and in the middle part of the west coast obviously indicate the impact of river discharges from inland areas with higher deposition values.<sup>2</sup> The relatively high concentrations of  $^{137}$ Cs found on the north and west coast of Åland, where the deposition values were in general very low, were probably caused by cesium transported by sea currents from the Gävle region, which was the main fallout area in Sweden.

The changes in the <sup>137</sup>Cs concentrations of surface water in different parts of the Baltic Sea since 1986 reflect the transport southwards from the Gulf of Bothnia and Gulf of Finland of fallout nuclides within the outflowing fresh surface waters (Fig. 2). In 1988, the concentrations of <sup>137</sup>Cs in the open waters of the Gulf of Finland and in the middle of the Bothnian Sea were about 20% and 55% of the corresponding maximum values detected in 1986. On the other hand, there was a clear increase in the concentrations found in the the Baltic Proper and in the Bothnian Bay, the latter being caused by retarded river discharges and the anticlockwise water currents in the Gulf of Bothnia.

Soon after the fallout situation, there was a marked difference between the concentrations found in the coastal and offshore areas, with the concentrations of radionuclides being highest near the coast. This was probably due to hydrological factors, such as quantity of rain and runoff, but also to the limited water exchange in the labyrinthine archipelago zone. Afterwards, there has been a general levelling of areal concentration differences, at least in sea water. However, considerable amounts of fallout nuclides have remained in the coastal zones due to accumulation in sediments and biota (Table I).

# 3 SINKING AND SEDIMENTATION RATE

The sinking of the fresh fallout through water layers was quite rapid soon after the Chernobyl accident. As early as the afternoon of April 28, considerable amounts of fallout nuclides had accumulated in <u>Fucus vesiculosus</u> growing at a depth of 2 m in the Olkiluoto area.<sup>4</sup> Also at open sea, the sinking rate was high owing to the concurrence of the end phase of the phytoplankton spring maximum, when radionuclides were transported downwards by dead plankton algae.

In the phytoplankton samples taken from the Loviisa area in 1986, the highest concentration of  $^{137}$ Cs (2500 Bq kg<sup>-1</sup> dry weight) was registered at the beginning of June.<sup>4</sup> Towards the autumn, the values were considerably lower (Fig. 3). The acute concentration factor for  $^{137}$ Cs from water to phytoplankton was approximately  $10^3$ .

According to our results, fresh fallout nuclides were found during the first half of May in water samples taken from a depth of 100 m in the southern Baltic Proper and in mid June from samples of surface sediment of the northern Baltic Proper (station Teili-1, depth 166 m). Since then, the primary halocline (at a depth of 50-70 m) may have limited the addition of fallout nuclides in the deep water layers of the Baltic Proper, while in the Bothnian Bay and Bothnian Sea, where no clear halocline is met, an effective mixing of water masses has been possible, and no marked difference was found between the surface water and near-bottom water in 1988.

It has been difficult to obtain an accurate picture of the sedimentation rate of radionuclides soon after the fallout situation, because the sedimentation process is very slow at many offshore stations (some millimetres per year or less) and there are many problems involved in sampling the uppermost loose sediment layers.<sup>5</sup> It has been possible, however, to follow up the sedimentation of fresh fallout by means of 1-cm-thick slices taken with the Niemistö corer in the coastal areas off Loviisa and Olkiluoto and at the Teili-1 station in the northern Baltic Proper, where the sedimentation rate is quite high (Figs. 5 and 6). In 1988, Chernobyl-originated <sup>137</sup>Cs was detected in the two uppermost centimetres of sediment at all these stations. in the amounts of long-lived radionuclides The distinct peaks provide useful pointers for sedimentation studies. At the Teili-1 station, the highest value of <sup>239,240</sup>Pu was in the 5-10 cm layer (Fig. 7), thus indicating the peak caused by nuclear arms testing conducted in the 1960s.

4

# INTERNAL RADIATION DOSE CAUSED BY THE CONSUMPTION OF BALTIC SEA FISH IN FINLAND

In comparison with concentrations in fresh water fish from Finnish lakes, the activity concentrations of  $^{137}$ Cs have been low in brackish water fish caught off the Finnish coast. The highest value (230 Bq kg<sup>-1</sup> fresh weight) was detected in perch caught in the Loviisa area in late summer 1986.<sup>3</sup> During the last two years, the concentrations in most species, have remained unchanged or have begun to decrese, but have continued to slowly increase in some predatory fish, such as pike and cod.<sup>5</sup> In 1988, the <sup>137</sup>Cs concentrations of pike were about 10-40 times higher than before the Chernobyl accident.

Figure 8 shows annual changes of 137Cs concentrations in four fish species at Loviisa, where the concentrations found in pike also began to decrease in autumn 1988. In Baltic herring, there has been a clear decrease of cesium values in winter, but a new increase has occurred in summer. This is probably associated with the seasonal migrations of Baltic herring.<sup>6</sup> In general, the species moves for the winter to deeper offshore waters, where the activity concentrations of 137Cs are lower.

Since the Chernobyl accident, the average intake of <sup>137</sup>Cs and the internal radiation dose caused by the consumption of Baltic Sea fish by the Finnish population have been as follows:

Year	Average intake	Internal dose
	of <sup>137</sup> Cs	( <sup>137</sup> Cs+ <sup>134</sup> Cs)
	Bq a <sup>-1</sup>	mSv a <sup>-1</sup>
1986	220	0.005
1987	330	0.008
1988	460	0.009

The estimate is based on an average per capita consumption of 7.7 kg of Baltic herring and 5.8 kg of other sea fish per year. Both cesium isotopes have been taken into account in the dose calculations.

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cer (Bg m <sup>-3</sup> ) May-June July-	GULF OF FINLAND Loviisa area 3000-5200 410-500 250-440 180-260	ND Offshore stations 640 230 120	BOTHNIAN SEA 01kiluoto area 1000-1100 260-370 230-400 220-280	Offshore stations 580 420 320
Sediment (Bq m <sup>-2</sup> )				
May-June July-	11000-18000 - 27000-35000	- 860- 3700 4300-22000 12000-13000	- - 9600-24000	- 5600 14000 16000
vesiculosus (Bq kg <sup>-1</sup> dr	Y wt.)			
May-June July-	2700-4900 700-1100 400-770 270-450	- - 370-410 210-340	700-2000 100-300 170-410 140-280	- - 150-160 160-180

Coastal-offshore comparison of  $^{137}$ Cs levels in the Gulf of Finland and Bothnian Table I.

a no samples

-87-

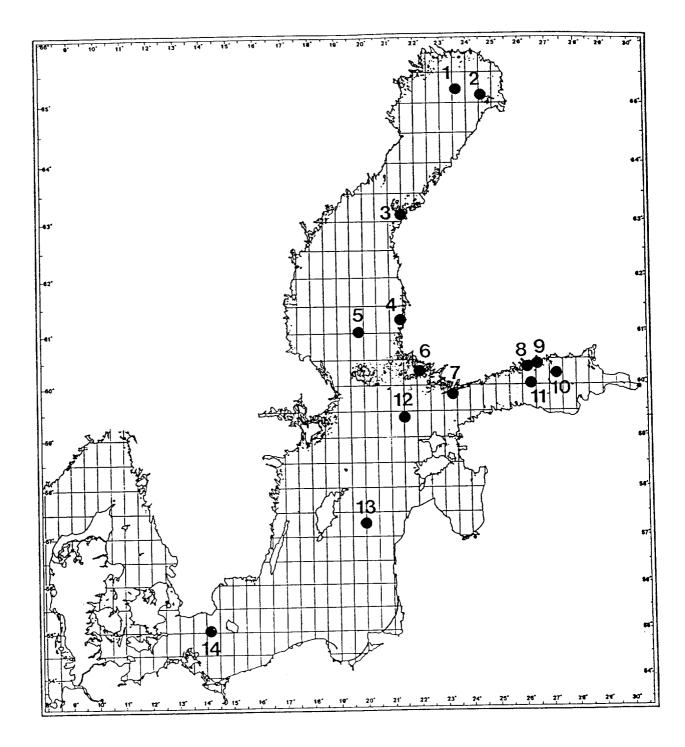


Fig. 1. The Finnish sampling stations for sea water (W), bottom sediment (S) and fish (F) in the Baltic Sea. 1. Station C VI (W+S), 2. Hailuoto (F), 3. Vaasa (F), 4. Station Olkiluoto 2 (W), 5. Station EB 1 (W+S), 6. Seili (F), 7. Tvärminne (F), 8. Station Pernaja R1 (W), 9. Station Loviisa 3 (W), 10. Station XV 1 (S), 11. Station LL 3a (W+S), 12. Station Teili-1 (W+S), 13. Station BY 15 (W+S), 14. Station BY 2 (W).

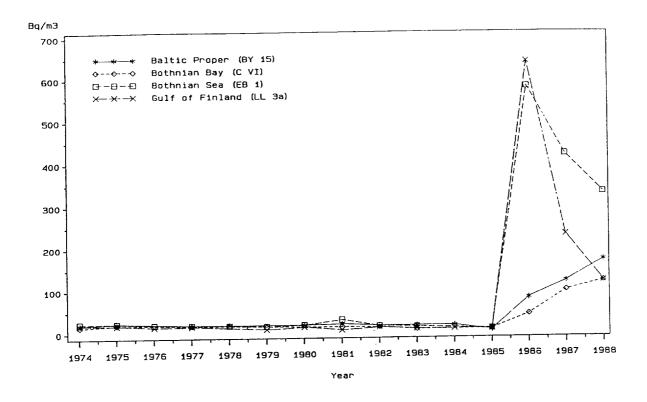


Fig. 2. Cesium-137 concentrations (Bq m<sup>-3</sup>) in sea water at four sampling stations in the Baltic Sea in 1974-1988.

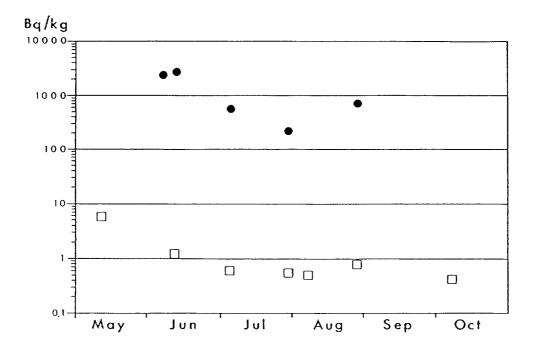


Fig. 3. Cesium-137 in phytoplankton (●) and sea water (□) at Loviisa in 1986.

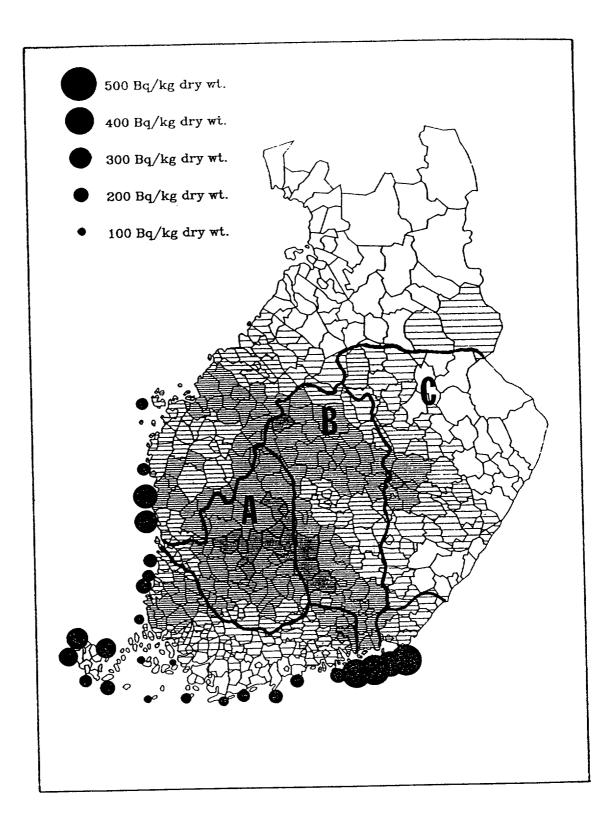


Fig. 4. Distribution of cesium-137 deposition in Finland and the cesium-137 concentrations in <u>Fucus vesiculosus</u> collected along the Finnish coast in 1987. The darkest areas received the most fallout. A, B and C are drainage areas of the three largest rivers Kokemäenjoki, Kymijoki and Vuoksi.

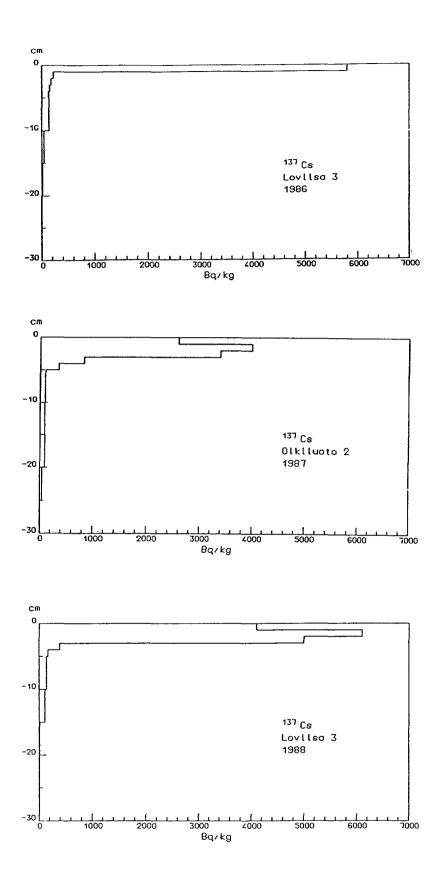


Fig. 5. Vertical distribution of cesium-137 (Bq kg<sup>-1</sup> dry wt.) in sediment samples taken at the stations Loviisa 3 and Olkiluoto 2 in 1986-1988.

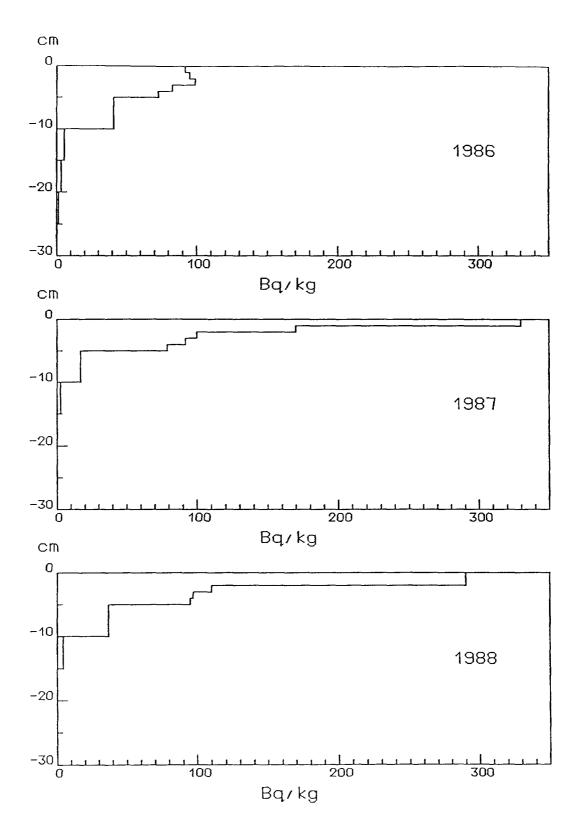


Fig. 6. Vertical distribution of cesium-137 (Bq kg<sup>-1</sup> dry wt.) in sediment samples taken at the Teili-1 station in 1986-1988.

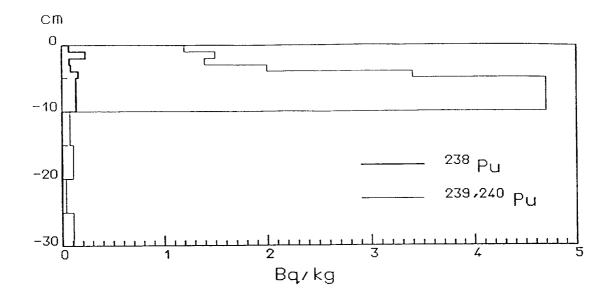


Fig. 7. Vertical distribution of plutonium-238 and plutonium-239,240 (Bq kg<sup>-1</sup> dry wt.) in sediment samples taken at the Teili-1 station in 1988.

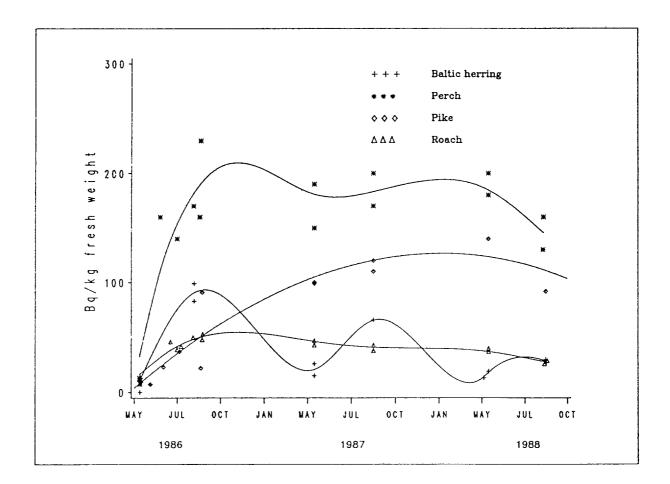


Fig. 8. Annual changes of cesium-137 concentrations (Bq kg<sup>-1</sup> fresh wt.) in edible parts of four fish species at Loviisa in 1986-1988.

THE DISTRIBUTION OF RADIONUCLIDES IN BOTTOM SEDIMENTS OF THE OPEN BALTIC SEA AND THE GREIFSWALD BODDEN

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#### ABSTRACT

Monitoring results of the radioactive contamination of sediments of the open Baltic Sea and the Greifswald Bodden, obtained in 1986 and 1987, are discussed and compared with results of previous years. The changes in sediment contamination and the vertical migration of contaminants are described for different types of sediments. A comparison between the sedimentation rate determined by means of the lead-210 dating method and the depth of penetration of artificial radionuclides since 1986 indicates that different factors have influenced the vertical migration of the deposited radionuclides. It seems that the diffusion is interfered by bioturbation and near bottom currents. In addition, any uncertainties during sediment sampling can also influence the accuracy of the measuring result.

### 1. INTRODUCTION

Investigations of sedimentation in seas, their spatial distribution and temporal changes provide basic information for estimating the mass balance of substances. Bottom sediments and suspended materials play an important role within the framework of radioecological investigations in the aquatic environment. They can act simultaneously as a source and a sink of radioactive substances. The high variability of the structure, the changing chemical behaviour and the abundance and activity of sediment dwelling organisms create a widespread inhomogeneity of the bottom sediments. Therefore it is difficult to make general assessments on sediment accumulation, fixation and remobilization of radionuclides. In addition , the techniques of sampling and treatment of sediment samples can evidently influence the accuracy of the measuring result.

Investigations of the content of artificial and natural radionuclides in bottom sediments of the Baltic Sea and of GDR coastal waters are an essential part of the radiological monitoring programme of the Baltic Sea carried out by the National Board for Atomic Safety and Radiation Protection of the GDR. In the past the investigations of sediments were focused on the measurement of the artificial radionuclides Sr-90 and Cs-137 and the natural radionuclides K-40, Pb-210 and Ra-226. Due to the radioactivity input into the Baltic Sea in the event of the Chernobyl accident, other radionuclides, especially Cs-134, became also important for sediment investigations. The isotope Cs-134 which previously either had not been detected at all or only in traces in Baltic sediments provided a peak-shaped input function. The temporal change of this input signal along a sediment core is well suited to inform of processes of distribution and mixing in sediments.

#### 2. METHODS

The sediment samples were taken by means of a Niemistoe-corer (diameter 5.5 cm, 6.0 cm or 8.0 cm), fractionated in slices of 1, 2 or 3 cm and than deepfrozen. Before measuring the samples were dried at 110°C or freeze-dried to a constant dry weight. The radioactivity of gamma-emitting radionuclides was measured by a multichannel analyzer. For sediment dating 2 to 5 g of dry matter were taken from selected samples, decomposed with HF and after precipitation on silver plates the radioactivity of the daughter nuclide Po-210 was analyzed by means of an alpha spectrometer. Sediment samples with a Cs-137 radioactivity below the gammaspectrometric detection limit and/or samples for Sr-90 analysis were ashed at 440°C, decomposed with HF, spiked with stable Sr and Cs and then a precipitation with carbonate was carried out. The solution contained the Cs-137 and the precipitate contained the Sr-90. The radioactivity of both radionuclides (for Sr-90 the daughter nuclide Y-90 was used and the Cs-137 radioactivity was precipitated as hexachloroplatinate) was determined by low level beta anticoincidence measurement.

## 3. RESULTS AND DISCUSSION

#### 3.1. Sediment dating by the lead-210 method

For dating of sediments aged less than 200 years the Pb-210 method seems to be the most common procedure. Based on the work of (GO61) this method became more advanced and was successfully used for studying sedimentation processes in marine and freshwater environs. The most advanced methods allow detailed investigations of processes of sediment mixing. The method used in this work is the c.r.s. method (PH79,CH87,GE89) which is also suited to interpret increases of the specific radioactivity of Pb-210 vs. mass depth. This phenomenon is visible as a snap-off or an increase of the Pb- 210 concentrations vs. sediment depth and has often been found in sediment samples of the Baltic Sea and the Greifswald Bodden.

For the calculation of the average age of a sediment slice the following equation can be used:

$$\gamma_{i} (M_{i}) = \frac{1}{\lambda} l_{n} \frac{A \omega}{A \omega - A_{i}}$$
(1)

where

$$A_{i} = \sum_{j=1}^{i} g_{j} a_{j} \Delta z_{j} \qquad (mBq/cm^{2})$$

The value of  $A \infty$  can be obtained by

- measuring results of the Pb-210 concentrations
- using the regression line of the c.i.c. method
- calculating on the flux of Pb-210 (if known).

The sedimentation rate (S) can be calculated either by differentials

$$S_{i} = \frac{M_{i+1} - M_{i-1}}{\gamma_{i+1} - \gamma_{i-1}}$$
(2)

or integrals by means of a regression analysis

$$\tau_i = \frac{1}{S} \cdot M_i + \gamma^* \tag{3}$$

where

$$M_{i} = \sum_{j=1}^{i} \varphi_{j} \Delta z_{j} \qquad (g/cm^{2})$$

The content of supported Pb-210 was determined either from the Ra-226 concentration or extrapolated from the profile of the Pb-210 concentration for  $z \rightarrow \infty$ .

The results of sediment dating of samples of the Baltic Sea and the Greifswald Bodden are summarized in Tab.I. A detailed interpretation of the results is given in (GE89). As the results show sediment mixing can be observed in a couple of samples. The sediment mixing results from bioturbation or mechanical sediment particle transport. At the sampling station GDR-023 (Bay of Mecklenburg) an increase of the sedimentation rate could also be assumed because the Peclet number of 1.4 indicates a predominantly advective transport.

# 3.2 The distribution of artificial radionuclides in sediments of the Baltic Sea before 1986

At the time before the radioactivity input from Chernobyl only the artificial radionuclides Sr-90, Cs-137 and some transurations have been detected in sediments which originated mainly from the global fallout of nuclear weapon tests. Figure 1 shows four profiles of the Sr-90 and Cs-137 concentrations in Baltic sediments. Due to the temporal changes of the concentration of these

radionuclides in water the maximum concentration measured in 1963-1965 should also be built up in sediments, if these radionuclides are strongly fixed at bottom sediments. This is not valid for Sr-90 and, as regards Cs-137, only one sample taken from the Arkona basin in 1984 showed a cocentration peak in the sediment layer between 10 and 19 cm (Fig.2). From the missing peak concentration in the other sediment samples it can be concluded that cesium is either more mobile in sediments than commonly assumed or that sediment mixing took place. The results of Pb-210 dating hint at the last interpretation.

For the determination of the cumulative deposition of the Cs-137 radioactivity at the Baltic Sea bottom, the Cs-137 concentration per unit area was calculated by summing up the concentration of each slice of a sediment core sample. These results are depicted in Figs.3a and 3b. As already indicated by the results of the determination of the sedimentation rates (s. Tab.I), the sediment accumulation rate seems to decline with increasing water depth. Consequently the deposition of Cs-137 at the sea bottom should also be inversely correlated to the water depth. A correlation analysis of the cumulative deposition of Cs-137 vs. water depth confirms this relationship (s. Fig.4) which can be described for the sedimentation period until 1985 by the following regression function:

 $y = 2,336 \cdot exp(-0.0072x)$  (r = 0.963)

where

У		cumulative	deposition	Οf	Cs-137	(Bq∕m²)
Х	==	water dept	h			(m)

This empirically derived regression function is valid for soft sediments of the Baltic Sea except for coastal waters and estuaries. Using this function it is possible to estimate the total storage of Cs-137 in the bottom sediments of the Baltic Sea before the new input of radioactivity in 1986. For this purpose the bottom areas corresponding with the zones of water depth were calculated from a bathymetric map (BS81). The percentages of soft bottom area were taken from (SA86) and set in order to each depth zone. Since a certain kind of hard bottom consisting of fine and medium-grained sands also involves small amounts of silt, clay or mud, the value of Cs-137 contamination was estimated to be about 10 % of the corresponding value of soft bottom. The results are summarized in Tab.II. Accordingly, the total storage of Cs-137 in the bottom sediments of the Baltic Sea amounted to 284 TBq in 1985. The Cs-137 deposition calculated by (SA86) was 277 TBc based on measured concentrations in sediment samples. Taking into account the difference between measured and calculated Cs-137 inventory of the watermass, and the soft bottom area only, the sediment-bound Cs-137 inventory of the Baltic Sea was estimated by (SA86) to be 323 TBq.

# 3.3 The distribution of artificial radionuclides in sediments of the Baltic Sea from 1986 to 1988

During the years 1986 to 1988, 11 sediment core samples from the open Baltic Sea and 4 core samples from the Greifswald Bodden were analyzed. The results are illustrated in Figs. 5a and 5b and summarized in Tab. III. It follows that the deposition of Cs-134 and Cs-137 in the event of the Chernobyl accident depends not only from the mentioned relationship between their concentration in sediments and the water depth but also from the contamination level of the water and the strength of the discontinuity layer. Therefore the highest values of sediment contamination with radiocesium can be observed in the sea areas in which the initial contamination of the water body in 1986 was highest, namely the Bothnian Sea and the Gulf of Finland. In contrast, the sediment contamination in the strongly stratified areas of the Baltic Froper was very low even in 1987. In the deepest parts of the Baltic Proper no or only traces of Cs-134 were detected (s. Fig. 5a and Tab. III).

In sediment samples taken at locations with high deposition rates of radiocesium also other Chernobyl burden radionuclides such as Co-60, Ru-106 and Ag-110m were detected. In sediment samples taken from the Greifswald Bodden Cs-134, Cs-137 and Ru-106 were found in measurable amounts. The Sr-90 radioactivity resulted from previous deposits.

Due to the very inhomogeneous level of the initial contamination of the Baltic Sea area and due to other factors influencing the radionuclide sedimentation an approach to calculating the total storage of radiocesium in the sediments, caused by the Chernobyl accident, in the same way described for deposits before 1986 is impossible. There exists no correlation between the radiocesium content in sediments and the water depth. Therefore a theoretical approach was taken into account according to (EV80) which calculates the percentage of the loss of radioactivity from the water body by sedimentation as follows:

$$K_{w,w} = \frac{K_{cl} \cdot S}{d_{w} (1 + K_{cl} \cdot SM)}$$
 (4)

where

Kw.s		fractional loss of radioactivity from water to sediment	(1/yr)
Ka	=	sediment distribution coefficient	(⊏m³/g)
S	=	sedimentation rate	(g/cm².yr)
SM	=	amount of suspended matter	(g/⊂m <sup>35</sup> )

The relationship of the  $K_{d}$  value of cesium vs. salinity of the ambient water can be expressed by:

(cm)

$$K_{e}(Cs) = 30,000 \cdot exp(-0.117 \cdot (%o))$$
 (5)

The parameters used for the calculation of the Cs-137 deposition at bottom sediments according to eq.(4) are listed in Tab.IV. The sedimentation rate (S) is conservatively considered as a constant value of  $0.05 \text{ g/cm}^2$ .yr (besides the inverse correlation to the water depth) and corresponds with the data given in Tab.I. The amount of suspended matter was assumed to be 0.001 g/l.

For the calculation of the Cs-137 deposition at the sea bottom it follows from eq.(4):

$$\overline{C}_{\infty} = \frac{\overline{C}_{\omega} \cdot \nabla \cdot K_{\omega,\infty}}{A}$$
(6)

where

Ē.	=	average deposition of Cs-137 radioactivity per year	(Bq∕m².yr)
Ē.	=	average concentration of Cs-137 in the ambient water	(Bq∕m <sup>™</sup> )
V	==	volume of the watermass	( m = )
A	=:	area of soft bottom	(m²)

The results compared with data measured in 1987 are listed in Tab.V. They demonstrate a good agreement between the calculated values and the measured ones of the Cs-137 deposition at the bottom of each Baltic Sea subarea. The total storage of Cs-137 on the Baltic Sea floor resulting in August 1987 from the fallout of 1936 was estimates to be 584 TBq. The value of 856 TBq given by (DA89) is based on the total area of the sea bottom without distinguishing hard bottom areas from soft ones.

# 3.4 The distribution of Cs-134 and Cs-137 along sediment core profiles

The vertical distribution of radiocesium concentrations among sediment cores can be seen in Figs. 6a to 6d. Fig.7 illustrates the temporal changes of the radiocesium concentration along the sediment cores taken from the Greifswald Bodden (station-"Ariadne") in 1985, 1986 and 1988. It is remarkable that the depth of penetration of Cs-134 concentration into sediments due

to the Chernobyl fallout widely varies from one to another location and sediment type. Assuming that cesium is immobile in bottom sediments and sediments are not mixed then the vertical penetration would result only from the present sedimentation rate the vertical sediment diffusion coefficient  $D_{x}$ and  $(D_z(Cs) =$ 1.1 E-08 to 3.0 E-09  $cm^2/s$ ). Consequently the total amount of Cs-134 should be found in the top layer of the sediment sample only. But this is not true for most of the analyzed samples. The patterns of vertical distribution of the Cs-134 and Cs-137 concentration in sediments indicate more or less extended disturbances due to different processes of sediment mixing.

#### 3.4.1 Bioturbation

The problem of sediment mixing and translocation of sediment particles due to the biological activity of sediment dwelling organisms is well known, but the effectivity and the velocity of this process differ widely. This depends, e.g., on the kind of the organisms, the abundance of the species, the depth of penetration of the organisms into the sediment, and the feeding habits.

well dwellt with benthic animals were found at the Sediments stations 11 (Kattegat), 37 (Aland Sea) and 50 (Bay of Bothnia). While the radionuclide Cs-134 at the Kattegat station has only been detected in the top layer (0 - 3 cm) it penetrates into the sediment up to a depth of 12 and 20 cm resp. at the other two stations (Fig.6). This penetration depth well agrees with the length of the bore holes of any kind of polychaetes and mussels. In contrast, the sediment from the Kattegat was dwellt mainly superficially. If it is assumed that the vertical penetration of radiocesium results from bioturbation only, this process has to be considered as very effective for sediment mixing. Within a one year period already 30% (Station 37) and 26% (Station 50) resp. of the deposited Cs-137 radioactivity was shifted from the surface layer (0 - 2 cm) to deeper sediment layers by bioturbation. Taking further into account that a growth rate of the sediment of 0.2 cm per year can be expected on the basis of a sedimentation rate of 29 mg/cm<sup>2</sup>.yr (Pb-210 dating, Station Bo3A, Bothnian Sea) and a dry mass of the top layer (0 - 1 cm) of  $0.1 \text{ q/cm}^2$ , the high importance of bioturbation in sediment mixing is obvious. The mixing in the top layer (0 - 2 cm) of the sediment core taken from the Bay of Bothnia should result from the biological activity of Mesitodea entomon, the most abundant macrobenthic animal of this area, while the mixing in deeper layers should be primarily effected by Macoma baltica and polychaetes.

In the sediment samples which were anoxic (redox layer within the surface sediment layer, at the sediment-water interface or in the water column) the penetration depth of Cs-134 was lower than in the above-mentioned well aerated sediments. This concerns the stations 25 (Bay of Gdansk), 96 (Arkona basin) and 101 (Bay of Luebeck). While the muddy sediments of the Arkona basin and the Bay of Luebeck are aerated at least within the top layer the existence of benthic animals can be assumed. Therefore, the penetration of Cs-134 into the sediment up to 2 or 3 cm could also result from bioturbation, but other processes affecting that cannot be excluded. In contrast, in the Bay of Gdansk, the redox layer is already within the water column and the bioturbation cannot act as a process of sediment mixing.

#### 3.4.2 Near bottom currents

The second process which can become important in sediment mixing is the near bottom current. Due to the small water depth and the current characteristics of both the Arkona basin and the Bay of Luebeck the vertical penetration of radiocesium can also be caused by this process in addition to bioturbation. According to (FU84) computer simulations of the current field of different depth zones of the Baltic Sea indicate a current velocity of appr. 2 to 3 cm/s in the near bottom water of the Bay of Luebeck and of the Arkona basin. These velocities are great enough to transport fine-grained sediment particles (silt, clay, mud) according to the Hjulstroem diagramme.

The sediment samples taken from the Greifswald Bodden at the same station successively in 1985, 1986 and 1988 indicate a very rapid and progressive mixing up to a sediment depth of 8 cm in 1988. The water depth at this location is about 10 m and full mixing of water by a wind-induced current takes place under special meteorological conditions. As the redox layer is located within the top sediment layer, the translocation of fine-grained materials should primarily be the result of near bottom currents. The bioturbation and the sedimentation of new settling material is of minor importance or neglegible.

### 3.4.3 Other factors

already mentioned above, a high sedimentation rate or a As sedimentation rate changing with time can also create irregularities in pollutant distribution patterns along a sediment core. This is true, e.g., if rivers transport sediment particles to coastal waters. The sediment load of the river water depends primarily on the flow rate and the run-off amount of the river. Fine-grained particles can be transported over great distances from the river mouth on and settle down at the sea bottom. This phenomena occurs in the Bay of Gdansk. At station 25 (Bay of Edansk) where the redox layer is within the water column and the near bottom current is very low the idendified Cs-134 radioactiwithin the surface sediment layer (0 - 1 cm and 1 - 2 cm)vity. could be of riverine origin. This assumption can also be supported by the calculated sedimentation rate of 90 mg/cm<sup>2</sup>.yr (Pb-210 method) in connection with a very low mass depth of the sediment top layer of 0.046 g/cm<sup>2</sup>. Also for station 101 (Bay of Luebeck) a recently higher sedimentation rate than some decades ago can be considered. because this phenomena was found in the nearby Mecklenburg Bay which has similar hydrological conditions.

Another factor influencing the vertical distribution patterns of a pollutant like Cs-134 is given by a partial disturbance of the sediment core during the sampling and fractionating proce-

dures. For example, at station 25 (Bay of Gdansk), one sample was taken with a Niemistoe corer and cut into slices of 1 cm thickness each but another one was taken with a box corer from which subsamples were taken with plastic tubes and fractionatad in slices of a thickness of 3 cm each. While the two uppermost slices of the first sample contained Cs-134 this radionuclide was not detected in the second sample (Fig.8). Using a Niemistoe corer the top of the core can be disturbed due to the very low density of this layer resulting in an apparent penetration of Cs-134 up to a sediment depth of 2 cm. On the other hand, by taking a sample with the box corer the top layer which contains the Cs-134, the radioactivity can be lost even before the subsamples were taken with the plastic tubes. In this case Cs-134 is not detectable. In.addition, if the radioactivity is deposited only within a few mm of the top slice, the greatest part of this slice contains no Cs-134 and this material which is not contaminated "dilutes" the Cs-134 radioactivity to such an extent that it cannot be detected. Therefore, the procedures of taking and pretreating sediment core samples have to be carried out carefully and information on the type of sampler must be given.

In contrast to the results of Cs-134 measurements of the above-mentioned sediment samples, the sediment cores taken at stations 33 (near Landsort depth) and 82 (Gothland depth) did not contain any Cs-134 radioactivity. These stations are located in the deepest parts of the Baltic Proper which are characterized by a permanent discontinuity layer within the water column and a very low sedimentation rate (10 - 15 mg/cm².yr). As regards the vertical distribution of Cs-134 concentration along the water column only trace amounts of this radionuclide (1 - 5 Bq/m²) were detected in the near bottom water in 1987. Consequently, no or only traces of Cs-134 settled town to the bottom in the period between the deposition at the sea surface in May 1986 and the date of sediment sampling (August 1987).

#### 4. CONCLUSIONS

- The soft sediments of the Baltic Sea act as an effective sink of radioactive substances. Like the nutrient cycle proceeding in the strongly stratified areas under anoxic conditions, the remobilization of sediment-bound radionuclides and their re-introduction into biological cycles is almost inhibited.
- The Baltic sediments can only be a source of radioactivity if they are oxic and dwellt by organisms, if the discontinuity layer of the water is only slightly or not developed or if the water current at the sea bottom is high enough to transport sediment particles.
- The results of sediment dating and the distribution patterns of the radiocesium contamination along sediment core profiles show that the upper part of the sediment is often exposed to more or less intensive sediment mixing proceses. The sediment mixing originate primarily from bioturbation and near bottom currents. In addition, high or fluctuating sedimentation rates

and disturbances of the sediment during sampling and pretreatment can influence the distribution patterns of pollutants.

- The Pb-210 method using the c.r.s. model in connection with the c.i.c. model seems to be the most appropriate method of dating sediments of an age of less than 200 years. The use of Cs-134 or Cs-137 for sediment dating is 1 imited because no constant flux of radiocesium can be assumed or becaus the peak of the Cs-137 concentration resulting from the high fallout deposition during 1963 1965 occurs in the sediment often indistinctly or vanished. Furthermore a recently higher or a changing sediment accumulation rate cannot be identified and due to mixing of sediment material at ... earlier time the peak concentration apparently occurs in a deeper sediment layer.
- The applicability of sediment dating procedures for a better unterstanding of the sediment chronology could be improved if it could be possible to achieve a quantification of the errors made during sediment sampling, fractionating and pretreatment and to compare quantitatively different types of sediment samplers. For this purpose further intercomparison exercises using different types of samplers have to be initiated.

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Station	B0-3A 64 <sup>0</sup> 18.3'N 22 <sup>°</sup> 21.5'E	Bay of Mecklenburg	<b>BY15</b> (GDR-271) GDR-113		GDR-113
Coordinates		54 <sup>0</sup> 10.0'N 11 <sup>0</sup> 24.0'E	57 <sup>0</sup> 18,3'N 20 <sup>0</sup> 04,6'E	54 <sup>0</sup> 55.5'N 13 <sup>0</sup> 30.0'E	
Date of sampling	1981-1X-27	1981-VII-15	1983-VII-3	1983-VII-5	1984-XI-19
Pb-210 concentration at <b>the s</b> ediment surface (mBq.g_d.w.)	348-42	135-16	350 <b>±</b> 56	239+26	219 <sup>+</sup> 26
mean Ra-226 concentra- tion '(mBq.g <sup>-d</sup> .w.)	44 <sup>±</sup> 12	24±3	108 <b>±</b> 32	23 <b>±</b> 4	31-5
Flux of Bb-210 (mBq.cm .a )	8.9	13.3	3.4	19.9	18.4
Mixing depth <b>(Z<sub>K</sub>)</b> (cm)	2.0	7.5	2.0	10.0	16.0
Mass depth at Z <sub>K</sub> (g.cm <sup>-2</sup> )	0.2	2.6	0.1	2.4	3.5
<u>C.R.Smethod</u> Sedimentation <b>rate(S<sub>1</sub>)</b>					
(mg.cm <sup>-2</sup> .a <sup>-1</sup> ) Sedimentation rate (S <sub>2</sub> )		68 <b>-</b> 6	14 <b>±1</b>	70-3	125 <b>-</b> 12
(mg.cm <sup>-2</sup> .a <sup>-1</sup> )	22+2	50 <b>±</b> 3		34 <b>±</b> 1	59 <b>-</b> 3

# Table I: Dating results of Baltic sediments with the Pb-210 method

Station	GCR-023	GDR-012	KW II	Ariadne	33
Co-ordinates	54 <sup>0</sup> 03.5'N 11 <sup>0</sup> 03.3'E	54 <sup>0</sup> 18.5'N 11 <sup>0</sup> 33.0'E	54 <sup>0</sup> 11.7'N 13 <sup>0</sup> 34.3'E	54 <sup>0</sup> 12.5'N 13 <sup>0</sup> 34.0'E	58 <sup>0</sup> 44.0'N 18 <sup>0</sup> 30.0'E
Oate of sampling	1984-XI-18	1934-XI-18	1985-X-3	1985-X-3	1987-VIII-11
Pb-210 concentration at the sediment surface (mBq.g <sup>-1</sup> d.w.) mean Ra-226 concen-	128 <b>-</b> 15	130 <sup>±</sup> 15	123 <sup>±</sup> 14	88 <b>-</b> 12	560±62
tration (mBe <sub>lux</sub> of Pb-210)	25±4	26±2	21-2	22±5	69 <b>±</b> 6
flux (mBq.cm <sup>-2</sup> .a <sup>-1</sup> ) of	16.0	13.4	9.6	4.9	21.3
Mixing depth (Z <sub>K</sub> ) (cm)	13.0	4.0	9.0	5.0	
(gssdep)th at Z <sub>K</sub>	4.2	1.8	2.8	1.2	
(gesdepth at Z <sub>K</sub> (q.cm <sup>-2</sup> ) C.R.Smethod Sedimentation rate (S <sub>1</sub> ) (mmg.cm <sup>2</sup> .a <sup>-1</sup> )		81 <b>-</b> 20	86+2	32 <b>-</b> 3	71 <sup>±</sup> 9
Sedimentation rate (S <sub>2</sub> ) (mg.cm <sup>-2</sup> .a <sup>-1</sup> )	50±3	45 <b>±</b> 7	45 <b>±</b> 4	33 <b>±</b> 5	

Table II: Calculated deposition of Cs-137 radioactivity at the Saltic-Sea bottom in dependence on the water depth(before 1986)

bottom area (km<sup>2</sup>) cumulative deposition of Cs-137 (TBq)

Depth zone (m)	B <b>elt</b> soft bo		Baltic soft bot	Proper hard tom	Bay o soft bot	f Riga hard tom	Gulf o: Finland soft bott	d hard	Dothn Sea soft bot	ian hard tom	Bay of Bothnia soft botto	hard	Baltic total soft bot	hard
1	2	• 3	4	5	6	7	8	9	10	31	12	13	14	15
0-20	4,000 8.08	10,00 2.02	0	39,800 8.04	500 1.01	5,000 1.01	1,000 2.02	7,800 1.58	-	20,700 4.18		11,400 2.30		94,700 19.10
20-40	3,000 5.46	4,000 0.73	5,300 9.65	32,600 5.93		4,000 0.73	4,000 7.28	4,000 0.73	1,600 2.91	7,200 1.31	3,000 5.46	6,900 1.26	19,800 36.00	58,700 10.70
40-60			13,300 22.50	20,800 3.39			4,700 7.67	1,200 0.20	5,700 9.29	) 5,600 0.94		1,700 0.28	30,400 49.60	29,500 4.81
GO-80			18,000 25.40	12,100 1.71			5,100 7.19		11,300 15.50	3,200 0.82	4,500 6.34		38,600 54.40	15,330 2.53
80-100			21,000 25.60	5,600 0 <b>.</b> 68			1,200 1.46		8,500 10.40	) 1,100 0.13	4,000 4.38		34,700 42.30	6,700 0.02
103-120			18,000 19.10	2,100 0.22			30 0.03		6,200 6.57	)	1,300 1.38		<i>25</i> ,530 27.10	2,100 0.22
120-150			13,900 12 <b>.</b> 80						5,000 4.60	)	200 0.18		19,100 17.60	
150-200			7,300 5.34						1,500 1.04	C			8,800 6.08	
>200			1,700 0.82						500 0.24	)			2,200 1.06	
sum of <b>Cs-</b> deposition	137 <b>-</b> 13.6	2.S	121.0	20.0	12.2	1.7	25.7	2.5	53.6	7.4	23.3	3.8	246.0	38.2

total: 284.2

Station	Oate	Water	Layer	Water	LOSS on	Sediment	Concent	ration (	Bq.kg	'd.w.)	
		depth (m)	(cm)	content (%)	ignition (%)	type	Cs-134	Cs-137	K-40	Pb-210	Ra-226
1	2	3	4	5	б	7	8	9	10	11	12
11	5.8.	56	0-3	35,38	3.73	clay	1.8	15,2	538		11,5
			3-6	35,38	3,16		-(Cl)	15,7			11.8
			6-9	32.06	3,19			14.5	660		10,6
			9-12	27.44	2.72			12.7	606		12,2
			12-15	26,99	2.46			7.6	523		11,6
			15-18	26.52	2.19			6.0	620		9.0
			18-21	26,22	2,43			3,4	520		9.8
25	7.8.	110	o-1	95,35	27.76	mud	141	516		470	
			1-2	88,68	18.87		56	316	820	457	
			2-3	87,88	18.76		<b>-(</b> <u>≤</u> 26)	136			
			3 - 4	89.86	18.75			134	866	348	23
			4-5	87,87	18.86			222		422	
			5-6	88,60	20.31			144		297	
			6 - 7	86,60	16.67			145	850	344	
			7-8	86,87	16.99			130	740	260	
			8 - 9	84,22	13.93			97.		217	
			9-10	80.91	13.73			60	720	236	
			10-12	80.60	13.23			62	900	182	
			12-14	80,60	13.13			39	960	204	
25	7.8.	110	14-16	76,90	12.37	mud		18	920	IIG	
			16-18	75,30	12.62			11		103	
			18-20	72,59	12.47			-(46)	880	148	32
			20-22	70.82	12.53				950	150	34
			22-24	74,41	12.45				860	160	43
			24-26	73,51	12.45					124	29
			26-28	68.63	13.06				870	91	34
			28-30	68 61	10 27				760	110	

# TableIII:Radionuclide Concentrations in Sediments of the Baltic Sea in 1987

			26-28	68.63	13.06				870	91	34
			28-30	68,61	10.27				760	116	
33	11.8.	319	o-1	93,16	20.16	mud	-(≤6)	96	740	449	67
			1-2	83.80	15,69			48		348	55
			2-3	79.22	9,30			18	695	-321	62
			3-4	75.58	9.35			9.0	942	196	76
			4-5	72.15	8,98			6,1	980	181	57
			5-6	74,93	8,91			3.0	965	168	80
			6-7	71,71	8,49	silt		9.2	980	187	79
			7-8	67.74	7,27			-(f-3)		148	78
			8-9	66,20	7,16			-(≤3)		145	82
			9-10	65.22	6,34			5.0		114	79
			10-11	62.00	5,41			-(≤3)		114	69
33	11.8.	319	11-12	61,63	5.42	silt					58
			12-13	60,05	6.15					101	63
			13-14	63.27	6.60						60
37	12.8.	279	o-2	76.44	9.14	silt	506	1533			70
			2-4	63.04	9.65		140	486			58
			4-6	69.10	8.77		14.3	132			49
			6 – 9	58.78	7.90		7.7	119			59
			9-12	62.37	10.27		5.3	62,2	-		44
			12-15	56.65	8.69		-(≤2)	26.6	-		49
			15-20	65.52				4.0	-		33

## <u>TableIII</u>:(continued)

1	2	3	4	5	6	7	8	9	10	11	12
0	14.8.	106	o-2	64.44	11.54	mud	191	579	760	-	70
			2-4	66.86	10.97		14.6	78.9	880	-	69
			4-6	59.59	8.67	silt	6.3	35.2	850	-	43
			6-9	61.56	8.14		5.6	22.0	920	-	67
			9-12	63.65	8.24		5.7	13.0	984	-	52
			12-15	61.73	8.57		4.9	9.6	914	-	61
			15-18	60.66	8.01		3.2	9.8	934	-	58
			16-21	59.39	9.45		3.2	6.0	960	-	68
			21-24	63.74	7.37	clay	-(≤1.5)	3.1	970	-	52
82	29.8.	170	o-1	94.59	22.24	mud	-(≤10)	267		-	60
			1-2	92.42	19.70			172		-	58
			2 - 3	64.41	13.49	clay		89		-	
			3 - 4	65.83	11.47			44		-	
			4-5	83.60	10.84			26		-	50
			5 – б	78.37	9.89			19		-	_;
			6-7	77.54	11.15			6		-	48
			7-6	76.89	10.74			-(≤6)		-	46
			6-9	75.29	9.13					-	69
			9-10	75.84	9.81					-	64
			10-12	61.46	13.39					-	
			12-14	80.55	12.60					-	61
96	31.8.	47	0-2	72.67	16.84	mud	17.8	111	813	-	22
			2 - 4	76.61	16.56		19.6	120	841	-	20
			4-6	66.68	16.30		12.1	104	648	-	25
			6 - 8	70.34	16.53		4.3	75.0	770	-	22
			8-10	65.95	15.93		2.9	64.6	850	-	26
			10-12	74.23	15.16		-(≤1.4)	25.2	850	-	21
			12-15	64.66	13.93			9.3	890	-	24
			15-18	72.37	13.78			4.5	a70	-	25
96	31.8.	47	18-21	65.93	13.71	mud		2.9	830	-	25
			21-24	73.48	14.34			2.0	790	-	19
			24-27	73.05	14.19			1.8	860	-	20
			27-30	68.22	13.69			1.8	840	-	21
101	1.9.	19	0-1	71.61	14.01	mud	22.0	154	660	-	27
			1-2	70.17	12.47		10.1	106	690	-	30
			2-4	68.28	11.11	_	3.9	62.6	738	-	16
			4-6	62.30	6.08	clay	-(52)	39.0	740	-	19
			6-9	58.90	8.02			20.0	740	-	19
			9-12	57.60	7.79			7.1	740	-	23
			12-15	53.95	5.32			3.7	690	-	21
			15-18	51.44	4.69			-(≤1.7)	690	-	18

Station Code	Depositio cs-134	n <b>(Bq.m</b> <sup>-2</sup> ) Cs-137 (total)	cs-137 <b>(1986/87)</b>	other nuclides
11	52	2,630	160	
25	53	1,110	160	
33	≤4	305	$(10)^{1}$	
37	3,710	14,170	11,240	Co-60: 62 Sr-90: <b>325<sup>2</sup></b> Ru-106: 1,480 <b>Ag-110m:</b> 210
50	1,320	4,250	4,000	
82	≤4	685	(10) <sup>1</sup>	
96	285	2,750	860	
101	190	2,950	575	

Table III a: Deposition of artifical radionuclides at the sea bottom ('referred to the date of sampling)

notes:<sup>1.</sup>(...) estimated value  $^{2}(...)$  Sr-90, total

Sea area	average water depth (m)	Salinity <b>corres-</b> ponding to water depth <b>(%o)</b>	value of K <sub>d</sub> (Cs-137)
Belt Sea	10	20.0	2.900
Baltic Proper	65	10.0	9.300
Gulf of <b>Riga</b>	23	7.0	13.200
Gulf of Finland	38	6.3	14.930
Bothnian Sea	62	5.5	15.800
Bay of Bothnia	43	4.5	17.700

<u>Table IV:</u> Calculated  $K_d$ -values of Cs-137 in dependence on the salinity

Table V: A comparison betweeen calculated and measured Cs-137 deposition at Baltic sediments in the period May 1986 to August 1987

Sea area	soft bottom area (m <sup>2</sup> )	Removal rate (yr <sup>-1</sup> )	Cs-137-depo calculated (Bq.m	measured
Belt Sea	7.3 E 09	0.14	560	575
Baltic Proper	9.9 E 10	0.36	1040	860
Gulf of Kiga	7.3 E <b>09</b>	0.28	1240	
Gulf of Finland	1.6 E 10	0.19	4070	5880
Bothnian Sea	4.3 E 10	0.12	8810	7530
8ay of Bothnia	1.6 E 10	0.22	3233	3960

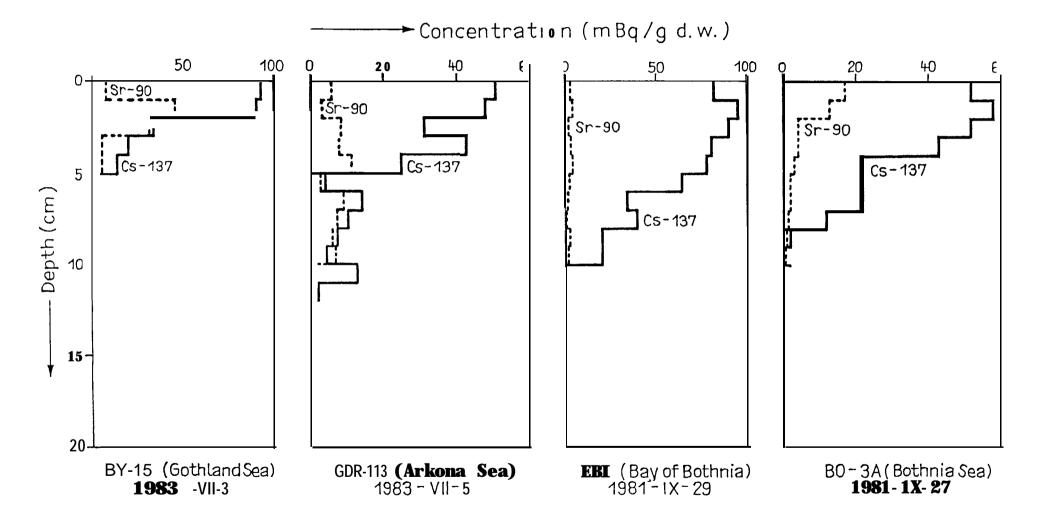
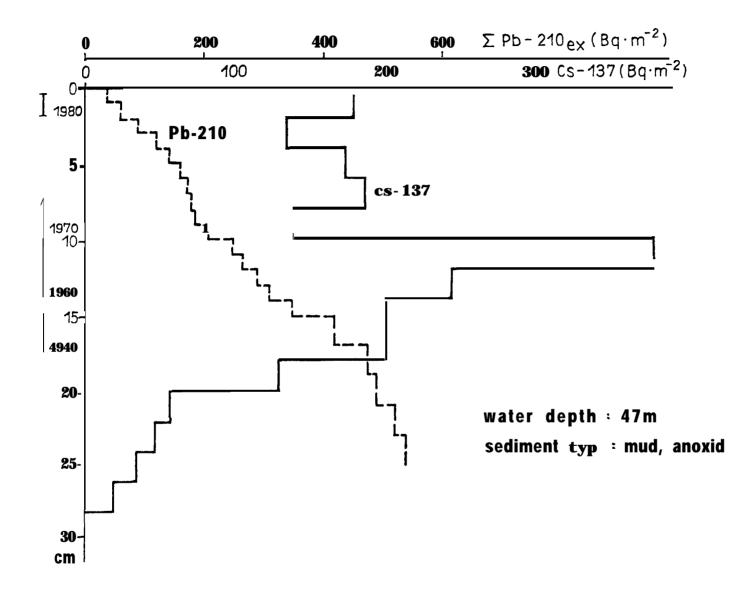
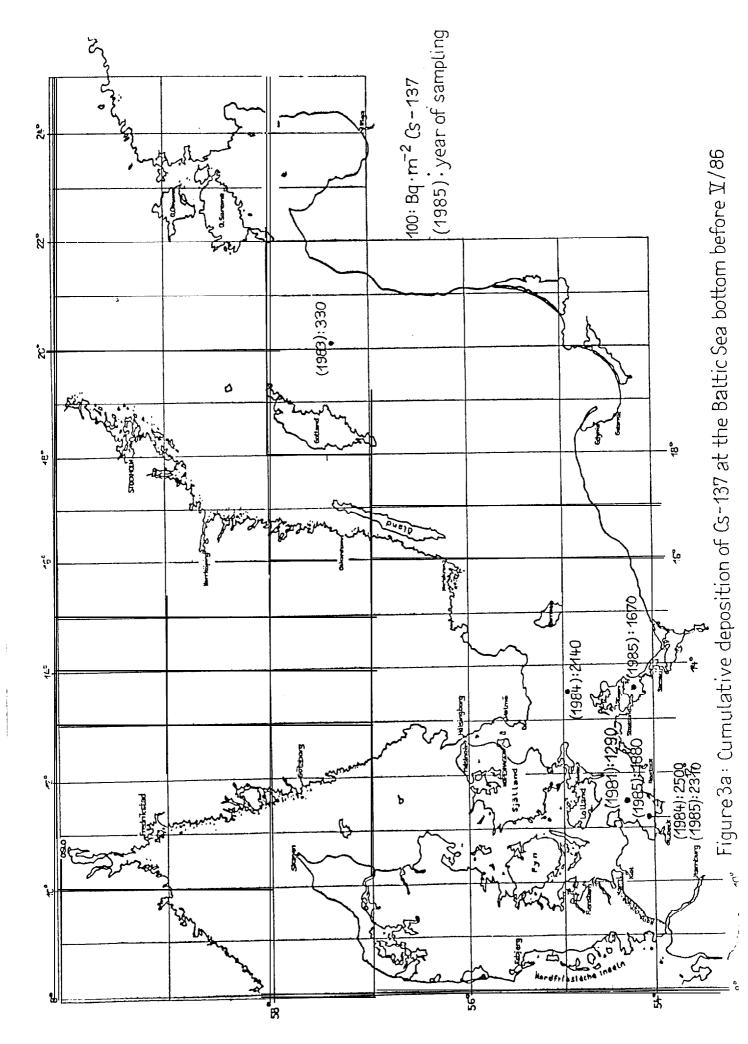


Figure 1: Distribution of Sr-90 and Cs- 137 along sediment cores from different parts of the Baltic Sea



# Figure 2 : Radionuclide distribution along a sediment core, Arkona basin , 1984



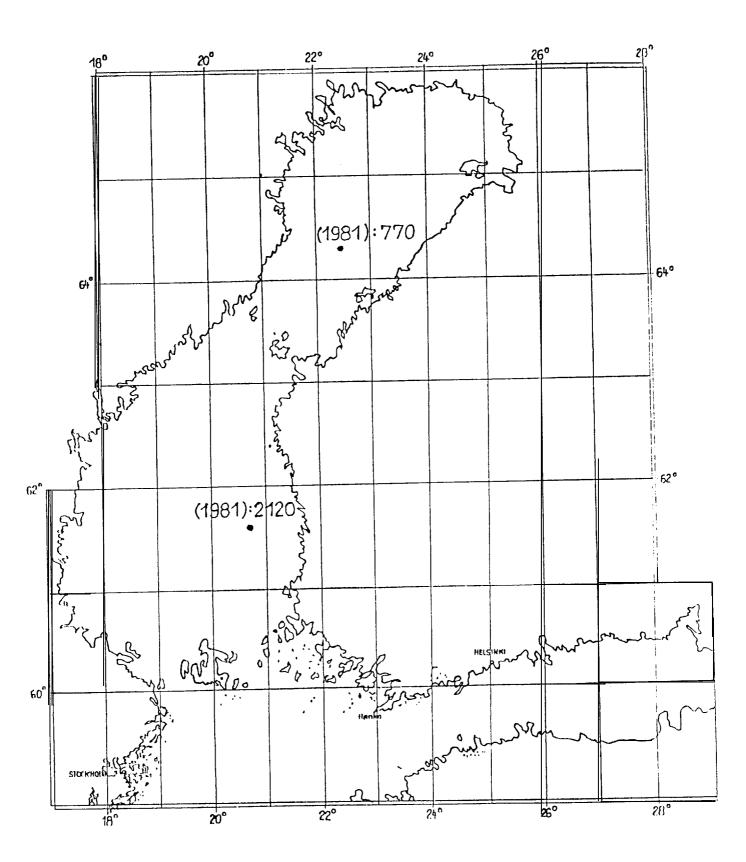
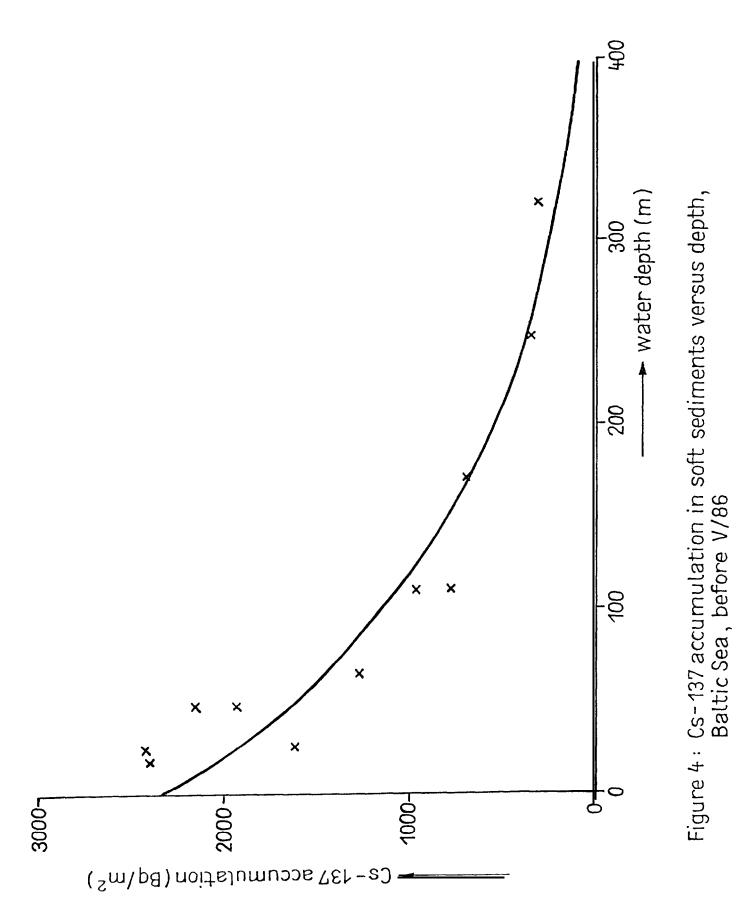
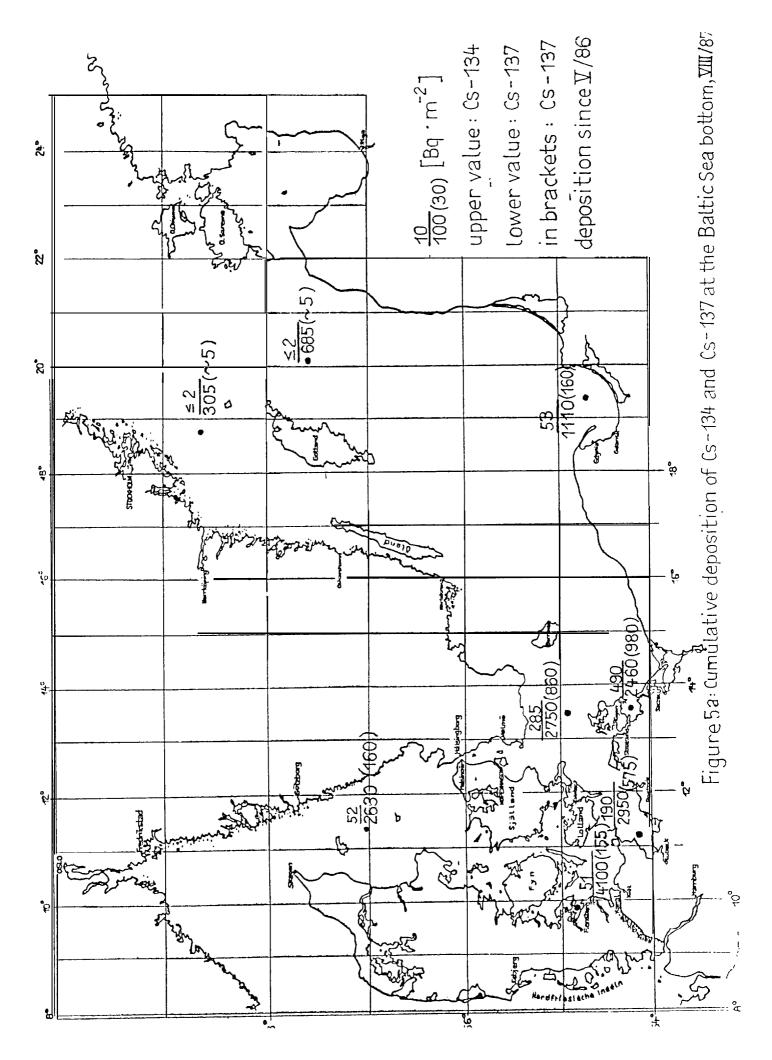


Figure 3b





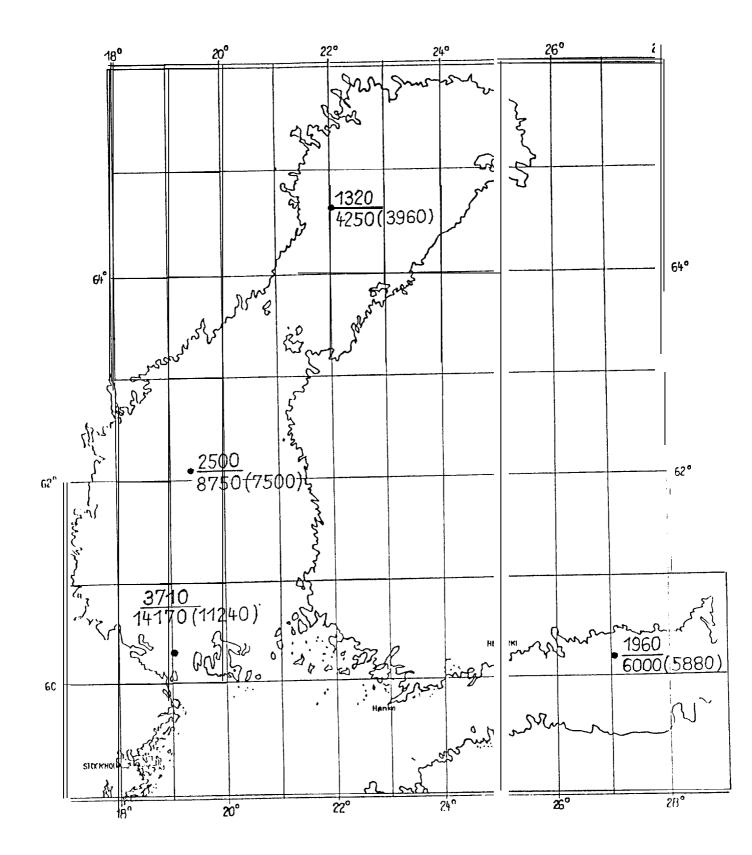
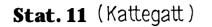


Figure 5b



Stat. 25 (Bay of Gdansk)

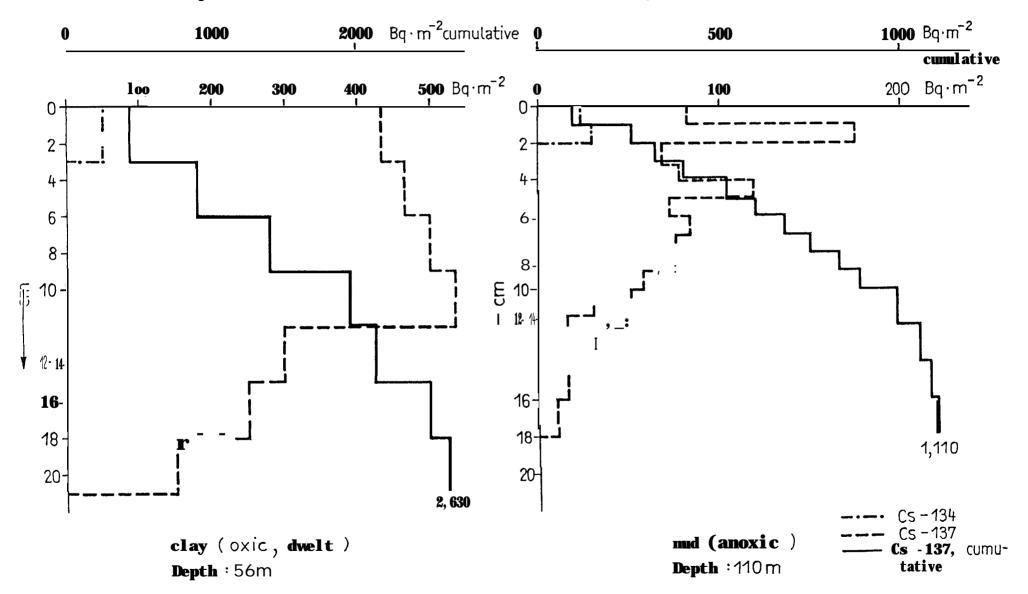
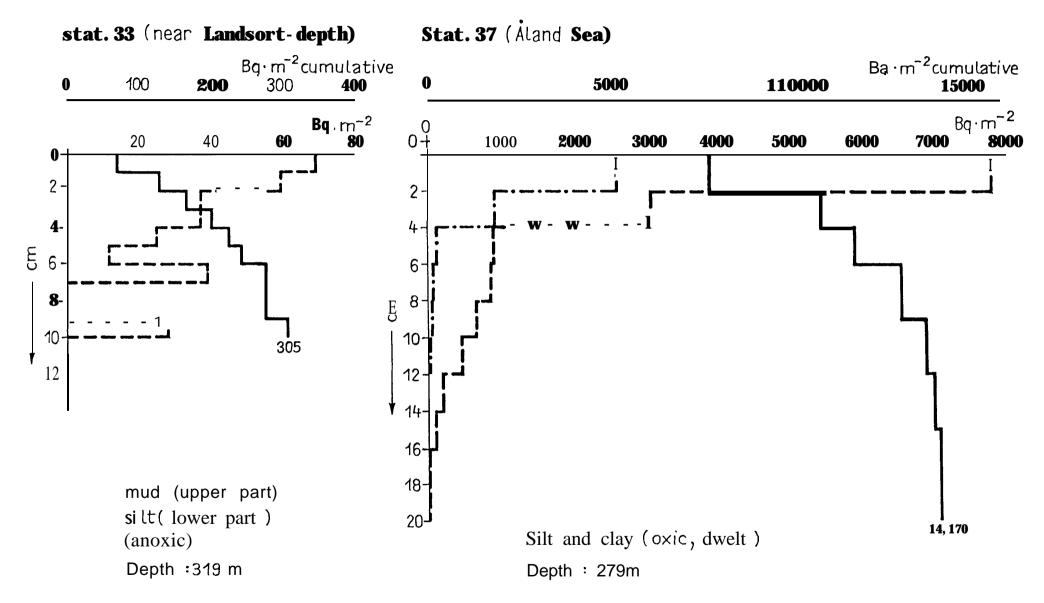


Figure 6a: Distribution of Cs-134 and Cs-137along sediment core profiles from the Baltic Sea, Aug. 1987



# Figure 6b

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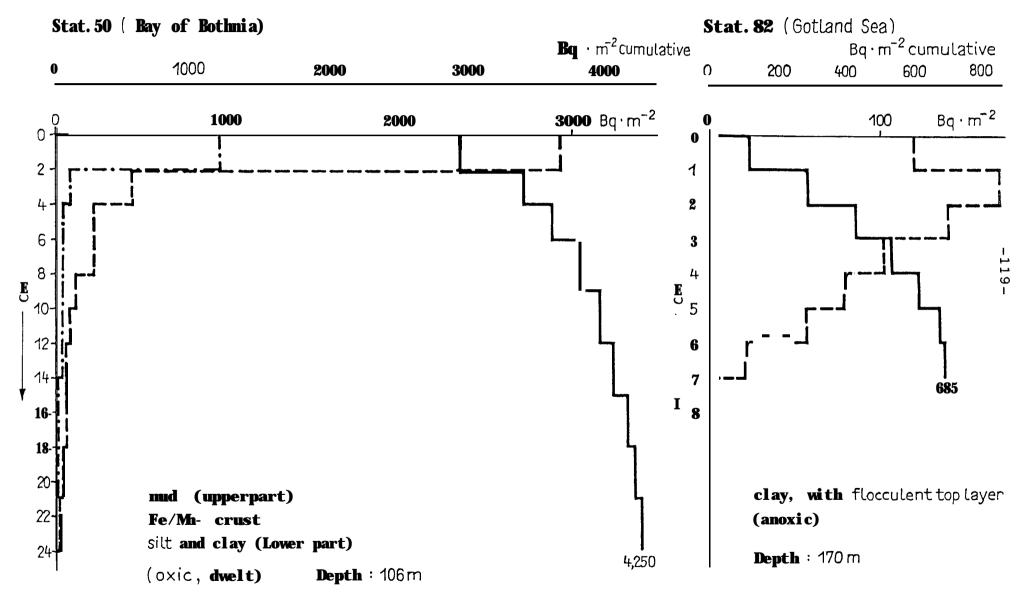
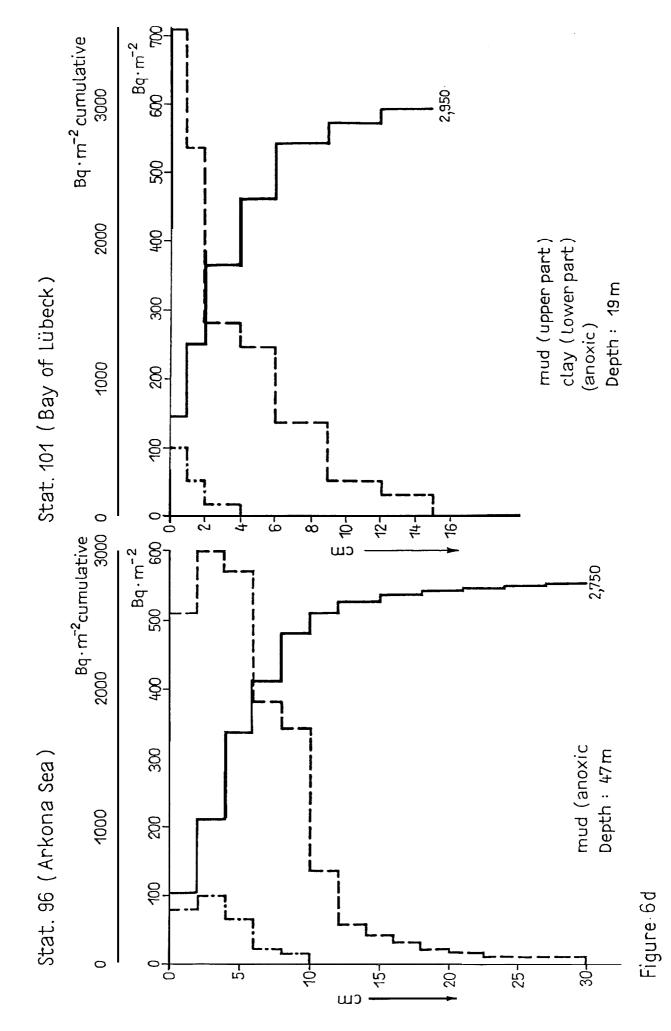


Figure 6c



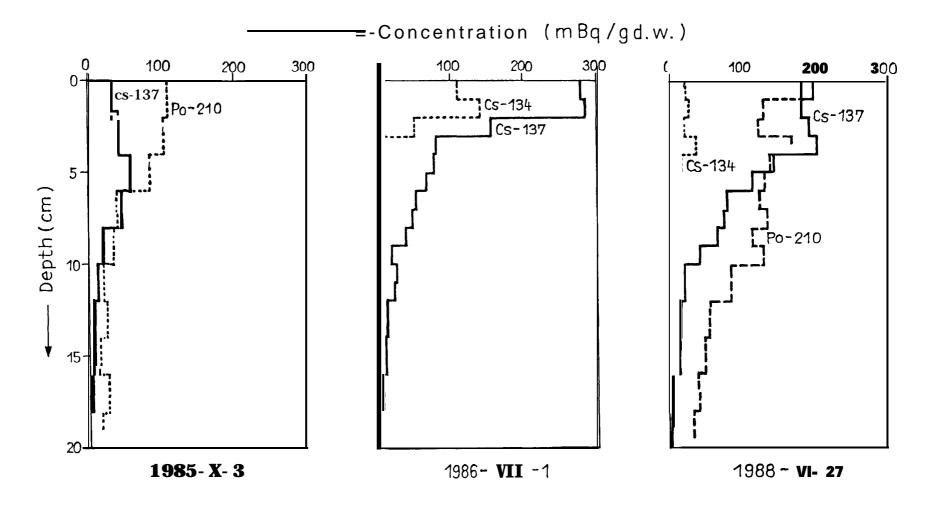


Figure 7: Depth penetration of radiocesium along a sediment core (Greifswald Bodden)

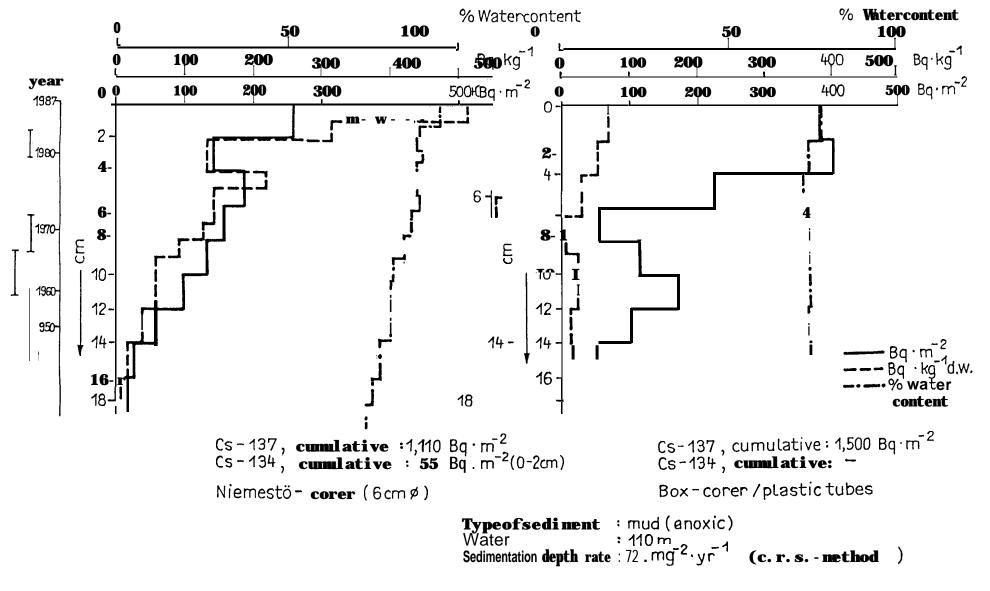


Figure 8: Distribution of Cs-137 along sediment core profiles in dependence on the type of sediment sampler, Stat. 25 (Bay of Gdansk), VIII/ 1987

#### Radioactivity in Biota from the Baltic Sea after the Chernobyl Accident

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#### 1. Introduction

As a result of the fallout after the Chernobyl accident the inventory of artificial radionuclides in the marine environment of the Baltic Sea increased.

To obtain information about the increase of radioactivity in marine biota, in 1986, **1987** and 1988 during three cruises of the FRV "Walther Herwig" to the Baltic Sea samples of fish, as well as few samples of benthic animals and aquatic plants were collected.

Fish was sampled in the region between 10° and 19° E.

#### 2. Materials and methods

On some 50 to 60 stations per cruise, each time carried **out** in December, the biota samples were collected by bottom trawls (1 hour per station). The fished areas were similar in the three years. Fig. 1 shows as example the stations of the 1987 cruise.

The most abundant species of marine fish was the cod. Herring was caught less frequently. Fishes were dissected on board (flesh, liver, skin, gonads or whole fish), and stored deep frozen after pooling several specimen into one sample. 1986 and 1987 at least one sample for a station, where fish could be caught, was obtained. In December 1988, single samples from two to four stations were pooled into one sample for measurement. Few samples of benthos and aquatic plants were collected in December of 1987 and 1988, but only in the western Baltic Sea.

For radioactivity measurements, each biota sample of 5 to 10 kg fresh weight was dried at 130°C and ashed at not more than 460°C. All ashed samples were measured by low-level gamma-ray spectrometry using Germanium detectors with relative efficiencies between 20 and 30 % (counting time between 24 to 70 hours). Depending on the amount of ash, radiochemical analyses of 90Sr and/or plutonium isotopes followed for the greater part of the samples. 90Y-oxalate precipitates were measured with low-background gas flow beta-counters (counted 4 times 500 min). For the measurement of 238Pu and 239/240Pu surface barrier detectors were used after chemical separation (counting time 7 to 11 days).

#### 3. Results of radioactivity measurements

#### 3.1 Results for fish

The main radionuclides, detected by gamma-ray spectrometry were 137Cs, 134Cs and 110mAg. The results for these nuclides as well as for the nuclides 90Sr, 238Pu and 239/240Pu determined radiochemically are presented in the tables 1, 2 and 3

for the years 1986, 1987 and 1988, respectively. In these tables, a simple statistical evaluation is given for each year without differentiating between sampling areas.

The most abundant fish was cod, from which in all three years fillet and liver was analyzed. 1987 also samples of skin and gonads were analyzed. Much less samples were obtained of herring being analyzed as fillet or whole fish samples. Few other fish species were dab, whiting, plaice and flounder.

The most important radionuclides in fish after the Chernobyl accident were the **ce**sium isotopes. Among all analysed fish, the highest activity contents were found in cod. Considering different organs, the cod fillet samples showed the highest activities of cesium. As can be seen from the tables 1 to 3, the cesium isotopes in cod fillet increased from Dec. 1986 to Dec. 1987, however, only **137Cs** increased until Dec. 1988, but not **134Cs.** A similar behaviour was found for cesium in cod liver. Other organs of cod, from which a larger number of samples was analyzed only in Dec. 1987 (table **2**), revealed lower activity contents for cesium compared to fillet.

The cesium contents in other fishes like herring, whiting or dab were found to be lower than in cod (data available only for 1986 and 1987). Cesium in herring fillets also increased from Dec. 1986 to Dec. 1987. Results for other fish species shown in the tables, cannot be discussed well because of their very low number of available samples.

In December 1986 significant amounts of the isotope <sup>110m</sup>Ag were detected in few samples of cod liver. The radioactivity of this nuclide, which can be clearly attributed to the Chernobyl fallout, was even higher than for <sup>137</sup>Cs in liver. Despite of its physical half-live of only 250 days, <sup>110m</sup>Ag increased from Dec. 1986 to Dec. 1987. However, it then decreased from Dec. 1987 to Dec. 1988 by a factor of 0.34, which is very similar to a value of 0.36 obtained for physical decay of this nuclide.

110mAg was not observed in cod fillet. One whole fish cod sample as well as measurements of other whole fishes (herring, dab, whiting) indicated, that it might be found in whole fish samples with low activities due to accumulation in certain organs like liver.

90Sr determinations from 1986 and 1987 (table 1 and 2) in cod fillet yielded very low mean activity contents of 4 to 5 mBq/kg wet weight. If analyses would have been made on edible parts, generally consisting of more than fillet (parts of the skin, etc.), the 90Sr activity would have been at least one order of magnitude higher, as can be seen from anlyses of whole fishes as well as from measurements of cod skin (table 2).

The **90Sr** activities measured in fillet of herring showed higher values, because here the fillet cannot be dissected in the same manner as for cod from other parts of organs generally having higher amounts of **90Sr** (small bones).

Measurements of plutonium isotopes were made only on a part of the samples. Apart from few weak detections of plutonium in cod fillet, which may be doubtful, most of the measurements yielded values below the detection limits. Thus, for the most important fish in the Baltic Sea, it can be concluded, that activity contents for 238Pu and 239/240Pu in fillet are at least lower than 1 mBq/kg wet weight. From the data presented in tables 1 and 2, it cannot be disproved, that this conclusion is also true for other fishes like herring and dab.

## 3.2 Results for low-activity nuclides in cod liver

To obtain better estimates of 90Sr and plutonium in cod liver, ashes of the 1987 samples from different stations were pooled to represent total fresh weights from 5 to 20 kg. The pooled ashes at first were remeasured by gamma-ray spectrometry with counting times of 9600 minutes and then analyzed for 90Sr and plutonium. The results for nuclides not already given for unpooled samples in tables 1 to 3 are presented in table 4. The nuclides 60Co, 65Zn and 108mAg were detected with very low activities.

Especially the long-lived 108mAg, clearly being identified by three gamma lines (half-live 127 years) emphasizes the silver accumulation in the cod liver, the latter already being indicated by accumulation of Chernobyl derived 110mAg. The mean value of 110mAg/108mAg was 2100 (13% la SD, referenced to the Chernobyl accident). 108mAg can partly be expected to be derived from nuclear weapon's fallout, as it was also found in decapods by investigations in the Northeast Atlantic before the Chernobyl accident (Feldt et al., 1985). However, the results of model calculations of reactor inventories with the KORIGEN code (Wiese, 1989; Fischer and Wiese, 1983), showed that the dominant part of the inventories of 110mAg and 108mAg resulted from neutron activation of many kilograms of natural silver used within neutron flux detectors (Van Dam, 1986; Rao et al., 1978). The resulting ratio 110mAg/108mAg depends on the activation duration; a value of 2100 would correspond to an silver activation of roughly half a year.

The mean of the **90Sr** activity is slightly lower than in cod fillet. **65Zn** was detected weakly in three samples from stations belonging to areas D and E (see fig. 1). **238Pu** and **239/240Pu** (only the latter is shown in table 4) were found to be less than the very low detection limits for all samples but one. Thus, **239/240Pu** in cod liver can be expected to be less than 0.1 mBq/kg wet weight.

### 3.3 Development of cesium isotopes in cod after Chernobyl

In the **preceeding** chapter, activity data were discussed without considering differences between areas. For the discussion here, five areas (coded A to E, see fig. 1) have been selected, where in all three cruises from Dec. 1986 to Dec. 1988 samples of cod were collected. Mean activity contents for 137Cs and 134Cs in cod fillet are presented in table 5, ordered by date and area as well as vice versa.

In Dec. 1986, the largest activities were found in area A, the most western area, where the water depth is lower than in more eastern parts of the Baltic Sea. In the following two years,  $^{137}Cs$  did not decrease significantly, however, because of physical decay this was the case for  $^{134}Cs$ . In Dec. 1986, higher cesium concentrations in seawater were observed in this area A (DHI, 1987) than in more eastern areas. Another reason for lower cesium activities in cod fillet in areas B to E in Dec. 1986 is the greater water depth in these areas. Because of incomplete vertical mixing, here the bottom feeding cod could accumulate only a smaller part of the Chernobyl fallout activity deposited to the water surface.

Until Dec. 1987, the activity of the two cesium isotopes in cod increased in areas B to E. Mean values for 137Cs ranged from 12 to 15 Bq/kg, which were significantly higher than in the area A, the higher values being attributed to the areas D and E (fig. 1). One reason for this increase is the southward directed transport of higher contaminated water masses from the southern part of the Bothnian Sea through the Aaland Sea (Nies, 1988) into these areas. Another reason is, that now more radioactivity is available to the cod by depth penetration of the nuclides within one year.

Until Dec. 1988, the activity of 137Cs in cod again slightly increased in areas B to E. Mean values now ranged from 16 to 19 Bq/kg, the higher values coming from areas D and E. However, 134Cs did not increase during this time.

#### 4. Results for gamma emitters in benthos and aquatic plants

Table 6 shows the results for artificial radionuclides obtained by gammaspectrometric measurements on few samples of benthic animals and aquatic plants. It is observed that besides 137Cs, 134Cs and 110mAg being detected in mussels and the sample of seastars, the nuclides 60Co, 106Ru and 125Sb were more or less good detectable in aquatic plants.

5. Estimation of dose to man **by** consumption of Baltic Sea fish

The estimation of the individual dose to man by consumption of fish is based on the results of radioactivity measurements in fish.

Differing estimates for the annual consumption of marine fish in our country lie between 12 and 14 kg. A smaller part thereof consists of cod and herring. For these two species, only a small part is caught in the Baltic Sea. Thus, using a value of 12 kg for the annual consumption of fish overestimates the consumption of Baltic Sea fish significantly. As cod is the dominant fish in the Baltic Sea with the higher cesium activity compared to herring, we use its cesium activity for calculations. With the mean values of 134Cs and 137Cs for Dec. 1988 from table 3, the annual effective dose equivalent is calculated to be roughly 0.004 mSv (0.4 mrem). Because of activity contents of 90Sr being lower by at least three orders of magnitude compared to 137Cs, the contribution of 90Sr can be neglected.

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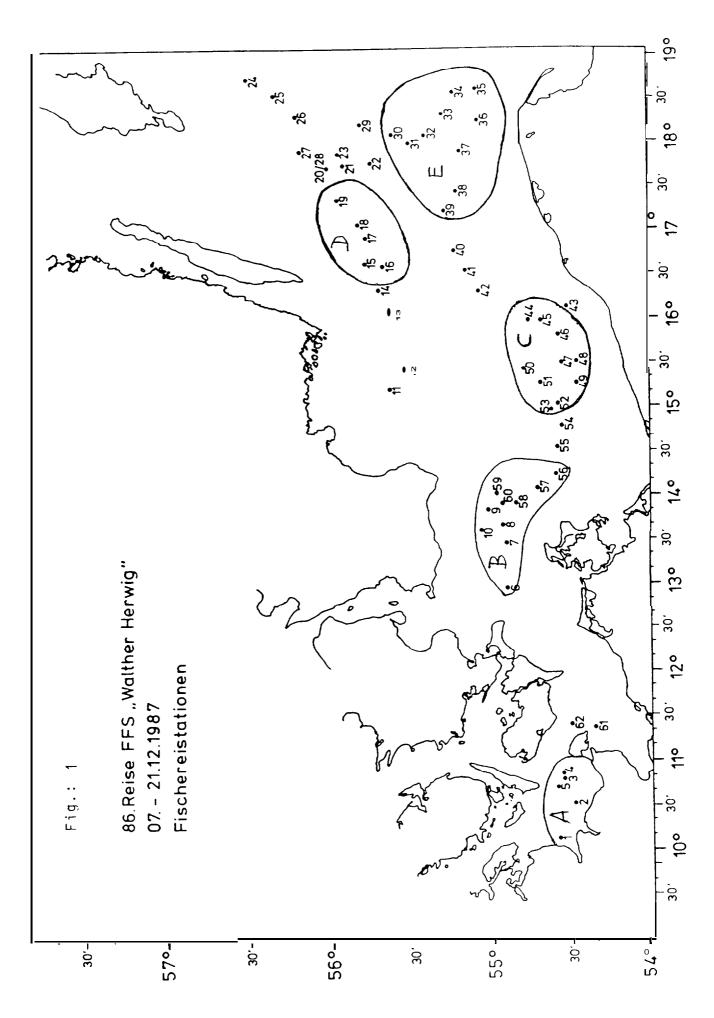


Table 1 : Radioactivity in Baltic Sea Fish in December 1986 Results from the "Walther Her-wig" cruise in December 1986; (N: number of measurements; nd: number < detection limit)

Sample	Nuclide N nd	min. value <b>Bq/kg ww</b>	max. value <b>Bq/kg ww</b>	
Cod: fillet <b>(Gadus</b> morhua)	Sr-90 28 12 cs-134 29 0 cs-137 29 0 Pu-238 17 16 Pu-239 17 15		2.6 9.3 0.00040	0.0040 1.4 5.9 < 0.00040 < 0.0028
Cod: whole fish	Sr-9010Ag-110m10cs-13410cs-13710			0.060 0.60 1.6 4.8
Cod: liver	Sr-9031Ag-110m50cs-13450cs-13750Pu-23822Pu-23922	<pre>&lt; 0.0010     1.7     0.21     0.80 &lt; 0.00014 &lt; 0.00019</pre>	0.0042 2.6 0.25 1.1 < 0.00015 < 0.00023	0.0027 2.1 0.23 0.91
Herring: fillet (Clupea harengus)	Sr-9060Cs-13460Cs-13760Pu-23833Pu-23933	0.014 0.80 3.5 < 0.00024 < 0.00030	0.22 1.1 4.3 < 0.0011 < 0.0018	0.053 0.93 3.9
Herring:whole fi	lsh Sr-90 2 1 <b>Ag-110m</b> 2 0 cs-134 2 0 cs-137 2 0 Pu-238 1 1 Pu-239 1 1	<pre>&lt; 0.0019             0.051             1.0             3.9 &lt; 0.00045 &lt; 0.00056</pre>	0.021 0.087 1.2 4.4	0.011 0.069 1.1 4.2
Dab: fillet (Limanda limanda)	Sr-90 1 0 cs-134 1 0 cs-137 1 0			0.048 1.3 4.4
Dab: whole fish	Sr-90       1       0         Ru-106       1       0         Ag-110m       1       0         cs-134       1       0         cs-137       1       0         Pu-238       1       1         Pu-239       1       0	< 0.00026		0.11 0.31 0.033 1.1 3.5 0.0013
Plaice / Flounder fillet (Pleuronectes pl. and Platichthys fl.)	Sr-90 3 0 cs-134 4 0 cs-137 4 0 Pu-238 1 1 Pu-239 1 1	0.022 0.79 2.7 < 0.00047 < 0.00078	0.035 0.89 3.5	0.030 0.82 3.1
Whiting: fillet (Merlangius mer]	Sr-90 1 0 Ag-110m 1 0 .) Cs-134 1 0 cs-137 1 0			0.0045 0.067 2.0 8.2

Table 2 : Radioactivity in Baltic Sea Fish in December 1987 Results from "Walther Herwig" cruise in December 1987; (N: number of measurements; nd: number < detection limit)

Sample	Nuclide N nd	min. value <b>Bq/kg ww</b>		
Cod: fillet <b>(Gadus</b> morhua)	Sr-90 50 4 <b>Ag-110m</b> 62 60 Cs-134 62 0 Cs-137 62 0 Pu-238 25 25 Pu-239 25 25	< 0.011 1.7 6.5 < 0.00021	0.041 7.6 23.8 < 0.0036	0.0052 4.0 13.8
Cod: liver	Ag-110m520Cs-134520cs-137520	0.31	1.1	2.74 0.69 2.36
Cod: gonads	Sr-9021Ag-110m53cs-13450cs-13750Pu-23822Pu-23922	< 0.044 1.0 3.8 < 0.00080	3.0 10.7 < 0.00090	0.0071 0.054 2.2 7.9
Cod: skin	Sr-9070Ag-110m1413cs-134140cs-137140Pu-23877Pu-23976	< 0.037 1.4 4.9 < 0.00050	3.2 10. < 0.0022	0.29 2.1 7.1
Herring: fillet (Clupea harengus)	Sr-9050cs-13490cs-13790Pu-23822Pu-23921	0.0046 0.40 3.2 < 0.00082 < 0.0010	2.6 8.9 < 0.00089	0.0088 1.8 6.8
Herring:whole fi	sh Sr-90 2 0 <b>Ag-110m</b> 2 0 cs-134 2 0 cs-137 2 0 Pu-238 1 1 Pu-239 1 1	0.064 0.11 2.4 8.2 < 0.00036 < 0.00060	0.075 0.11 2.7 8.8	0.069 0.011 2.6 8.5
Dab: fillet (Limanda limanda)	Sr-90 1 0 cs-134 1 0 cs-137 1 0			0.037 0.92 4.1
Sugar kelp (Laminaria sacch (Bq/kg dry !!)	Sr-90 1 0 Ag-110m 1 0 a. Cs-134 1 0 cs-137 1 0 Pu-238 1 0 Pu-239 1 0			1.4 0.46 5.5 23. 0.012 0.063

Sample	Nuclide	N	nd	min. value <b>Bq/kg ww</b>	max. value <b>Bq/kg ww</b>	Mean <b>Bq/kg ww</b>
Cod: fillet (Gadus morhua)	Sr-90 Cs-134 Cs-137 Pu-238 Pu-239	7 8 8 4 4	0 0 0 4 4	0.0028 1.40 7.70 < 0.00024 < 0.00042	0.0117 4.81 20.9 < 0.00025 < 0.00043	0.0056 3.77 16.7
Cod: liver	<b>Ag-108m</b> <b>Ag-110m</b> cs-134 cs-137	5 5 5 5	0 0 0 0	0.0088 0.45 0.34 1.70	0.015 1.45 0.73 3.17	0.011 0.93 0.57 2.57
Flounder: fillet (Platichthys fl.)	cs-134 cs-137	1 1	0 0			1.72 7.87
Dab: fillet (Limanda limanda)	Sr-90 cs-134 cs-137	1 1 1	0 0 0			0.032 0.66 3.55
Herring: fillet (Clupea harengus)	Sr-90 cs-134 cs-137	1 3 3	0 0 0	0.337 2.40	2.46 10.8	0.0034 1.21 5.84

Table 3 :Radioactivity in Baltic Sea Fish in December 1908<br/>Results from "Walther Herwig" cruise in December 1988;<br/>(N: number of measurements; nd: number < detection limit)</th>

 Table 4 : Further nuclide activities in pooled samples of cod liver from December 1907

Samples were pooled over different stations

Sample	60C0	65Zn	90Sr	108mAg	239/240Pu
number		<b>Bq/kg</b> wet w	eight <b>±</b> lo SD	(X)	
12052 12053 12054 12055 12056 12057 12058	5.09E-3 ±18. 5.23E-3 ±22. 5.85E-3 ±18. 3.97E-3 ±28.	2.87E-2 f23. 2.25E-2 ±25. <1.89E-2 <1.71E-2	<2.33E-3 2.16E-3 ±14. 2.58E-3 ±7.4 3.88E-3 ±7.0	9.60E-3 ±10. 6.663-3 ±8.2 5.82E-3 ±9.0 6.76E-3 ±9.4 5.23E-3 ±9.4 6.82E-3 ±8.9 8.45E-3 ±9.4	<5.1E-5 <7.7E-5 <4.0E-5 <4.9E-5 <7.7E-5

# Table 5 : **Mean cesium** contents **(Bq/kg** wet) in <u>cod fillet</u> from 5 different areas in the Baltic Sea

(134Cs/137Cs is the ratio of the means; activity values calculated for date of sampling; see fig. 1 for the 5 areas )

Date	Area	Stations	137Cs	134Cs	134Cs/1 37cs
ord	ered i	by date <i>and area</i>			
12/86	A B C D E	1-9 12, 58, 59 44-46 27-32 49-56	8.0 6.4 5.6 5.9 4.8	2.3 1.7 1.4 1.5 0.8	0.29 0.27 0.25 0.25 0.17
12/87	A	1-5	8.0	2.0	0.25
	B	6-10, 56-60	11.9	3.2	0.27
	C	44-53	13.3	3.8	0.29
	D	15-19	15.1	4.5	0.30
	E	30-39	14.5	4.2	0.29
12/88	A B C D E	1-2 6-8 15-17, 41-43 30-33 <b>47-50,23-24,26-28</b>	7.7 17.1 15.9 19.4 19.3	1.4 3.9 3.6 4.5 4.4	0.18 0.23 0.23 0.23 0.23 0.23
ord	ered j	by area and date			
12/86	A	1-9	8.0	2.3	0.29
12/87		1-5	8.0	2.0	0.25
12/88		1-2	7.7	1.4	0.18
12/86	В	12, 58, 59	6.4	1.7	0.27
12/87		6-10, 56-60	11.9	3.2	0.27
12/88		6-8	17.1	3.9	0.23
12/86	С	44-46	5.6	1.4	0.25
12/87		44-53	13.3	3.8	0.29
12/88		15-17, 41-43	15.9	3.6	0.23
12/86	D	27-32	5.9	1.5	0.25
12/87		15-19	15.1	4.5	0.30
12/88		30-33	19.4	4.5	0.23
12/86	Ε	49-56	4.8	0.8	0.17
12/87		30-39	14.5	4.2	0.29
12/88		<b>47-50,23-24,26-28</b>	19.3	4.4	0.23

Table 6 : Artificial gamma emitters in benthic **animals** and in aquatic plants Samples from area A (Fig. 1)

Sample	Base	Date	60C0	106Ru	110mAg	125SD	134Cs	137Cs
Mussels Cyprina islandica Cyprina islandica	wet wet	12/88 12/88	0.039 ±31. <0.035	<0.26 <0.24	0.36 <b>±5.2</b> 0.10 <b>±10.</b>		0.084 f13. 0.070 <b>±13.</b>	0.51 f7.7 0.36 <b>±7.9</b>
Seas <i>tars</i> Asterias <b>rubens</b>	dry	12/88	0.21 f33.	<1.1	0.41 <b>±11.</b>	<0.32	0.68 f8.5	3.66 <b>±7.4</b>
Aquatic plants Laminaria sacchar. 1) Laminaria sacchar. Fucus vesiculosus	dry dry dry	12/87 12/88 12/88		3.2 ±14. 3.0 ±18. 20.7 ±8.0	<0.16	2.0 <b>±11.</b>	5.45 <b>±5.3</b> 3.36 <b>±6.1</b> 2.27 <b>±12.</b>	22.9 f7.4 16.8 f7.0 14.7 f7.1

Activities in Bq/kg wet or dry  $\pm$  la SD (%)

1) see table 2 for 90Sr and plutonium

THE RADIOACTIVE CONTAMINATION OF FISH FROM THE BALTIC SEA AND THE DOSE EQUIVALENT TO THE POPULATION OF THE GDR RESULTING FROM FISH CONSUMPTION DURING 1986 - 1988

K. Jarmatz, D. Weiss

Staatliches Amt für Atomsicherheit und Strahlenschutz Waldowallee 117 DDR-Berlin 1157 German Democratic Republic As in previous years, also in the period from 1986 to 1988, fish samples were taken by the National Board for Atomic Safety and Radiation Protection from commercial catches in the Baltic. These fish samples were fractionated into the fractions flesh, skin plus bones and guts, and examined by means of gamma spectrometry.

In the following only the results of the flesh fraction are discussed because the others play no role for consumption. In 1986, fish samples from coastal waters of the GDR were only taken on 7 and 8 May since in that year sampling from inland waters had priority. Cesium enrichment in fish is inhibited by high potassium concentration as found in coastal waters. Hence, compared with freshwater with a lower content of potassium, the radiocesium concentrations in fish at the same level of water contamination were expected to be lower. Despite a relatively high water contamination of 300 mBq/1 the Cs-137 concentration in the samples taken in May 1986 still agreed with the values of previous years. This is due to the fact that cesium is mainly taken in by food, and the active ion regulation via the gills is negligible. This is why cesium-134 with concentrations of about 0.5 Bq/kg (wet weight) could only be detected in few samples.

Due to the mentioned influence of potassium on cesium enrichment the Cs-134 and Cs-137 concentrations in fish flesh determined in 1987 and 1988 were distinctly lower than in the same species of fish from inland waters (Fig. 1).

As the accumulation of cesium in fish also depends on the position of the species within the food chain, the piscivorous fish perch, pike-perch and pike consequently have the highest radiocesium concentration. This is illustrated by the example of an inland *water* of the GDR (Lake Mueggelsee) in Pig. 2.

As regards the mentioned piscivorous freshwater fish from coastal waters, radiocesium concentrations of between 7 Bq/kg (wet weight) and 79 Bq/kg (wet weight) were measured.

In contrast, the concentrations in non-piscivorous fish only ranged from **6** Bq/kg to 19 Bq/kg (wet weight). The lowest enrichment was found in marine fish with concentrations in fish flesh lying between 5 Bq/kg and 9 Bq/kg. A higher radiocesium concentration of 16 Bq/kg was only detected in cod flesh (piscivorous marine fish).

Strontium-90 measurements in selected fish samples in the period from 1986 to 1988 showed no detectable increases compared with measurements of previous years.

For the determination of the ingestion dose resulting from the consumption of fish from the Baltic Sea and the GDR's coastal waters, only the radionuclides **Cs-134** and **Cs-137** were considered because strontium in fish flesh is not enriched under brackish-water and salt-water conditions.

For the year 1986 in which fish samples were taken from the above-mentioned waters only on 7 and 8 May, the ingestion dose during the period from January to May 1986 was estimated on the basis of the 1985 values. For the period from June to December **1986**, the radiocesium concentration of samples taken in 1987 was used. This calculation can be substantiated by the results of fish samples from inland waters. From autumn 1986 to late 1987, a nearly unchanged fish contamination was stated, The real values of radiocesium concentration in the fish flesh of the analyzed samples served as the basis for calculation for the years 1987 and 1988.

The calculation of the individual effective dose equivalent resulting from the consumption of fish from the Baltic Sea and the GDR's coastal waters is given in Fig. 3 where a differentiation is made between persons with a medium and a maximum consumption rate. The group with the maximum consumption rate comprised about 3000 persons representing fishermen and their families. Fig. 3 shows that in 1986 the ingestion doses at a medium and a maximum consumption rate are higher by a factor of 3 and 4, respectively and in 1987 by a factor of 7 and 6, respectively than in 1985. On the basis of values measured in fish samples in 1988 a slight decrease in the fish flesh contamination could be established. Compared with 1985, the effective ingestion dose equivalents for medium and maximum consumption rates in 1988 were higher by a factor of 3 and 4, respectively. As against 1987, the ingestion dose to the population only amounted to 53 per cent and to the group of fishermen to 79 per cent in 1988. The reason for it is to be seen in the reduction of water contamination due to mixing up, in the appearance of steady states and in the decrease of Cs-134 concentration due to physical decay.

The ingestion dose caused by the consumption of fish from the Baltic Sea and the coastal waters corresponds in about to that resulting from the medium consumption rate for fish from inland waters (adults 8kg/year of which about 65 per cent of saltwater fish).

This result is not in contradiction to the contamination values shown in Fig. 1, since the consumption of fish from extensively used inland waters is very low. Most of the fish come from aquacultures where they are fed with only slightly contaminated fodder so that the contamination values are very low too. In this way the relatively high contamination values of wild fish are compensated.

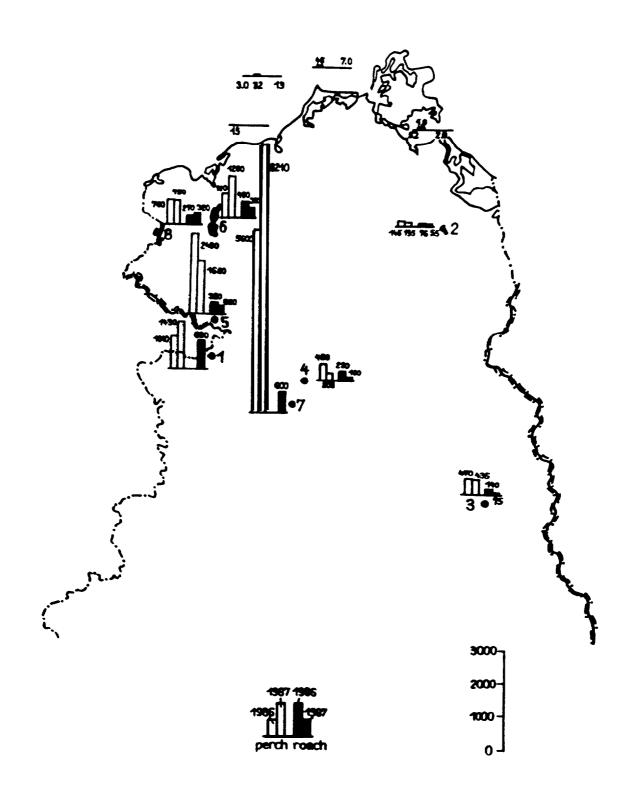
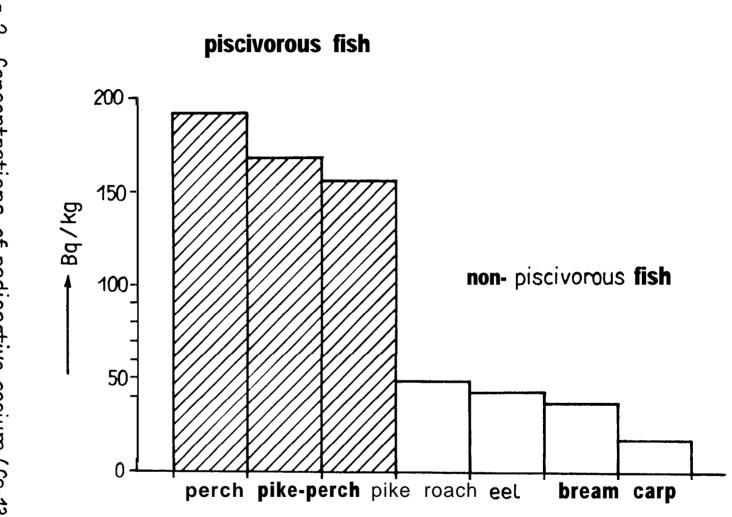


Fig. 1 Concentrations of radioactive cesium (Cs-134, Cs-137) in perch and roach (flesh) in 1986 and 1987 (maximum values)





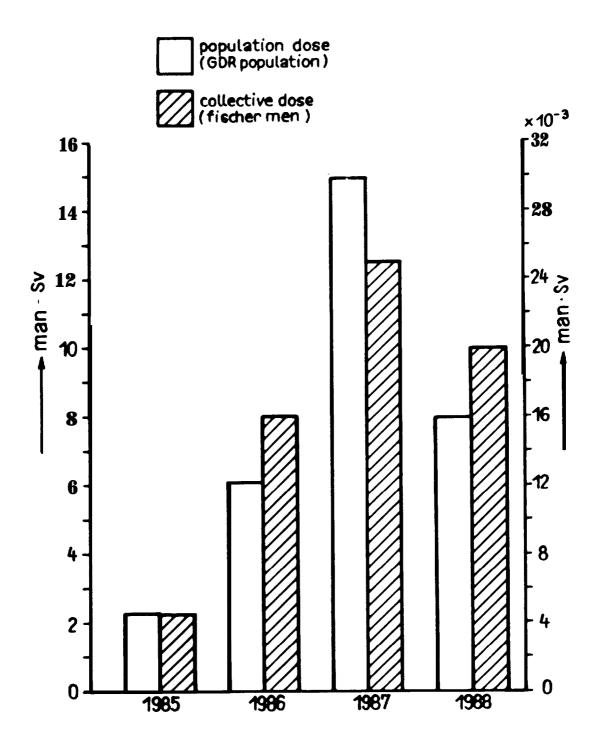


Fig.3 Ingestion dose resulting from consumption of Baltic fish (1985 - 1988)

fish species	<b>Concentration</b> cs-134 cs-137 (Bq.kg <sup>-1</sup> f.w.)		Consumption rate (kg per man and	Ingestion (Bq.y <sup>~1</sup> ) Cs <b>-</b> 134 <b>cs-137</b>		per capita effective equivalent dose (Sv)		
	( 1 )	- /	year)				(57)	
non-piscivorous fish, narine	0. 7	4. 4	1.80	1.26	7. 92	1.24	E- 7	
spratt, herring, flounder, dab			8.00	5.60	35. 20	5. 53		
piscivorous fish, mrine								
cod	1.5	9.2	0.30 3.00	0. 45 4. 50	2. 76 27. 60	4. 35 4. 35		
non-piscivorous fish, fresh-water	1.5	5. <b>8</b>	0. 20	0. 30	1.16	2. 02	E- 8	
roach, bream			1.00	1.50	5.80	1.01	E- 7	
piscivorous fish, fresh-water	5. 2	21. 2	0. 40	2.07	8. 47	1.45	E- 7	
perch, pike-perch, pike			8. 50	44. 20	180. 20	3. 09	E- 6	
other species	1.8	6. 7	0. 30 9. 50	0. 54 17. 10	2. 01 63. 65	3. 52 1. 12		
lower val		consumptio imm consum 11.			Sum Sum 146 man.Sv			

<u>Table 1:</u> Calculation of ingestion doses resulting from consumption of Baltic fish in 1986

fish species	Concentrat Cs-134 (Bq.kg <sup>-1</sup> f.		<b>Consumption</b> (kg per man year)		/ <sup>-1</sup> )		<b>capita effective</b> 7 <b>alent dose</b> (S∨)
non-piscivorous fish, narine	1.2	5.6	1. <b>80</b>	2. 16	10. 08	1.68	E- 7
spratt, herring, flounder, dab			8. 00	9.60	44. 80	7. 45	E- 7
piscivorous fish, marine	2.6	13. 7	0. 30	0. 78	4. 11	6. 67	E- 8
cod			3.00	7.80	41.10	6. 67	
non-piscivorous fish, fresh-water	2.5	9.0	0. 20	0. 50	1.80	3. 19	E-8
roach, bream	-		1.00	2. 50	9.00	1.601	E-7
piscivorous fish, Fresh-water	8. 9	32.9	0. 40	6. 67	<b>32. 8</b> 3	5. 77	E- 7
perch, pike-perch, pike			8. 50	75.65	279,65	4. 93	E- 6
other species	3.0	10. 0	0. 30	0. 90	3.00	5.44	E- 8
			9. 50	28. 50	95.00	1. 72	E- 6
Renarks: s. Table	8		Popul : Col l e		Sum Sum 14. 997 nan. Sv	8. 98 8. 22	

Table 2	Calculation	of	ingestion	doses	resulting	from cons	umption	of	Baltic	fish	in	1987

	<b>capita ivalent</b> (Sv)		1)	Ingesti (Bq.y <sup>-1</sup> Cs-134	<b>Consumption rate</b> (kg per man and year)	Cs-137	<b>Concentr</b> cs-1 <b>34</b> (Bq.kg <sup>-1</sup>	fish species
7	4 E-7	<b>1. 94</b>	11.88	2.34	1.8	6.6	1.3	non-piscivorous fish, marine
7	3 E-7	8.63	52.8	10.40	8.0			herring, flounder
8	0 E-8	3. 90	4.02	0.78	0. 30	13. 4	2.6	piscivorous fish, marine
7	5 E-7	6. 55	40.20	7.80	3.00			cod
B	5 E-8	2.65	1.62	0.32	0.20			non-piscivorous fish, fresh-water
7	2 E-7	1.32	8.1	1.60	1.00	8.1	1.6	roach, bream
	6 E-7 7 E-6		11.64 247.35	2.64 56.1	0,40 8.50	29.1	6.6	piscivorous fish, <u>fresh-water</u> perch, pike-perch
8	4 E-8 2 E-7	2. 34	1.41 44.65	0.30 9.50	0.30 9.50	4.7	1.0	other species
	<b>9 E-7</b> 56 E-6		Sum: Sum: man.Sv		Popul at i on de			
5	6 E-6			ose				

<u>Table 3:</u> Calculation of ingestion doses resulting from consumption of Baltic fish in 1988

### Intercomparison of Radionuclides Determined in a Baltic Sea Sediment Sample

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### Introduction

In 1988 the International Laboratory of Marine Radioactivity (ILMR) prepared and distributed 3 new marine materials with quantities of some man-made and natural radionuclides for intercomparison and certification. One was a sample of Baltic Sea sediment (IAEA-306) that was distributed to 102 scientists who were requested to prepare 3 aliquots from the sample and measure as many man-made and natural radionuclides as possible using analytical methods available to them. All participants were informed that the expected range in concentration for the man-made radionuclides was 0- 100 Bq kg<sup>-1</sup>. Results have been received from 84 participants including 15 sets of results from laboratories in member states with representatives to the Group of Experts on Monitoring Radioactive Substances in the Baltic Sea (MORS).

An interim report *issued* in March 1989, indicated that some of the reported concentrations were far from the acceptable mean values determined from a statistical analysis of the data base then available. All participants were informed of the interim results and urged to look over their data and inform us of any errors. There were several replies and the affected data was corrected according to the instructions provided. All other results remain as originally submitted.

Some of the concentrations of radionuclides in the global data base and the MORS data base are compared herein to assess the quality of results being generated by the MORS member state laboratories participating in this exercise. A more detailed analysis of results from individual laboratories is being prepared and will be appear in a subsequent IAEA publication from this laboratory during 1989.

### SampleCollect ion and Processing

The 0 to 5 cm surface layers from a series of sediment cores collected in the Baltic Sea during a cruise in October-November 1986 on the R/V GAUSS (Deutsches Hydrographisches finstitut of Hamburg, Federal Republic of Germany), were combined to yield a sample of approximately 30 kg in weight. The material was returned to the laboratory where it was freeze-dried and ground in a laboratory mill. The powdered sediment was next passed through a Imm diameter sieve and further homogenized by mixing in a stainless-steel rotating drum for one week. An aliquot of the material was removed and sieved. 85% of the material was found in the  $63\mu$ m to  $250\mu$ m size range while the remaining material was equally divided between the fractions  $<63\mu$ m and  $>250\mu$ m. The moisture content was determined to be 0.9% of the dry weight at the time of sample preparation. Plastic bottles were filled with 100 g of the sediment and labelled with the code name !AEA-306.

### Homogeneity Tests and Results

Prior to releasing any Intercomparison material from ILMR, it is a requirement to assure that the radionuclides are distributed among the samples in a manner that satisfies a homogeneity test. Ten bottle were selected at random from the prepared lot of 140 bottles. The amount of 40K and 134, 137Cs present in the individual samples was determined instrumentally by gamma spectrometry. Plutonium and americium were chemically separated from the samples, plated onto stainless steel disks, and measured by alpha spectrometry. Homogeneity was determined using one way analysis of variance and it was concluded that the material satisfied the homogeneity criteria for the radionuclides measured.

# Evaluation of Results

The concentrations of the specific radionuclides measured by the 84 participants were collated and all "less than" values were segregated from the results. The remaining concentrations of the specific radionuclides were checked for the presence of outliers by a non parametric statistical test. The application of this test to sets of data is performed on a computer using a commercially available statistical software application. Table 1 is a summary of only of the results for the radionuclides that were determined by MORS member state participants showing the number of reported and accepted results, number of outliers, most reliable estimates of the mean-median values, and the confidence interval about the true median (summaries for radionuclides reported by other laboratories but not by MORS participants will appear in the final report from this laboratory). The confidence intervals were determined using the number of measurements that passed the statistical test for outliers and a statistical table defining the two-sided 95% confidence limits for the median of any continuous distribution. Computed median concentrationns are considered to be the most reliable estimators of the true values. Further details of the data evaluation procedure for AEA 306 w i be described in our final report but previous reports on the certification procedure used at ILMR may also be consulted for information (for example, IAEA/AL/O 12, Dec. 1988).

### **Results and Discussion**

Table 2 shows the MORS member state contributors identified by an

Summary of Data for Radionuclide Concentrations in Intercomparisan Baltic Sediment. IAEA 306.

Reference date 1 Jan 1988.

		Ba/kg dry weight							
Radionuclide	* of Reported	• of Outliers or	Accepted	Mean	Standard	coefficient of	Median	Confidence limits	
Measured	Results	<u>.ess than Value!</u>	Results	Concentration	Deviation 1	Variation <b>X</b>	:oncentration	a ≖ 0.05	
239Pu	30	4	26	5.8	0.5	8	5.8	5.5-6.3	
238Pu	23	5	18	0.16	0.04	24	0.17	0. 14- 0. 19	
241Am	29	9	20	1.9	0.3	17	1.9	1. 6- 2. 1	
40K	69	8	61	799	105	13	785	757-827	
137cs	76	6	70	201	17	8	201	194-206	
134Cs	72	а	64	52	6	12	53	50-54.3	
106Ru	23	16	7	18	10	55	16	2. 1-31	
125Sb	14	0	14	4.8	0.9	18	4.9	3. 8- 5. 4	
905r	18	4	14	4.6	1.2	25	4.8	3. 5-6. 0	
2350	11	0	11	7.5	3.8	50	6.4	2.7-12.4	
238U	15	3	12	73	6	9	75	65-77.4	
234Th	7	0	7	83	22	27	77	65-127	
234U	8	1	7	81	4	5	82	76-86	
230Th	7	0	7	60	4	6	62	53-63	
226Ra	25	4	21	87	29	33	84	64-102	
214Pb-Bi	34	9	25	71	16	23	70	61-81	
2 10Pb	16	1	15	462	103	22	435	381-535	
<b>2</b> 10Po	13	0	13	393	113	29	407	305-489	
232Th	13	1	12	49	4	8	48	46. 6-53	
228Ra-Ac	25	0	25	51	7	14	50	47-54.5	
228Th	17	2	15	50	5	10	SO	45-54	
212Pb-B1,208T1	24	2	22	43	16	36	48	29- 54	

Table 1

	Radionuc	lide Conce	Iration Ba	/kg dry w	1	Detected	by: * allp	ha spectro	metry;	amma si	pectron	netry; beta_	<u>un t</u> ing
Code #	239PU	238Pu	Am241	40K	137cs	134cs	106Ru	125Sb	905r	235U	_238U_	234Th-234mP≀	<u>234U</u>
3	5.5±0.1*	0.18±0.01*	1.6±0.1*	655±36*	170±9*	47±3°	31±6*				67*1*		78±2*
-4	5.1±0.3*	0.13±0.02*	1.61±0.20*	711±7°	183±3°	45±3*	12±2*	6.1±0.5*		6.5±0.7*		95±6*	
			2.4±0.2*										
13	5.7±0.2*	0.14±0.03*	1.96±0.4*	701±13°	179±3°	51±1*		3.8±1.1°	3.5±0.2			77±3*	
22				830±10*	218±3*	62±2*				11±1°		127±15*	
23			4.2±3.1°	754±6*	190±16*	49±4*		5.3±1.5°		11.5±0.6*			
30											65±2*		76±2*
31	5.7±0.3*	0.19±0.03*	1.7±0.1*	766±7*	199±2°	56±1*			5.6±0.1				
33	6.3±0.9*	5.9±08*		821±17°	209±7°	54±2*			99±6'	2.7±0.5*	77±3*		83±4*
36			5.1±0.7*	1135±39°	218±4*	80±1*							
50				763±19°	178±2*	43±1°							
64	6.0±0.3*			896±6*	230±2*	62±3*			6±2'				
65	5.4±0.2*	0.18±0 <b>04</b> '	2.4±0.2*		105±4'	19±2'			4.4±0.1'		69±4*		79±3*
6′3				690±80*	174±11*	37±5*			6±1′				
75				711±18°	192±2*	58±1*				12.4±1.3*			
16				407±19°	_228±19*_	29±5°						-	
			230Th	<u>226Ra</u>	<b>2</b> 14Pb-Bi	210Pb	210Po	232Th	228Ra-A(	228Th	l - 1208	-	
3			59±5*		82±5*		241±7*	47±3*	38±6*	54±2*			
4					53±2*	358±2*	407±7*		48±2*				
13				102±5°		381±52			45±1°	44±1*			
22					94±4				60±2*				
23													
30							501±9*						
31							513±18*		47±4*				
33				51±3*		535±34'				45±2*			
36													
50				83.8±3*							56±11*		
64				113±3'	76±2*	119±32	418±18*	44±1*		49±2*	54±1*		
65			63±6*					45±4*		45±4*			
69			56±4*										
75					51±3*				46±2*		19±2*		
81	I				74+3*	I			I				

Table 2. Results from MORS Member State Participants-IAEA 306

assigned code number, the analytical measurement techniques used; and concentrations of specific radionuclides reported. The MORS results are a subset of the global data base and outliers, as identified from the statistical tests with all results, are shown in italics. Each MORS participant is encouraged to compare their individual results with the computed median values for the radionucides listed intable 1 and to review their procedures if outliers are evident in the data.

# 40<sub>K</sub>

All values for  ${}^{40}$ K concentrations from the 69 reporting laboratories, including 13 participants from MORS member states, were determined by gamma spectrometry. Eight results failed the non parametric test and were designated as outliers. Two outliers were reported by labs 36 and 81 from MORS member state participants.

# 134-137<sub>Cs</sub>

 $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  concentrations were reported by 76 and 72 investigators, respectively. Except for iab 65, all participants determined the concentration of the two radionuclides by gamma spectrometry. Six values were rejected as outliers including the one low concentration from lab 65. A t-test was used to compare the sample means of the global data set and the MORS data set. The correlation among the sets of results was significance showing that the two samples (MORS and Global data sets) come from a population of data with the same mean. This was also the case when the two sets of results for  $^{40}\text{K}$  were compared.

# 90<sub>Sr</sub>

Only 18 measurements of 90Sr were provided for the Baltic Sea sediment of which 14 were accepted after removing outliers. One value from MORS participating lab #33 was approximately 20 times higher than the median value computed from the accepted results.

### Plutonium 239+240 and Americium 241

Plutonium was determined by 30 laboratories and only 4 values were rejected at outliers. No results from the MORS participants were rejected.

The situation for 241 Am is less satisfactory. The mean (median) value of 1.9 Bq/kg shown for 241 Am in table 1 (global data set) was generated after removing 9 less than numbers and outliers from the combined alpha and gamma spectrometry results. A t-test was used to compare the mean value for 241 Am measured directly by gamma spectrometry with the value determined by alpha spectrometry following radiochemicai separation. The

mean (and median) value determined from the concentrations measured by gamma spectrometry (2.94) was higher than the mean value determined by alpha counting (1.83) and the probability of the two data sets coming from the same population of results is very small (significance .053). There appears to be a systematic difference between the mean values for the low concentration of  $^{241}$ Am determined by the different analytical methods. At this time we recommend that the computed median value for  $^{241}$ Am shown in table 1 be viewed as an information value. Most results from the MORS participants were determined by gamma spectrometry.

### Radlonuclides in the Uranium-238 and Thorium-232 Decay Series

The well known daughter radionuclides of the  ${}^{238}$ U decay series are shown in table 3. There was 15 reported measurements of  ${}^{238}$ U by alpha spectrometry along with 8 determinations of the  ${}^{234}$ U alpha activity in 7 instances the gamma activity from the decay of  ${}^{234}$ Th( $t_{1/2}$ =24.1d) was measured and there were 7 determinations of the  ${}^{230}$ Th alpha activity Outliers from the data sets were rejected and t-tests were conducted to compare computed mean values between the first 4 members of the decay chain. The  ${}^{238}$ U shows a strong correlation with  ${}^{234}$ Th, the most immediate daughter, but is poorly correlated with the  ${}^{234}$ U (significance = .005). Likewise the  ${}^{230}$ Th activity is taken from a population of data different from that of  ${}^{234}$ U. These analytical results indicate that  ${}^{234}$ U is enriched over both its parent ( ${}^{238}$ U) and daughter ( ${}^{230}$ Th) in the sediment.

The 25 reported results for  $^{226}$ Ra were generated either by the radon emanation technique or by direct measurement of the 186 keV photopeak from the decay of  $^{226}$ Ra. Another 34 participants measured the gamma activity from the  $^{214}$ Pb or  $^{214}$ Bi daughter decay products. We assume equilibrium exists among all 226Ra daughter products in the sample but a t-test indicates that the  $^{226}$ Ra data set and the  $^{214}$ Pb-Bi daughter data set are from different populations; the mean from the daughter gamma data being 23% lower than the mean of the  $^{226}$ Ra activity measured directly. We have no immediate explanation for this difference. Again we suggest that these "best" median values be used only for information at this time.

210 Pb was determined from the direct measurement of the 47 keV gamma activity by gamma spectrometry and, assuming equilibrium, from the 210 Po alpha activity following radiochemical separation. Both data sets are correlated and come from populations with the same mean. Therefore the best value for the 210 Pb concentration is generated from the combined results generated by both methods. The best values are for: mean,  $439\pm100$ ; median, 434; and a confidence internal of 384-489.

Excellent correlations are evident among the  $^{232}$ Th decay series radionuclides determined both by alpha counting following radiochemical separations and by direct gamma counting techniques. In this sample of Baltic Sea sediment all the  $^{232}$ Th daughter products appear to be in equilibrium with the parent radionuci ide.

### Table 3. Uranium Decay Products

238U (alpha 4.15 & 4.20 MeV)

234Th (gamma .063 & .093 MeV; 3.5 & 4%; & B)

**234mPa** (B)

234U (alpha 4.72 & 4.77 MeV)

230Th (alpha 4.62 & 4.68 MeV)

226Ra (aipha 4.60 & 4.72 MeV; gamma 0.186-4%)

222Rn (alpha 5.49 MeV)

218Po (alpha 6.00 MeV)

214Pb (gamma 0.295- 19%; 0.352-36%;&ß)

214Bi (gamma 0.609-47%;1.12-17%;1.76- 17%; &B)

214Po (alpha 7.69 MeV)

**21**0Pb (gamma 0.047-4%; &B)

**210Bi** (ß)

2 1 OPo (alpha 5.305 MeV)

206Pb stable

### Conclusion

Although a few outliers were identified in the MORS data set, the majority of results reported are of reasonable quality and in agreement with the global set of results, There appears to be some unresolved differences in concentrations for specific radionuclides determined by more than one detection technique. This is not a problem unique to the MORS data set but applies to all reported results for radionuclides such as  $^{241}$ Am and  $^{226}$ Ra (and daughters). The results in this report may be subject to some further small revision that will appear in the final report for IAEA-306.

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#### BALTIC SEA ENVIRONMENT PROCEEDINGS

- No. 1 JOINT ACTIVITIES OF THE BALTIC SEA STATES WITHIN THE FRAMEWORK OF THE CONVENTION ON THE PROTECTION OF THE MARINE ENVIRONMENT OF THE BALTIC SEA AREA 1974-1978 (1979)\*
- No. 2 REPORT OF THE INTERIM COMMISSION (IC) TO THE BALTIC MARINE ENVIRONMENT PROTECTION COMMISSION (1981)
- No. 3 ACTIVITIES OF THE COMMISSION 1980
   Report on the activities of the Baltic Marine Environment
  Protection Commission during 1980
   HELCOM Recommendations passed during 1980
  (1981)
- No. 4 BALTIC MARINE ENVIRONMENT BIBLIOGRAPHY 1970-1979 (1981)
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No. 19 BALTIC SEA MONITORING SYMPOSIUM Tallinn, USSR, 10-15 March 1986 (1986)

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- No. 21 SEMINAR ON REGULATIONS CONTAINED IN ANNEX II OF MARPOL 73/78 AND REGULATION 5 OF ANNEX IV OF THE HELSINKI CONVENTION National Swedish Administration of Shipping and Navigation; 17-18 November 1986, Norrkoping, Sweden (1987)
- No. 22 SEMINAR ON OIL POLLUTION QUESTIONS 19-20 November 1986, Norrkoping, Sweden (1987)
- No. 23 ACTIVITIES OF THE COMMISSION 1986
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- No. 26 ACTIVITIES OF THE COMMISSION 1987
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- No. 27B GUIDELINES FOR THE BALTIC MONITORING PROGRAMME FOR THE THIRD STAGE; PART B. PHYSICAL AND CHEMICAL DETERMINANDS IN SEA WATER (1988)
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<sup>\*</sup> out of print

- No. 29 ACTIVITIES OF THE COMMISSION 1988
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- No. 30 SECOND SEMINAR ON WASTEWATER TREATMENT IN URBAN AREAS 6-8 September 1987, Visby, Sweden (1989)

 BALTIC SEA ENVIRONMENT
 THREE YEARS OBSERVATIONS OF THE LEVELS OF SOME RADIONUCLIDES IN

 PROCEEDINGS No. 31
 THE BALTIC SEA AFTER THE CHERNOBYL ACCIDENT

 Seminar on Radionuclides in the Baltic Sea, 29 May 1989,
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