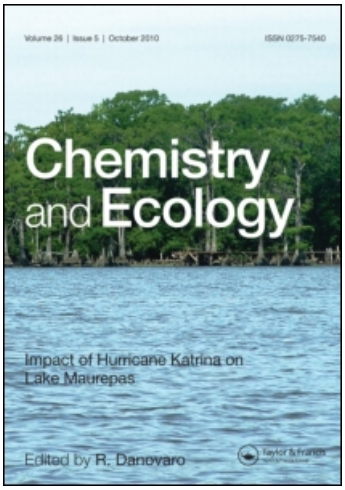

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Radioactivity in the Baltic Sea

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RADIOACTIVITY IN THE BALTIC SEA

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The Baltic Sea is, like the Mediterranean, a marginal sea, which with the Black Sea, were marine environments contaminated from the Chernobyl accident.

Radiocaesium and plutonium isotopes were studied in water, sediment and macroalgae in the Baltic Sea since 1982. The inventory of ^{137}Cs in the Baltic increased from 0.65 PBq to 5.85 PBq following the Chernobyl accident. The corresponding increase for $^{239+240}Pu$ was less significant and yielded 1.5 TBq to a total value of 16.5 TBq.

For plutonium, 98% is trapped in the sediment and the net-exchange of this element through the Baltic straits is very small (1 GBq/year), while for radiocaesium, 45% is in the water phase and there is a net-loss of 60 000 GBq annually into the adjacent water (Kattegatt).

KEY WORDS: Baltic sea, radiocaesium, source term, radionuclide balance.

INTRODUCTION

The Baltic Sea is a semi-enclosed and shallow sea in which dissolved substances will remain for a relatively long period compared to that of other seas. This leads to a greater accumulation of radioactive materials than in other seas of the Atlantic basin. The influence of rivers is relatively large compared to other seas, which will be of importance for the long term balance of radionuclides in the Baltic Sea following an accidental release. Tables I and II give the dimensions and water balance of radionuclides in the Baltic Sea.

It is well known that macroalgae are excellent bioindicators for dissolved species of radionuclides such as ^{99}Tc and ^{137}Cs in sea water and can thus be used to trace water after contamination from nuclear facilities over long distances (Dahlgaard *et al.*, 1986, Aarkrog *et al.*, 1987, Mattsson *et al.*, 1980). In the vicinity of nuclear power plants the pathways and distribution of acitivation products have been investigated with great success particularly by using species of brown algae (*Fucus vesiculosus, F. serratus*) as bioindicators (Carlson, 1990). The macroalgae have been used primarily for studying marine contamination arising from liquid discharges to the sea from nuclear facilities, while the response of the algae to atmospheric deposition has been little investigated. Global fallout of ^{137}Cs and transuranium elements from the testing of nuclear weapons, however, has been investigated but long after the events (Ballestra, 1980).

Table I Dimensions of the Baltic Sea area and its sub areas.

	Area (km^2)	*Volume (km^3)*
Baltic proper	209 200	13 600
Gulf of Riga	18 100	410
Gulf of Finland	29 600	1 130
Bothnian Sea	80 300	4 950
Bothnian Bay	36 800	1 490
Kattegatt + Belt Sea	43 900	800
Total	417 900	22 380

Table II The water balance of the Baltic Sea.

	$km^3\ a^{-1}$
Precipitation	+ 200
Evaporation	− 200
Inflow through Danish Straits	+ 433 (S ‰ 17–19)
Outlfow through Danish Straits	− 866 (S ‰ 8–9)
Run-off	+ 433

MATERIALS AND METHODS

Random samples of *F. vesiculosus* have been collected in Sweden during 1982, 1983, 1986, 1987 at different localities from Simpenäs (1) to Saltö (40) (Fig. 1). During July 1991, the sampling was repeated at the different localities on the Swedish east, south and west coasts as well as on Gotland. The samples of *Fucus* (1–3 kg wet weight) were generally collected randomly within an area of a few hundred metres.

In addition, since 1967 *Fucus* sp. have been collected yearly, and during later years monthly, at Särdal on the Swedish west coast (56.76 °N, 12.63 °E) since 1967.

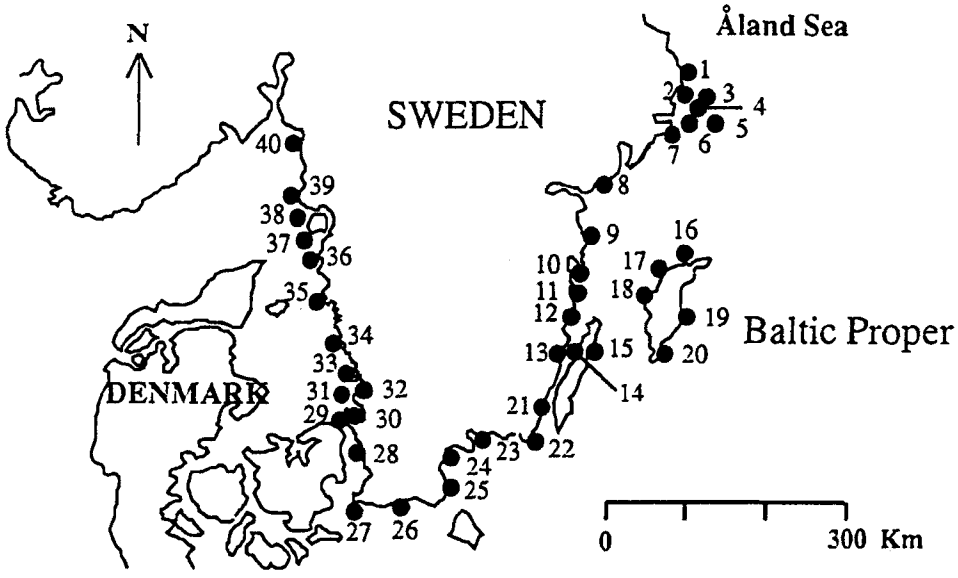
During the Gauss expedition, 1983, an extensive sampling of sediment was performed. Twenty eight stations in the Baltic Sea were covered. The sediment cores were sliced in 3 cm sections on board.

The algae and sediments were air-dried and ground after return to the laboratory. Water samples were preconcentrated on board by co-precipitation of plutonium as hydroxides and radiocaesium with ammonium molybdophosphate. Plutonium was determined after radiochemical separation and alpha spectrometry using solid state silicon detectors (Holm, 1984), while radiocaesium was measured with Ge-gamma spectrometry.

RESULTS AND DISCUSSION

Radiocaesium, ^{137}Cs ($T_{1/2} = 30$ a)

Pre-Chernobyl data (1982–1983) for *Fucus* were for ^{137}Cs about 10–15 Bq kg^{-1} dry weight in *F. vesiculosus* (IAEA, 1986), and in surface sea water 12–15 Bq m^{-3} on the



1 Simpnäs	12 Dragskärbadet	22 Svanhalla hamn	33 Görvik
2 Furusund	13 Revsudden	23 Matvik	34 Glommen
3 Bromskär	14 Ekerum	24 Landön	35 Getterön
4 Glyxnäs	15 Kapelludden	25 Baskemölla	36 Lerkil
5 Sandhamn	16a Ainesriv	26 Svarte	37 Marstrand
6 Stavsnäs	16b Hallshuk	27 Kämpinge	38 Gullholmen
7 Dadlarö	17 Stenkyrkehuk	28 Barsebäck	39 Smögen
8 Hummelvik	18 Ygne	29 Lerhamn	40 Saltö
9 Fyrudden	19 Grogarn	30 Arild	
10 Horns udde	20 Grunnet	31 Torekov	
11 Kråkelund	21 Ekenäs	32 Kattvik	

Figure 1 Sampling localities of *Fucus vesiculosus* along the Swedish coast during May-August 1986.

Swedish east coast and 15–30 Bq m^{-3} on the west coast. The higher salinity bottom water originating from the North Sea contaminated from European reprocessing facilities showed higher concentrations (a factor of 2) especially on the west and south coast.

Until 1986, activity concentrations on the Swedish west coast reflected releases from European reprocessing facilities with a delay of about 4 years (Mattsson and Erlandsson, 1991). Due to decrease of release rates from the Sellafield plant in the Irish Sea, activity concentrations of radiocaesium at the Swedish west coast also decreased significantly after 1982. It has been possible to detect radiocaesium (but with difficulty) in the bottom water up to Gotland (Duniec *et al.*, 1984), hence the major activity in the Baltic proper and the Bothnian Bay up to 1986 came mainly from nuclear test fallout where outflow from rivers played an important role. The coincidence that the activity concentrations in *Fucus vesiculosa* were about the same

in different areas, in spite of different water concentrations, is a result of higher concentration factor at lower salinity.

Following the Chernobyl accident, activity concentrations increased over all areas but especially in the Åland Sea where we have our most northern location for collection of *Fucus*. The actual deposition over the sea was not recorded but it is well known that the land area adjacent to the Åland Sea received the highest deposition. In Figure 2, it can be seen that up to 600 Bq kg^{-1} of ^{137}Cs was recorded in *Fucus vesiculosus*. The activity concentrations can reflect differences in the deposition pattern but also runoff during May-August 1986. The deposition was rather "patchy" and single hot spots were observed.

Before the Chernobyl accident, the total inventory of ^{137}Cs in the Baltic Sea was estimated to be about 650 TBq of which 55% was present in the water column. In 1986 this value rose to about 5 200 TBq in the water and 650 TBq in the sediments. The corresponding figures for 1987 were 3 800 TBq and 1 200 TBq respectively. The total lower inventory for 1987 can be explained by outflow to the North Sea but reflect also the uncertainties in such estimations.

The *Fucus* samples were collected in July 1986, in August-September 1987 and in July 1991. We know that there is a strong seasonality for radiocaesium in *Fucus* with maxima during spring-summer and minima during the winter (Mattsson and Erlandsson, 1991) reflecting biological activity/uptake. August-September values would accordingly be slightly lower than for samples collected in July.

It is interesting to note that activity concentrations were higher in 1987 than in 1986 except for the Åland Sea. The anti-clockwise circulation of the Baltic Sea would move activity from the Åland Sea down along the Swedish east coast. From the Swedish south coast, activity concentrations of ^{137}Cs in *Fucus* are decreasing with a semiexponential relationship with distance northwards along the west coast (location 26–40). This is the opposite, compared to the situation before the Chernobyl

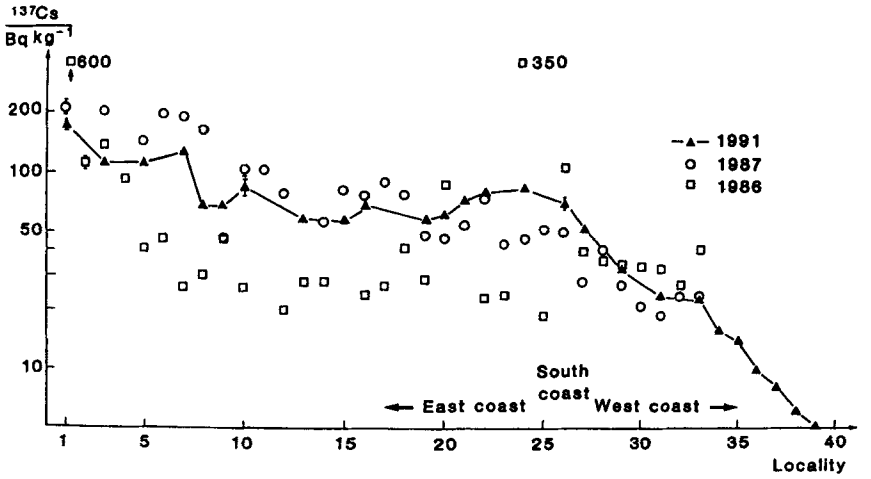


Figure 2 Activity concentrations of ^{137}Cs in *Fucus vesiculosus* along the Swedish coast in July 1986, August-September 1987 and July 1991.

accident when the major activity along this coast originated from European reprocessing facilities. It has also been shown that concentrations in the outgoing surface water at present are higher than in the inflowing bottom water. We therefore have a situation with a net outflow of radiocaesium from the Baltic Sea to the North Sea in contrast to that before the accident (Aarkrog and Hansen, 1991).

Similar studies were performed in the Mediterranean showing a rather rapid decrease of activity concentrations in water and *Fucus* for all Chernobyl derived radionuclides showing a mean residence time in water of about 20 days only and an ecological half-life of 60–120 days (70 days for radiocaesium) in different macroalgae (Holm, 1994). The normal mean residence time of radionuclides in *Fucus* when translocated to "clean" water after contamination is about 100 days (Dahlgaard, 1994).

No such conclusions can be taken from our studies of *Fucus* and water during 1986–1991. The outflow from rivers is much more important for the radionuclide balance in the Baltic Sea compared to the Mediterranean which has a total volume 180 times that of the Baltic Sea. Furthermore, the anti-clockwise circulation in the Baltic Sea brings activity released from the largest rivers (Neva, Vistula, Daugava, Neman, Kemijoki and Oder) situated south-east and east of Sweden to the east coast of Sweden. These rivers drain a total area of 733 000 km^2 and have a total outflow of 6 000 m^3s^{-1} which corresponds to about half of the total run-off. In addition, the inflow to/ventilation of the Baltic Sea from the North Sea has been relatively small until recently, e.g. the winter 1992/93 when we had a significant ventilation.

Radiocaesium was determined in water from 11 sites where *Fucus* was collected and the results are given in Figure 3. The activity concentrations in water follow the same trend as the results for *Fucus*, showing the highest concentration at location 1 (177 mBq l^{-1}) and the lowest at the west coast, location 40 (28 mBq l^{-1}). At one location, Kämpinge (location 27), a more detailed study was performed during 1987 and 1988. Those results showed that the ^{137}Cs activity concentration in water varied between 60 and 140 mBq l^{-1} with distinct maxima during April-July. The concentrations in *Fucus* essentially followed the changes in the water concentration with minima during September-October (25 Bq kg^{-1}) and maxima during May-June (80–90 Bq kg^{-1}). From the same study, the observed activity concentration ratios *Fucus*/water at that particular location were between 400 and 700 (based on dry weight for *Fucus*), with the highest values during June-August. This is in perfect agreement with the value 700 for July, 1991 obtained in this study.

From Figure 3 we observe the activity concentration in water, *Fucus* and the observed activity ratio *Fucus*/water at the 11 locations. It is obvious how the decreasing water concentrations and increasing salinity along the west coast work in the same direction to result in a strong decrease of the concentrations of radiocaesium in *Fucus* from locality 25 to 40.

Salinity concentrations in the Baltic Sea vary with season, with the highest values during winter and the lowest during summer. It has been shown that the concentration factor, activity ratio *Fucus*/water at steady state, is a factor 5 higher at a salinity of 8‰ (location 27) compared to 24‰ at location 40. This is in agreement with our findings.

In Figure 4 the present balance for ^{137}Cs in the Baltic Sea is given.

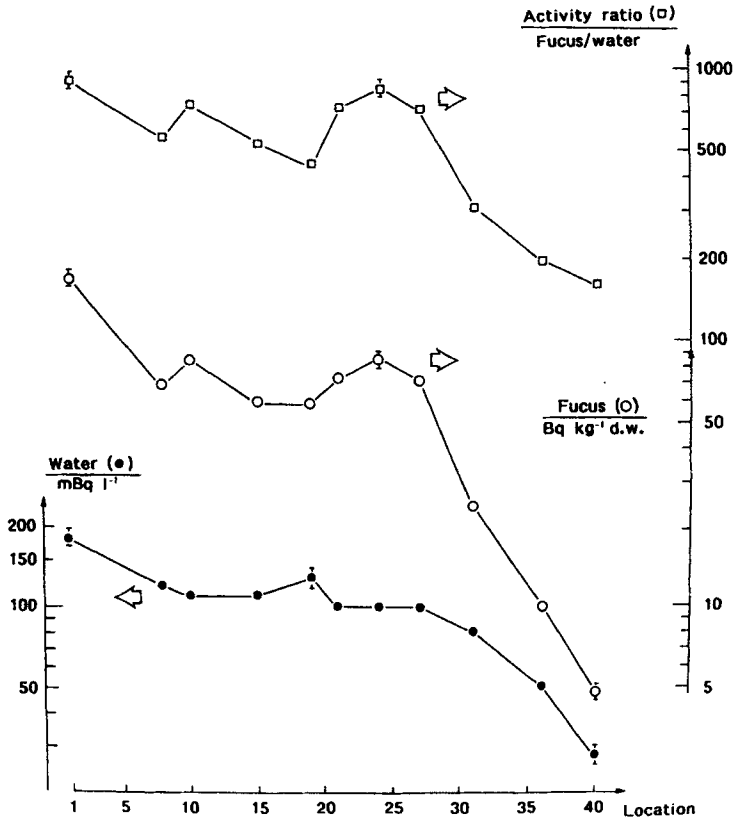


Figure 3 Activity concentration of ^{137}Cs in *Fucus vesiculosus*, activity concentration of ^{137}Cs and the observed activity ratio *Fucus*/water along the Swedish coast in July 1991. The arrows indicate respective axis (concentration in *Fucus* or water) to be used.

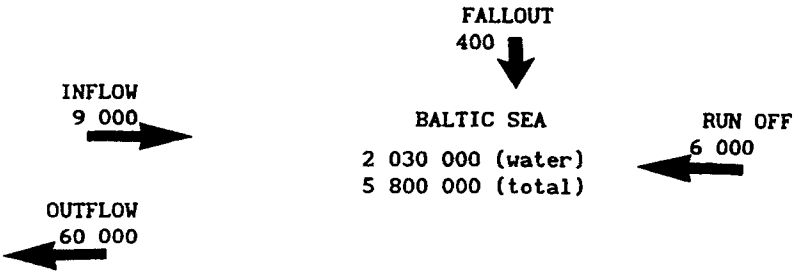


Figure 4 Balance of ^{137}Cs in the Baltic Sea (GBq).

Caesium, ^{134}Cs ($T_{1/2} = 2.05$ a)

Different values for the activity ratio $^{134}Cs/^{137}Cs$ in Chernobyl fallout have been given. The differences are probably to some extent due to systematic errors unable to correct for coincidence effects during gammaspectrometry. We will here use the ratio of 0.54 on April 26, 1986 based on different samples at different distances from Chernobyl given by Aarkrog (1988). For *Fucus* collected in July 1986 the average value was 0.44 ± 0.01 ($n = 32$, 1 S.E) and in August-September 1987 0.35 ± 0.01 ($n = 33$, 1 S.E.) and in July 1991 0.095 ± 0.02 ($n = 25$, 1 S. E.). The theoretical values in Chernobyl fallout at these times corrected for physical decay are 0.50, 0.35 and 0.11, respectively. In water collected in July 1991 we found an average ratio of 0.091 ± 0.01 ($n = 11$, 1 S. E.).

The results are displayed in Figure 5 and it is obvious that in July 1986 the fraction originating from nuclear test fallout can be demonstrated and is calculated as an average of 13% of the total ^{137}Cs. During 1987 the data indicate that essentially all radiocaesium originated from the Chernobyl accident except for some fraction on the west coast where the activity ratios are lower than what is derived from decay corrected Chernobyl fallout. The truth is, of course, that some ^{137}Cs from the nuclear tests during the late 1950's and early 1960's still remain in the Baltic Sea, but was in 1987 masked by the signal from Chernobyl fallout. During 1991 a significant fraction of ^{137}Cs originated from nuclear test fallout.

Plutonium, $^{239+240}Pu$ ($T_{1/2} = 24410$ a, 6550 a)

The source term of plutonium can be identified by isotopic ratios. The $^{238}Pu/^{239+240}Pu$ activity ratio in nuclear test fallout is about 0.025. In addition, the

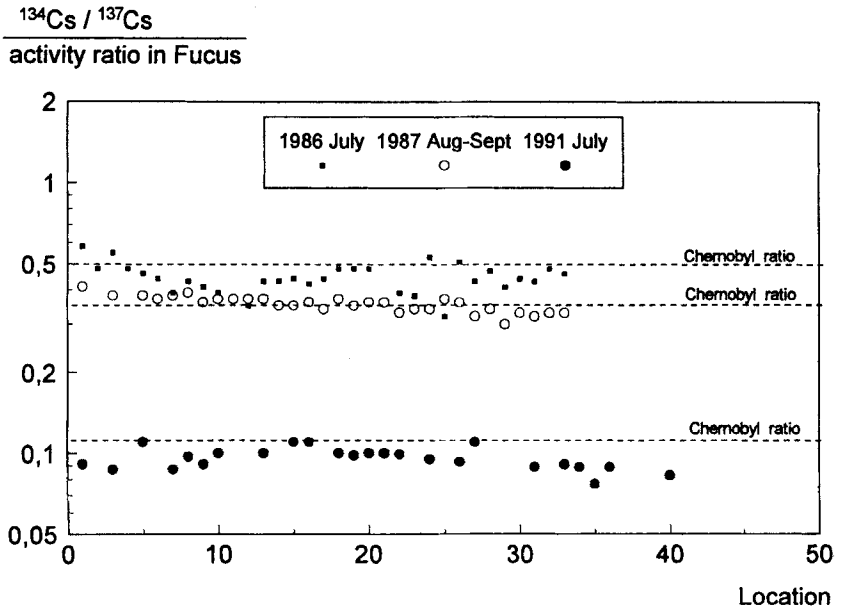


Figure 5 The activity ratio $^{134}Cs/^{137}Cs$ in *Fucus vesiculosus* along the Swedish coast in July 1986, August-September 1987 and July 1991.

satellite failure of SNAP 9A in 1964, over the Mozambique channel, increased the $^{238}Pu/^{239+240}Pu$ in world-wide fallout to a total value of about 0.045 on the Northern hemisphere. This should be compared with a value of 0.25 in releases from nuclear fuel reprocessing plants, 0.47 in Chernobyl fallout and 0.016 in weapon grade plutonium. The corresponding activity ratios for $^{241}Pu/^{239+240}Pu$ are 16, 25, 86 and 4, respectively (Holm, 1988).

The major source for plutonium in the Baltic Sea is nuclear test fallout although other sources can also be identified. The area deposition of $^{239+240}Pu$ in the Baltic Sea area is around 40–50 Bq m^{-2} or the inventory 16–18 TBq for the whole Baltic Sea.

The influence of plutonium on the Swedish west coast from European reprocessing plants can be expected to have been much smaller than for radiocaesium. Only a minor fraction (4%) of the releases of plutonium are present in the soluble oxidized forms (+V, +VI) (Pentreath *et al.*, 1985). Fallout of refractory elements such as plutonium in fallout from the Chernobyl accident was very small compared to that for radiocaesium. In the most contaminated areas of Sweden, the total integrated deposition increased with a factor of 2 compared to 50–100 for ^{137}Cs and in "low contamination" areas it increased by 1% or less (Holm, 1991). This means that the major source for plutonium in the Baltic Sea for most areas is recirculating activity from nuclear tests.

In the Baltic Sea 99% of plutonium is rapidly transferred to and associated with the sediments. Analysis of Fe/Mn concretions from the Baltic Sea show no preferential association of plutonium in these nodules over surrounding sediments (Sanchez *et al.*, 1988). Other inorganic and organic scavenging processes play a more important role.

Plutonium concentrations in water are low in accordance to the transfer of plutonium to sediments. Before the Chernobyl accident they were about 4 μBq l^{-1} on the east coast and about 8 μBq l^{-1} on the west coast. This gives concentration factors for *Fucus* (dry)/water of 42 000 on the east coast and 12 500 on the west coast. As for radiocaesium, concentration factors for plutonium increase at lower salinity but are not that pronounced. Applying these data on the results for *Fucus vesiculosus* collected in 1991 gives water concentrations between 0.6–2 μBq l^{-1} on the east coast and 2–6 μBq l^{-1} on the west coast in agreement with data from Finland (Saxén *et al.*, 1989, Ilus *et al.*, 1993). Plutonium was not measured in the water samples collected in 1991.

In Table III, the activity concentrations of plutonium in *Fucus vesiculosus* collected at the sampling stations in 1982, 1983, 1986, 1987 and 1991 shown in Figure 1 are given. The activity concentrations in *Fucus vesiculosus* were for example, 100 ± 30 mBq kg^{-1} on the west coast and 171 ± 50 mBq kg^{-1} on the east coast in 1983, before the Chernobyl accident. The corresponding values for 1986 were 49 ± 7 and 75 ± 10 Bq kg^{-1}, for 1987, 43 ± 4 and 63 ± 6 Bq kg^{-1} and for 1991, 54 ± 5 and 48 ± 6 Bq kg^{-1}. It is obvious that the Chernobyl accident had no significant impact regarding plutonium in *Fucus* and consequently not on concentrations in water.

The results from plutonium in *Fucus serratus* at a sampling site on the Swedish west coast during 1967–1992 are displayed in Figure 6. As can be seen, the concentrations decreased from 500–600 mBq kg^{-1} dry weight 1967–1970 to 50–100 mBq kg^{-1}

Table III $^{239+240}$Pu in *Fucus vesiculosus* along the Swedish coast in 1982, 1983, 1986, 1987, and 1991 (mBq kg^{-1} dry weight). Estimated total analytical errors are about 20%.

Site No.	*1982 Oct*	*1983 May/June*	*1986 July*	*1987 Aug/Sept*	*1991 July*
1	21		120	56	57
2			35		
3			121	57	50
4			58		
5			81	36	26
6			42	25	
7	54		100	51	48
8	53		69	49	30
9	83	175	75	36	43
10	98		64	45	25
11	110	152		90	
12	47	382	82	45	
13	61	137	113	74	85
14	54	181	75	86	
15		210	62	84	65
16a			38	47	31
16b					15
17			30	67	
18			31	60	31
19			40	43	70
20			96	156	69
21	139			111	85
22	193	214	52	82	49
23	180	108	45	43	
24	51		268	44	25
25	39	83	36	44	
26	34		40	40	30
27	65		54	46	41
28		49	26		
29		115	55	27	23
30			54	67	–
31			45	35	50
32		70	36	31	–
33		259	75	47	33
34		120			55
35		81			71
36					40
37					43
38					84
39		77			51
40		53			

in 1980. This value has then been more or less constant since then. The source of $^{239+240}$Pu in the late sixties was mostly nuclear test fallout while European reprocessing facilities had a more significant impact on the Swedish west coast during the late seventies and early eighties. The transit time from Sellafield to the Swedish west coast is estimated to be 3 years (Dahlgaard *et al.* 1986).

The total inventory of plutonium in the Baltic Sea in 1986 was estimated to be 16.5 TBq of which only 1.5 TBq originated from the Chernobyl accident. Nevertheless, we would expect to observe an increase in activity concentrations in *Fucus*,

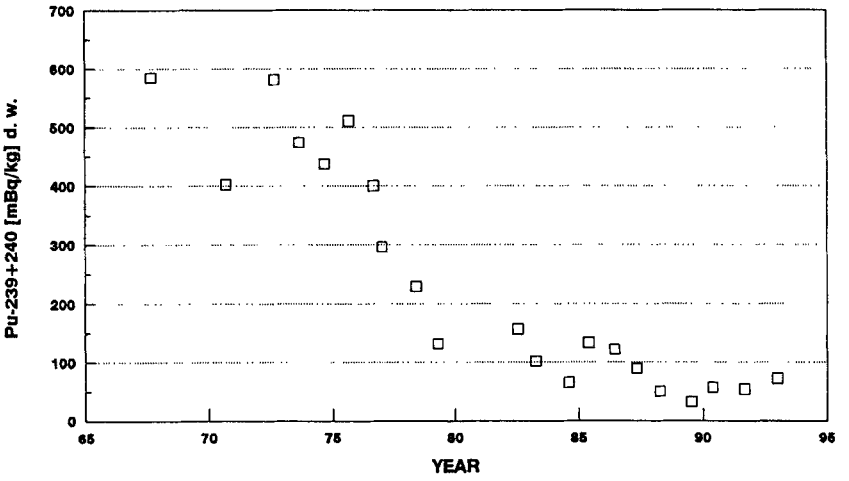


Figure 6 Activity concentrations of $^{239+240}$Pu in *Fucus serratus* at Särdal on the Swedish west coast during 1967–1992. Total errors about 20%.

at least in the most contaminated areas, but this does not seem to be the case, i.e., the increase is not statistically significant even directly after the accidental deposition, in July 1986, 3 months later.

We also know that only about 1% of plutonium was present in the water column before the accident compared to 56% for ^{137}Cs. After the accident, during the summer 1986, over 80% of ^{137}Cs was present in the water column (Dahlgaard *et al.*, 1986). Furthermore, outflow of plutonium from rivers is much less important and/or is associated with particles, or will become so rapidly. The plutonium will thus be incorporated into the sediments and is not available to biota such as *Fucus*.

The maximal concentrations of plutonium are found at a depth of 4–10 cm in undisturbed sediments compared to a depth of 1–2 cm for radiocaesium. The maxima of plutonium correspond to the time of fallout from nuclear explosion tests and shows the relatively small impact of plutonium from the Chernobyl accident. Inventories are generally higher in sediments than in integrated fallout on land, i.e., 40–130 Bq m^{-2}. This is again in agreement with Finish data.

On basis of our knowledge, the present radionuclide balance of plutonium in the Baltic Sea can be assessed as in Figure 7. The figure is based on data for plutonium in river water, levels in precipitation, and total levels in/out in flowing in surface water and bottom water. If levels were constant with the present input, the residence time for plutonium in the Baltic Sea is about 9 years. We have not taken resuspension and remobilisation from sediments into account.

Plutonium, ^{238}Pu ($T_{1/2} = 88$ a) and ^{241}Pu ($T_{1/2} = 14.5$ a)

The assessment of ^{238}Pu is rather difficult since the alpha particle energy is almost equal to that for ^{228}Th generally present in much larger amounts even after radiochemical separation. ^{228}Th might interfere in the α-spectrometry.

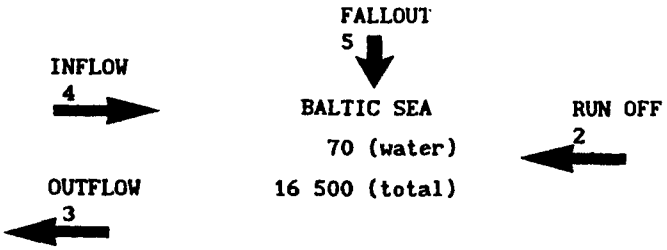


Figure 7 Balance of $^{239+240}$Pu in the Baltic Sea (GBq).

Table IV Inventories of plutonium in the Baltic Sea (GBq).

	Nuclear test fallout	*Chernobyl*	*Total*
$^{239+240}$Pu	15 000	1 500	16 500
^{238}Pu	675	725	1 400
^{241}Pu	60 000	129 000	189 000

The assessment of ^{241}Pu is also rather difficult but can be done by beta counting or by alpha particle measurement of the daughter product ^{241}Am. Nevertheless, as described above, both ^{238}Pu and ^{241}Pu are tools for determining the source term.

The ^{238}Pu/$^{239+240}$Pu ratio in *Fucus* samples from the Särdal site during 1972–1977, was 8.0±0.5% (corrected for decay of ^{238}Pu back to 1975) and was 6.7±0.4% in 1982. This indicates that about 30% of the plutonium at that time at the Swedish west coast in water (and *Fucus* sp) originated from European reprocessing facilities.

Values for ^{238}Pu in *Fucus* sp. on the Swedish east coast ranged from 4–16% of those for $^{239+240}$Pu and should be taken with great precaution and not directly be translated to be influenced significantly from the Chernobyl accident. In a later study we will combine several samples from the same area in order to improve counting statistics and improve decontamination from ^{228}Th.

Plutonium-241 was measured in pre-Chernobyl samples of different matrices but only in air-filters and precipitation in samples contaminated from the Chernobyl accident. Data shows that the activity ratio ^{241}Pu/$^{239+240}$Pu was 16 in fresh fallout from nuclear testing, perhaps about 4 today, but as high as 86 in Chernobyl fallout based on activity ratios. Table IV gives the inventories of plutonium isotopes in the Baltic Sea.

CONCLUSIONS

The investigation shows that it is possible to use *Fucus* sp. as a bioindicator for the study of turnover of radionuclides in the Swedish coastal zone after the Chernobyl accident.

The results show the importance of outflow from rivers to the Baltic for the radionuclide balance. The large catchment area is contaminated from the Chernobyl

accident and will provide radiocaesium (and radiostrontium) to the Baltic Sea over several years. The anti-clockwise circulation in the Baltic Sea enhances these effects and supplies radioactivity to the central Sea from the large rivers draining the Baltic States and Russia. The ecological residence time is long and further investigations are needed so as to provide a model for the turnover of radionuclides in the Baltic Sea. This model is of great importance for the estimation of the radiological impact to the Baltic Sea and its catchment area.

It is also obvious that a significant fraction of radiocaesium from rivers entering the Baltic Sea in a soluble form or is dissolved in the interphase between fresh and saline water. The impact of plutonium from the Chernobyl accident was small and plutonium is rapidly associated with the sediments.

At present we have a significant net outflow of radiocaesium to the North Sea by the Baltic surface water which has higher concentrations than the more saline bottom water, while plutonium is mainly trapped in the sediments in the Baltic Sea. This is an opposite situation to that before the Chernobyl accident when the North Sea provided a net inflow of radioactivity to the Baltic Sea by bottom water contaminated from European reprocessing facilities.

Acknowledgement

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