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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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 ^{90}Sr and/or plutonium isotopes followed for the greater part of the samples. ^{90}Y-oxalate precipitates were measured with low-background gas flow beta-counters (counted 4 times 500 min). For the measurement of ^{238}Pu and $^{239/240}Pu$ 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 ^{137}Cs, ^{134}Cs and ^{110m}Ag. The results for these nuclides as well as for the nuclides ^{90}Sr, ^{238}Pu and $^{239/240}Pu$ 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 cesium 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 ^{137}Cs increased until Dec. 1988, but not ^{134}Cs. 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 ^{110m}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 ^{137}Cs in liver. Despite of its physical half-live of only 250 days, ^{110m}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.

^{110m}Ag 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.

^{90}Sr 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 ^{90}Sr 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 ^{90}Sr 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 ^{90}Sr *(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 ^{238}Pu and $^{239/240}Pu$ 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 ^{90}Sr 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 ^{90}Sr and plutonium. The results for nuclides not already given for unpooled samples in tables 1 to 3 are presented in table 4. The nuclides ^{60}Co, ^{65}Zn and ^{108m}Ag were detected with very low activities.
Especially the long-lived ^{108m}Ag, 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 ^{110m}Ag. The mean value of $^{110m}Ag/^{108m}Ag$ was 2100 (13% la SD, referenced to the Chernobyl accident). ^{108m}Ag 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 ^{110m}Ag and ^{108m}Ag 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 $^{110m}Ag/^{108m}Ag$ depends on the activation duration; a value of 2100 would correspond to an silver activation of roughly half a year.
The mean of the ^{90}Sr activity is slightly lower than in cod fillet. ^{65}Zn was detected weakly in three samples from stations belonging to areas D and E (see fig. 1). ^{238}Pu and $^{239/240}Pu$ (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, $^{239/240}Pu$ 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 ^{137}Cs and ^{134}Cs 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 ^{137}Cs 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 ^{137}Cs 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, ^{134}Cs 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 ^{137}Cs, ^{134}Cs and ^{110m}Ag being detected in mussels and the sample of seastars, the nuclides ^{60}Co, ^{106}Ru and ^{125}Sb 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 ^{134}Cs and ^{137}Cs 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 ^{90}Sr being lower by at least three orders of magnitude compared to ^{137}Cs, the contribution of ^{90}Sr can be neglected.

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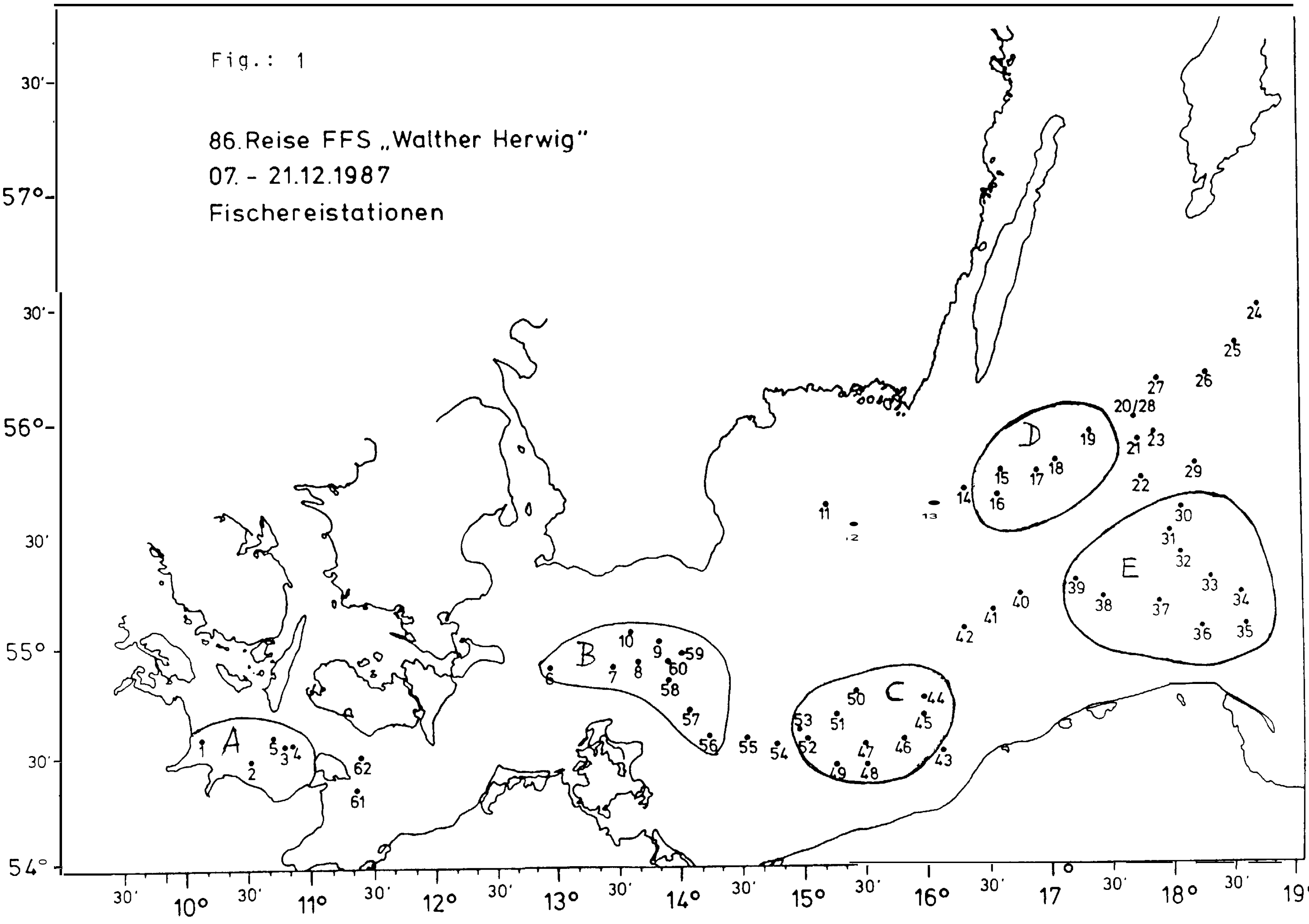

Fig.: 1

86. Reise FFS „Walther Herwig"
07. - 21.12.1987
Fischereistationen

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 Bq/kg ww	max. value Bq/kg ww	Mean Bq/kg ww
Cod: fillet	Sr-90	28	12	< 0.0015	0.0091	0.0040
	cs-134	29	0	0.67	2.6	1.4
(Gadus morhua)	cs-137	29	0	4.4	9.3	5.9
	Pu-238	17	16	< 0.00011	0.00040	< 0.00040
	Pu-239	17	15	< 0.00018	0.0028	< 0.0028
Cod: whole fish	Sr-90	1	0			0.060
	Ag-110m	1	0			0.60
	cs-134	1	0			1.6
	cs-137	1	0			4.8
Cod: liver	Sr-90	3	1	< 0.0010	0.0042	0.0027
	Ag-110m	5	0	1.7	2.6	2.1
	cs-134	5	0	0.21	0.25	0.23
	cs-137	5	0	0.80	1.1	0.91
	Pu-238	2	2	< 0.00014	< 0.00015	
	Pu-239	2	2	< 0.00019	< 0.00023	
Herring: fillet	Sr-90	6	0	0.014	0.22	0.053
	Cs-134	6	0	0.80	1.1	0.93
(Clupea harengus)	Cs-137	6	0	3.5	4.3	3.9
	Pu-238	3	3	< 0.00024	< 0.0011	
	Pu-239	3	3	< 0.00030	< 0.0018	
Herring: whole fish	Sr-90	2	1	< 0.0019	0.021	0.011
	Ag-110m	2	0	0.051	0.087	0.069
	cs-134	2	0	1.0	1.2	1.1
	cs-137	2	0	3.9	4.4	4.2
	Pu-238	1	1	< 0.00045		
	Pu-239	1	1	< 0.00056		
Dab: fillet	Sr-90	1	0			0.048
	cs-134	1	0			1.3
(Limanda limanda)	cs-137	1	0			4.4
Dab: whole fish	Sr-90	1	0			0.11
	Ru-106	1	0			0.31
	Ag-110m	1	0			0.033
	cs-134	1	0			1.1
	cs-137	1	0			3.5
	Pu-238	1	1	< 0.00026		
	Pu-239	1	0			0.0013
Plaice / Flounder	Sr-90	3	0	0.022	0.035	0.030
fillet	cs-134	4	0	0.79	0.89	0.82
(Pleuronectes pl.	cs-137	4	0	2.7	3.5	3.1
and	Pu-238	1	1	< 0.00047		
Platichthys fl.)	Pu-239	1	1	< 0.00078		
Whiting: fillet	Sr-90	1	0			0.0045
	Ag-110m	1	0			0.067
(Merlangius merl.)	Cs-134	1	0			2.0
	cs-137	1	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 Bq/kg ww	max. value Bq/kg ww	Mean Bq/kg ww
Cod: fillet	Sr-90	50	4	< 0.0029	0.013	0.0052
	Ag-110m	62	60	< 0.011	0.041	
(Gadus morhua)	Cs-134	62	0	1.7	7.6	4.0
	Cs-137	62	0	6.5	23.8	13.8
	Pu-238	25	25	< 0.00021	< 0.0036	
	Pu-239	25	25	< 0.00036	< 0.0045	
Cod: liver	Ag-110m	52	0	0.91	10.6	2.74
	Cs-134	52	0	0.31	1.1	0.69
	cs-137	52	0	1.25	3.87	2.36
Cod: gonads	Sr-90	2	1	< 0.013	0.0076	0.0071
	Ag-110m	5	3	< 0.044	0.10	0.054
	cs-134	5	0	1.0	3.0	2.2
	cs-137	5	0	3.8	10.7	7.9
	Pu-238	2	2	< 0.00080	< 0.00090	
	Pu-239	2	2	< 0.0011	< 0.0013	
Cod: skin	Sr-90	7	0	0.23	0.35	0.29
	Ag-110m	14	13	< 0.037	0.096	
	cs-134	14	0	1.4	3.2	2.1
	cs-137	14	0	4.9	10.	7.1
	Pu-238	7	7	< 0.00050	< 0.0022	
	Pu-239	7	6	< 0.00084	0.0031	
Herring: fillet	Sr-90	5	0	0.0046	0.013	0.0088
	cs-134	9	0	0.40	2.6	1.8
(Clupea harengus)	cs-137	9	0	3.2	8.9	6.8
	Pu-238	2	2	< 0.00082	< 0.00089	
	Pu-239	2	1	< 0.0010	0.0030	
Herring:whole fish	Sr-90	2	0	0.064	0.075	0.069
	Ag-110m	2	0	0.11	0.11	0.011
	cs-134	2	0	2.4	2.7	2.6
	cs-137	2	0	8.2	8.8	8.5
	Pu-238	1	1	< 0.00036		
	Pu-239	1	1	< 0.00060		
Dab: fillet	Sr-90	1	0			0.037
	cs-134	1	0			0.92
(Limanda limanda)	cs-137	1	0			4.1
Sugar kelp	Sr-90	1	0			1.4
	Ag-110m	1	0			0.46
(Laminaria saccha.	Cs-134	1	0			5.5
	cs-137	1	0			23.
(Bq/kg dry !!)	Pu-238	1	0			0.012
	Pu-239	1	0			0.063

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

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

Table 4 : Further nuclide activities in pooled samples
of cod liver from December 1907
Samples were pooled over different stations

Sample number	^{60}Co	^{65}Zn	^{90}Sr	^{108m}Ag	$^{239/240}Pu$
	Bq/kg wet weight ± lo SD (X)				
12052	9.26E-3 ±23.	<3.50E-2	3.68E-3 ±27.	9.60E-3 ±10.	<2.5E-4
12053	4.14E-3 f25.	2.87E-2 f23.	2.41E-3 ±21.	6.663-3 ±8.2	<5.1E-5
12054	5.09E-3 ±18.	2.25E-2 ±25.	<2.33E-3	5.82E-3 ±9.0	<7.7E-5
12055	5.23E-3 ±22.	<1.89E-2	2.16E-3 ±14.	6.76E-3 ±9.4	<4.0E-5
12056	5.85E-3 ±18.	<1.71E-2	2.58E-3 ±7.4	5.23E-3 f9.4	<4.9E-5
12057	3.97E-3 ±28.	<2.01E-2	3.88E-3 ±7.0	6.82E-3 ±8.9	<7.7E-5
12058	5.00E-3 ±24.	4.38E-2 ±19.	4.09E-3 ±8.0	8.45E-3 f9.4	1.3E-4±18.

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

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

Date	Area	Stations	^{137}Cs	^{134}Cs	$^{134}Cs/^{137}Cs$
ordered *by* date *and area*					
12/86	A	l-9	8.0	2.3	0.29
	B	12, 58, 59	6.4	1.7	0.27
	C	44-46	5.6	1.4	0.25
	D	27-32	5.9	1.5	0.25
	E	49-56	4.8	0.8	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	l-2	7.7	1.4	0.18
	B	6-8	17.1	3.9	0.23
	C	15-17, 41-43	15.9	3.6	0.23
	D	30-33	19.4	4.5	0.23
	E	47-50,23-24,26-28	19.3	4.4	0.23
ordered *by area and* date					
12/86	A	l-9	8.0	2.3	0.29
12/87		l-5	8.0	2.0	0.25
12/88		l-2	7.7	1.4	0.18
12/86	B	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	C	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	E	49-56	4.8	0.8	0.17
12/87		30-39	14.5	4.2	0.29
12/88		47-50,23-24,26-28	19.3	4.4	0.23

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

Activities in Bq/kg wet or dry ± la SD (%)

Sample	Base	Date	^{60}Co	^{106}Ru	^{110m}Ag	^{125}Sb	^{134}Cs	^{137}Cs
Mussels								
Cyprina islandica	wet	12/88	0.039 ±31.	<0.26	0.36 ±5.2	<0.084	0.084 f13.	0.51 f7.7
Cyprina islandica	wet	12/88	<0.035	<0.24	0.10 ±10.	<0.063	0.070 ±13.	0.36 ±7.9
Seas *tars*								
Asterias rubens	dry	12/88	0.21 f33.	<1.1	0.41 ±11.	<0.32	0.68 f8.5	3.66 ±7.4
Aquatic plants								
Laminaria sacchar. 1)	dry	12/87	<0.24	3.2 ±14.	0.46 ±13.		5.45 ±5.3	22.9 f7.4
Laminaria sacchar.	dry	12/88	0.27 ±25.	3.0 ±18.	<0.16	2.0 ±11.	3.36 ±6.1	16.8 f7.0
Fucus vesiculosus	dry	12/88	2.18 ±6.1	20.7 ±8.0	0.79 ±10.	19. ±5.8	2.27 ±12.	14.7 f7.1

1) see table 2 for ^{90}Sr and plutonium