Pre- and post-Chernobyl accident levels of ^{129}I and ^{137}Cs in the Southern Baltic Sea by brown seaweed *Fucus vesiculosus*

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ABSTRACT

^{129}I is a very long-lived radionuclide ($T_{1/2} = 15.7 \times 10^6$ years) that is present in the environment both because of natural and anthropogenic sources. In this work ^{129}I concentration and ^{129}I/^{127}I ratio have been determined in seaweed *Fucus vesiculosus* collected in the Southern Baltic Sea during 1982 and 1986 (post-Chernobyl accident). The resulting data were evaluated in terms of ^{129}I concentrations, ^{129}I/^{127}I and ^{129}I/^{137}Cs ratios. ^{129}I concentrations were found to be in the order of $(0.82–5.89) \times 10^9$ atoms g^{-1} in 1982 and $(1.33–38.83) \times 10^9$ atoms g^{-1} in 1986. The ^{129}I/^{127}I ratios ranged from $(22.7–87.8) \times 10^{-10}$ for seaweed collected in 1982 and from $(26.1–305.5) \times 10^{-10}$ for seaweed collected in 1986. Also a linear relationship was established for ^{127}I concentrations in seawater and salinity in this area, enabling the estimation of concentration factors for ^{127}I in *F. vesiculosus*.

The high levels of ^{129}I and ^{129}I/^{127}I in the Kattegat and their gradually decreasing trend to the Baltic Sea indicates that the most important contribution to the ^{129}I inventory in the Baltic Sea area comes from Sellafield and La Hague reprocessing plants. With respect to Chernobyl accident, ^{129}I concentrations in samples collected in 1986 were not much higher than those expected in less contaminated samples from 1982. This supports the view that the contribution of the Chernobyl accident to ^{129}I in the Baltic region was not significant.

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1. Introduction

^{129}I is a long-lived cosmogenic radionuclide ($T_{1/2} = 15.7 \times 10^6$ years) for which the natural abundances have been altered in a significant way by human actions. In nature, it is produced by cosmic rays interactions with Xe in the atmosphere, by spontaneous fission of ^{238}U and ^{235}U in the lithosphere (Eisenbud and Gesell, 1997) and, to a minor extent, by neutron-induced reactions on ^{128}Te and ^{130}Te in the Earth crust (NCRP, 1983). These processes lead to typical isotopic ratios ^{129}I/^{127}I between 10^{-12}

(Kilius et al., 1992) and 6×10^{-13} (Fabryka-Martin et al., 1985). The natural inventory of ^{129}I has been estimated to be ~230 kg, most of which resides in the deep oceans (Rao and Fehn, 1999). ^{129}I has also been produced anthropogenically during the last 60 years during fission processes associated with civil and military nuclear activities and accidents, such as the Chernobyl accident (April 26th, 1986). An estimated 50–150 kg of ^{129}I was released to the atmosphere during the main period (1945–1964) of nuclear weapons testing (Raisbeck et al., 1995; Wagner et al., 1996) and a further ~6 kg were released due to the Chernobyl accident (Gallagher et al., 2005). However, the main sources of anthropogenic ^{129}I are nuclear fuel reprocessing plants (La Hague-France and Sellafield-England, see Fig. 1). It has been estimated that, by 2004, 1371 kg of ^{129}I had been discharged into the NE Irish Sea from the Sellafield plant, with an additional 182 kg released directly to the atmosphere in gaseous form (Gray et al., 1995; López-Gutiérrez et al., 2004). By the end of the same year, 3119 kg of ^{129}I had been released from La Hague plant as liquid discharges into the English Channel, with an

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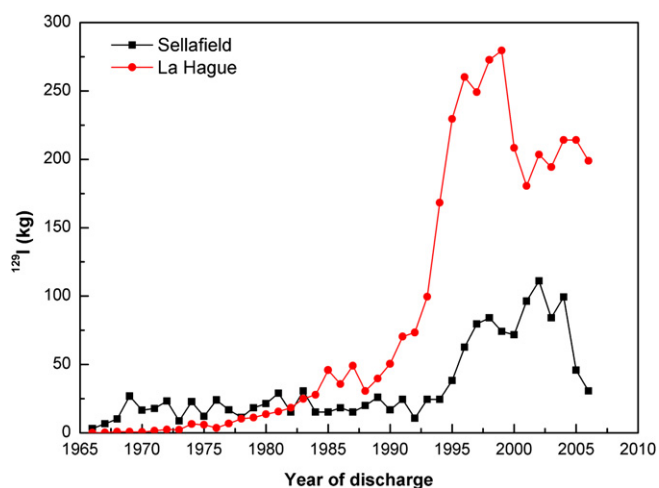
Fig. 1. Annual [129]I release to the sea from the reprocessing facilities in Sellafield and La Hague (compilation until 2004 from López-Gutiérrez et al. (2004) and the rest by Monitoring our Environment (2006) and Areva Rapport Environnement (2006)).

additional 68 kg released in gaseous form (López-Gutiérrez et al., 2004; Aldahan et al., 2007). These large releases of [129]I from reprocessing facilities, along with the conservative behaviour of iodine in seawater (Chai and Zhu, 1994) give a residence time of iodine in the ocean of 4×10^5 years, make [129]I an important oceanographic tracer.

Seaweed is useful as a bioindicator for monitoring radioactive contamination of the sea, due to its property of concentrating various elements present in the water. It is well known that it can concentrate iodine to a high level, with concentration factors as high as 10^4 in some brown seaweed (Hou and Yan, 1998). It has therefore been used as a bioindicator of radioiodine, such as ^{131}I and [129]I (Druehl et al., 1988; Cooper et al., 1998; Hou et al., 2000a). In addition, the relatively high stability of iodine in seaweed and the convenience of storing dry seaweed present a unique possibility to use archived seaweed collected at appropriate times and locations. This provides useful records of the variation of the anthropogenic [129]I levels.

It has been well established that the discharged radioactive effluents from the reprocessing facilities at La Hague are first transferred northwards to the coast of north-western Europe via the English Channel, mixed with effluent from Sellafield at the North Sea, and then further transferred by the Norwegian Coastal Current to the Arctic (Alfimov et al., 2004). Meanwhile, there is an inflow of saline sea water from the North Sea that enters the Skagerrak bringing anthropogenic pollutants from the European reprocessing facilities. [129]I in the Baltic Sea is mainly due to fallout from the atmospheric emissions from nuclear facilities and emissions from contaminated seawater as iodide (especially forming organic compounds, such as CH_3I (Aldahan et al., 2006)), and iodate (Lehto et al., 2012).

The Baltic and Black Seas became contaminated by the Chernobyl accident with ^{90}Sr, [134]Cs, ^{137}Cs and [239,240]Pu. The dose rates to the public from ingestion of ^{137}Cs from Chernobyl in aquatic food have been estimated to be low, at least one order of magnitude lower than those due to natural [210]Po (Povinec et al., 1996). The highest dose from the Chernobyl accident was delivered to the critical group in the Baltic Sea region, who received a maximum dose of 0.08 mSv during 1986 (Nielsen and Keith-Roach, 2004).

While the distribution and behaviour of caesium isotopes have been widely studied, less is known about the iodine isotopes, which

had a substantially different initial release history and also different physical mechanisms governing their deposition. Thus, in the 10 days period after the accident, the initial large release from the Chernobyl reactor contained mainly the more volatile radionuclides such as noble gases, iodine and some caesium. The second large release was associated with the high temperatures reached in the core melt, and contained most of the particulate caesium (Brant et al., 2002).

Here we present a study of [129]I concentration in brown seaweed *Fucus vesiculosus* sampled around the coastline of Sweden at 33 different stations in the years 1982 and 1986 (post-Chernobyl accident). The resulting data were evaluated in terms of [129]I concentrations, [129]I/[127]I and [129]I/^{137}Cs ratios.

2. Sampling and radiochemical method

Brown seaweed *F. vesiculosus* was sampled at locations selected to provide a representative geographical distribution on all aspects of the Swedish coast (Fig. 2). Sampling took place in years 1982 (during the month of October) and 1986 (during the month of July), and about 1 kg of each sample was collected, dried, ground and homogenised.

All samples were chemically prepared using the microwave-digestion method described by Gómez-Guzmán et al. (2011). Typically, 0.5 g of sample and 10 mL of 60% (v/v) HNO_3 suprapure quality (Merck, Darmstadt, Germany) were introduced into the vessels. The operational conditions were: 200 °C for 10 min (ramp of 10 min), 200 °C for 20 min and 20 min for cooling to room temperature. After digestions, the chemical separation consisted of an organic compound extraction followed by an aqueous solution extraction. Iodine was extracted from digested samples three times with 10 mL $CHCl_3$ and then back-extracted into aqueous solution by addition of 20 mL 0.1 M $NaHSO_3$. Then the solution was stirred and the SO_3^{2-} and SO_4^{2-} ions were precipitated by addition of 10 mL of saturated $Ba(NO_3)_2$ solution followed by centrifugation. Finally, iodine was precipitated as AgI by adding 10 mL 0.1 M $AgNO_3$ solution, washed twice with distilled water and dried at 60−80 °C. The mean recovery of the radiochemical procedure was found to be 90%. The AgI was mixed with about the same volume of Nb powder and loaded in a copper target holder for the measurement of [129]I. After the digestion and before the addition of 2 mg of Woodward iodine as carrier ([129]I/[127]I $\approx 7 \times 10^{-14}$ (Roberts et al., 1997)), a 1 mL aliquot of each sample was used for [127]I measurement. These aliquots were diluted in adequate volume of double deionised water (18 MΩ cm) to obtain maximum stable iodine concentrations in the order of 20 μg L^{-1}.

3. Experimental

3.1. Measurement of [129]I by AMS

Determination of [129]I/[127]I ratios in seaweed samples was conducted at the 1 MV multielement compact Accelerator Mass Spectrometry (AMS) system at the Centro Nacional de Aceleradores (CNA) in Seville (Spain). Details of the measurement and the system have been previously described (Gómez-Guzmán et al., 2012; Klein et al., 2006, 2007), so only a short description will be given here. A beam of negative iodine ions is extracted from the AgI + Nb target using the Cs^+ high-intensity sputter source at 35 keV. At the terminal of the tandem, negative iodine ions are changed to positive iodine ions by the stripper (pressure at 6×10^{-3} mbar of argon, which corresponds to a mass thickness of about 0.15 μg cm^{-2}) and they are then accelerated. Following acceleration, selected positive ions (charge state +3) are analyzed by mass spectrometer. The [127]I
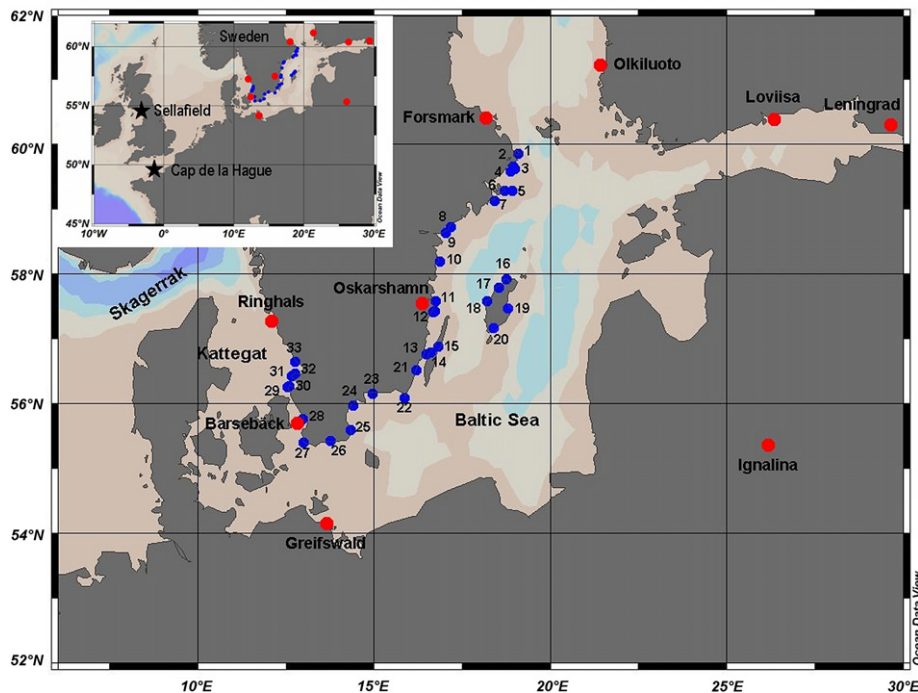
Fig. 2. Sampling sites (blue dots) of *Fucus vesiculosus* extracted from Carlson and Holm (1992). Red dots represent the nuclear facilities in the drainage are of the Baltic Sea. The small map in the left upper corner shows the location of Sellafield and La Hague reprocessing facilities with respect to the sampling sites. [For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.]

current beam is measured in a Faraday cup, and $^{129}I^{3+}$ ions are counted in a gas ionization chamber. AMS measurements were performed relative to a standard with known $^{129}I/^{127}I$ isotopic ratio which was made by repeated dilutions from NIST SRM 3230 Iodine Isotopic Standard Level I.

The isotopic ratios obtained directly by AMS measurement of the samples (including carrier) were typically of the order of $^{129}I/^{127}I \sim 10^{-10}–10^{-11}$. Background effects were evaluated through frequent digestions of iodine blanks (Woodward Iodine, from Woodward Iodine Corporation, USA), which measured a $^{129}I/^{127}I$ isotopic ratio of $(3–4) \times 10^{-13}$ when 2 mg carrier was used, meanwhile the limit of detection of the CNA AMS system was found to be $^{129}I/^{127}I = (3.11 \pm 0.16) \times 10^{-13}$ (Gómez-Guzmán et al., 2012). This is typically 2 to 3 orders of magnitude lower than the isotopic ratios measured for real samples.

3.2. Measurement of ^{127}I by ICP-MS

Measurements of ^{127}I content in seaweed samples were performed at the Inductively Coupled Plasma Mass Spectrometry (ICP-MS) Unit of the Servicio de Investigación Agraria (University of Seville, Spain). Details of the measurement and the system have been previously described (Gómez-Guzmán et al., 2011), so only a short description will be given here. A Quadrupolar ICP-MS (Thermo X7, Thermo, Madrid, Spain) equipped with a concentric quartz nebulizer (Meinhard Associates, Golden, USA) was used for ^{127}I determination. The instrument was used at normal resolution and set to detect the signal intensity at m/z 127. Samples were analyzed using recommendations provided by US-EPA 200.8 methodology adapted for iodine. Various data quality tests were used during every instrumental running, such as replicate, matrix matching, memory effects, and both digestion analysis and laboratory reactive blanks checking. The instrumental detection limit of ^{127}I calculated as three times the standard deviation of the blank, was $0.045 \ \mu g \ L^{-1}$.

The methodology used to measure the ^{127}I content in seaweed samples by this technique was previously validated by measuring ^{127}I content in standard reference materials from the National Institute of Standards and Technology (NIST), the International Atomic Energy Agency (IAEA) and the Community Bureau of Reference (BCR) (Gómez-Guzmán et al., 2011).

4. Results

Concentrations of ^{129}I and ^{127}I, $^{129}I/^{127}I$ ratios and the activity of ^{137}Cs in seaweed samples collected in 1982 and 1986 around the Swedish coastline are shown in Table 1. It can be seen that the concentration of stable iodine (^{127}I) in the seaweed varies significantly from 54 to 178 $\mu g \ g^{-1}$ for 1982 and from 37 to 372 $\mu g \ g^{-1}$ for 1986 dry weight. This variation has also been observed in other species of seaweed (Hou and Yan, 1998; Hou et al., 2000b). The ratio $^{129}I/^{127}I$ instead of ^{129}I concentration in seaweed could be used as an index of the ^{129}I level in the surrounding seawater of sampling location, which is based on the assumption that seaweed has a similar capability to take up and assimilate ^{129}I and ^{127}I, as previously reported by Hou et al. (2000b).

^{129}I concentrations in seaweed samples were found to be in the order of $(0.82–5.89) \times 10^9$ atoms g^{-1} in 1982 and $(1.33–38.83) \times 10^9$ atoms g^{-1} in 1986 (Table 1, Fig. 3b). The $^{129}I/^{127}I$ ratios ranged from $(22.7–87.8) \times 10^{-10}$ for seaweed collected in 1982 and from $(26.1–305.5) \times 10^{-10}$ for seaweed collected in 1986 (Table 1, Fig. 3a). For both years these $^{129}I/^{127}I$ levels are still two or three orders of magnitude higher than the fallout background level of $\sim 10^{-10}$ (Edmonds et al., 1998; Cooper et al., 1998).

From Table 1 and Fig. 3 it can be seen that there is a geographical enhancement of ^{129}I concentrations and $^{129}I/^{127}I$ ratios in samples collected in the Kattegat area (positions 27–33) with respect to seaweed samples collected in the Baltic Sea coastline (positions 1–26). The effect of the horizontal gradients in salinity has to be

Table 1
^{129}I and ^{127}I concentrations (dry weight), ^{137}Cs activities and ^{129}I/^{127}I ratios in *Fucus vesiculosus* sampled around the Swedish coastline in 1982 and 1986. Quoted uncertainties are ±1σ. Uncertainties related to the ^{129}I/^{127}I ratios come from the error propagation theory. ^{137}Cs data extracted from Carlson and Holm (1992).

Sampling site	1982				1986			
	^{129}I ($\times 10^9$ atoms g^{-1}) (measured)	^{127}I (µg g^{-1}) (measured)	^{129}I/^{127}I ($\times 10^{-9}$) (calculated)	^{137}Cs (Bq kg^{-1}) (measured)	^{129}I ($\times 10^9$ atoms g^{-1}) (measured)	^{127}I (µg g^{-1}) (measured)	^{129}I/^{127}I ($\times 10^{-9}$) (calculated)	^{137}Cs (Bq kg^{-1}) (measured)
1					3.49 ± 0.16	38 ± 5	19.6 ± 3	600 ± 29
2					1.77 ± 0.09	61.6 ± 1.6	6.0 ± 0.3	110 ± 6
3					3.87 ± 0.22	40 ± 4	20.2 ± 2.4	138 ± 7
4					1.80 ± 0.11	35.9 ± 0.2	10.6 ± 0.7	91 ± 5
5					1.33 ± 0.06	54 ± 4	5.2 ± 0.5	41 ± 2
6					1.55 ± 0.21	51 ± 3	6.4 ± 0.9	46 ± 3
7	1.03 ± 0.06	75 ± 6	2.9 ± 0.3	8.2 ± 0.5	1.81 ± 0.15	51.4 ± 0.6	7.4 ± 0.6	26 ± 1
8	0.82 ± 0.05	77 ± 3	2.27 ± 0.16	7.3 ± 0.7	1.59 ± 0.07	74 ± 6	4.5 ± 0.4	30 ± 2
9	1.18 ± 0.06	85 ± 5	2.92 ± 0.23	9.3 ± 0.5	1.88 ± 0.18	53 ± 4	7.4 ± 0.9	46 ± 3
10	0.93 ± 0.05	61 ± 7	3.2 ± 0.4	7.5 ± 0.4	1.72 ± 0.10	41.9 ± 2.0	8.6 ± 0.7	26 ± 1
11	0.96 ± 0.05	67 ± 5	3.0 ± 0.3	7.0 ± 0.4				
12	1.12 ± 0.09	90 ± 4	2.66 ± 0.24	7.5 ± 0.7	1.60 ± 0.08	129 ± 5	2.60 ± 0.17	20 ± 1
13	1.44 ± 0.10	102 ± 4	2.97 ± 0.24	7.2 ± 0.5	1.50 ± 0.09	78.±4	4.1 ± 0.3	28 ± 2
14					1.9 ± 0.3	104 ± 3	3.8 ± 0.7	28 ± 2
15					2.55 ± 0.13	84 ± 3	6.4 ± 0.4	57 ± 3
16	2.11 ± 0.12	132 ± 12	3.4 ± 0.4	7.5 ± 0.4	1.82 ± 0.11	71 ± 4	5.4 ± 0.4	24 ± 1
17	2.09 ± 0.10	84 ± 3	5.3 ± 0.3	8.3 ± 0.7	1.97 ± 0.19	35 ± 3	12.0 ± 1.5	27 ± 2
18					1.9 ± 0.3	106 ± 8	3.8 ± 0.7	42 ± 2
19					2.19 ± 0.11	115 ± 10	4.0 ± 0.4	29 ± 2
20	2.48 ± 0.13	144 ± 6	3.65 ± 0.24	8.6 ± 0.7	2.81 ± 0.16	163 ± 3	3.64 ± 0.22	89 ± 5
21	3.39 ± 0.15	131 ± 2	5.5 ± 0.3	9.3 ± 0.5				
22	2.51 ± 0.11	134 ± 9	3.9 ± 0.3	8.7 ± 0.9	2.89 ± 0.21	142 ± 5	4.3 ± 0.4	23 ± 1
23	2.81 ± 0.16	68 ± 7	8.8 ± 1.0	14.6 ± 0.8	2.1 ± 0.4	67 ± 6	6.8 ± 1.3	24 ± 1
24	2.18 ± 0.09	72 ± 4	6.4 ± 0.4	8.0 ± 0.7	1.88 ± 0.13	50 ± 3	7.9 ± 0.7	352 ± 19
25	2.05 ± 0.10	54 ± 8	7.9 ± 1.3	8.2 ± 0.9	2.66 ± 0.12	55 ± 4	10.2 ± 0.9	19 ± 1
26	3.22 ± 0.23	118 ± 8	5.7 ± 0.6	6.9 ± 0.4	3.36 ± 0.17	68.4 ± 1.9	10.4 ± 0.6	107 ± 5
27	4.02 ± 0.21	109 ± 8	7.8 ± 0.7	10.4 ± 0.6	6.9 ± 0.3	114 ± 5	12.7 ± 0.8	40 ± 2
28	5.89 ± 0.15	178 ± 15	6.9 ± 0.6	13.0 ± 1.3	18.0 ± 0.8	132 ± 5	28.6 ± 1.6	36 ± 2
29					29.6 ± 1.2	341 ± 24	18.3 ± 1.5	34 ± 2
30					28.8 ± 1.2	331 ± 12	18.4 ± 1.0	34 ± 2
31					31.0 ± 1.6	372 ± 18	17.6 ± 1.3	33 ± 2
32					29.4 ± 2.0	279 ± 7	22.2 ± 1.6	27 ± 1
33					38.8 ± 2.1	269 ± 15	30.5 ± 2.4	41 ± 2

evaluated, but previous results indicate that a significant amount of discharge from the reprocessing facilities had been transferred from the North Sea to the inner Danish waters and reached the west coast of Sweden through the Kattegat and Skagerrak. However, only a minor fraction of this contaminated seawater is then transferred through the Kattegat further to the Baltic Sea; most of it returns to the North Sea (Hou et al., 2000b).

To compare these ^{129}I concentrations and ^{129}I/^{127}I ratios on a global scale, Table 2 presents a comparison of ^{129}I levels and ^{129}I/^{127}I ratios in seaweed from other locations worldwide. For example, Osterc and Stibilj (2008) reported results obtained in seaweed *Fucus virsoides* sampled during the summers of 2005 and 2006 in the coastline of the Adriatic Sea. ^{129}I concentrations and ^{129}I/^{127}I ratios in these samples ranged between (1.5–3.4) × 10^9 atoms g^{-1} and (8.3–21.1) × 10^{-10}, respectively. Results reported by Fréchou and Calmet (2003) and obtained in seaweed sampled between 1998 and 1999 in Goury, sited in the vicinity of La Hague nuclear reprocessing plant, show very high ^{129}I concentrations and ^{129}I/^{127}I ratios. Both range between (1.4–3.2) × 10^{13} atoms g^{-1} and (1.01–1.94) × 10^{-5}, respectively, between four and five orders of magnitude higher to that found in this study. Hou et al. (1999a) reported ^{129}I concentrations and ^{129}I/^{127}I ratios in seaweed sampled in Denmark and Iceland between 1988 and 1992. These values ranged between (0.6–115) × 10^9 atoms g^{-1} and (4–640) × 10^{-10}, respectively, and were in the same order of magnitude as results found in this study. Results reported by Cooper et al. (1998) showed ^{129}I/^{127}I ratios ranged between (2.2–80) × 10^{-10} in seaweed sampled in Novaya Zemlya between 1967 and 1993. This Russian archipelago has been influenced by the impact of the Soviet nuclear

tests and, indirectly, its waters are contaminated by Sellafield and La Hague liquid emissions that arrive through the NCC (Norwegian Coastal Current) (Arctic Monitoring and Assessment Programme, 2003).

Activity concentrations of ^{137}Cs varied from 600 Bq kg^{-1} at the northernmost locality (1), to 20–25 Bq kg^{-1} at the southeast coast in July 1986. For 1982, the mean activity concentration of ^{137}Cs was 9 ± 2 Bq kg^{-1}. Thus, the ^{137}Cs concentration increased by 2–5 times in most of the seaweed samples (Table 1).

5. Discussion

5.1. ^{127}I

The Baltic Sea is a semi-enclosed water body connected to the North Sea through the Strait of Kattegat. Salinity distributions in its surface layer (0–10 m) show strong horizontal gradients, with values close to 30 psu at the Kattegat area. This value decreases towards the Bothnian Bay and the gulfs of Finland and Riga, where values below 3 psu are found (Feistel et al., 2010). Vertical stratification is also noticeable (Hou et al., 2001). ^{127}I is naturally present in seawater with concentrations that increase with the salinity, as shown in Fig. 4, with data from a set of water samples from the Baltic Sea area (Hou et al., 2001; Lehto et al., 2012). Fig. 4 shows that iodine behaviour is conservative, as expected taking into account that this radionuclide is generally soluble in seawater and poorly adsorbed onto particulates and sediment. Both iodide (I$^-$) and/or iodate (IO$_3^-$) anions can be present, but the latter predominates (Pentreath, 1985).
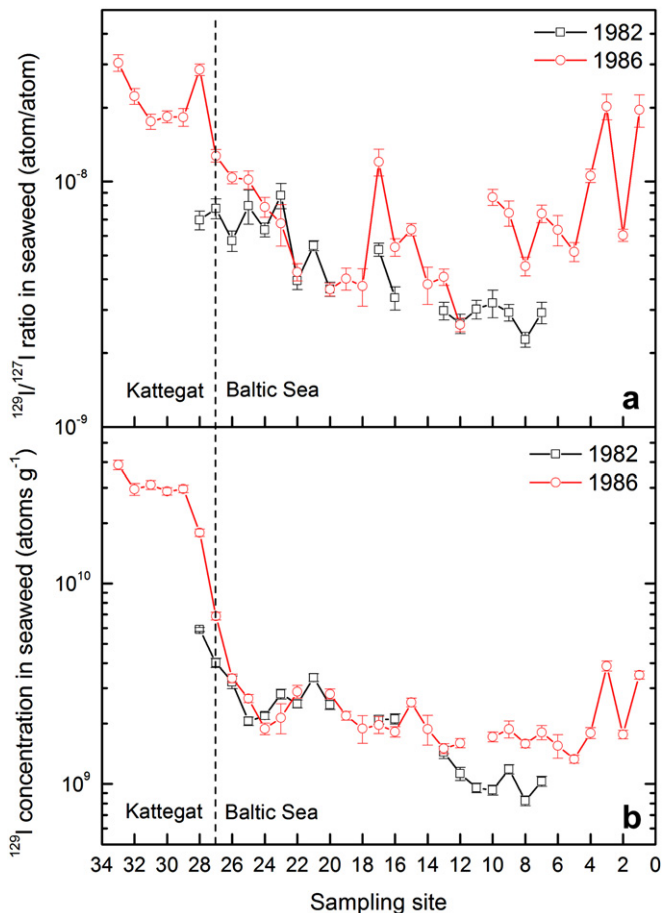
Fig. 3. (a) $^{129}I/^{127}I$ ratios and (b) ^{129}I concentration (dry weight) in seaweed *Fucus vesiculosus* sampled around the Swedish coastline in 1982 and 1986.

Table 2
^{129}I concentrations and $^{129}I/^{127}I$ reported in various seaweeds sampled worldwide.

Seaweed	Sampling zone		^{129}I ($\times 10^9$ atoms g^{-1})	$^{129}I/^{127}I$ ($\times 10^{-10}$)	Reference
	Date	Location			
F. Vir.	Sept. 2005	Slovenia	2.0 ± 0.6	424.5 ± 34.0	(Osterc and
F. Vir.	June 2006	Italy	1.5 ± 1.1	405.3 ± 28.9	Stibilj, 2008)
F. Vir.	Oct. 2006	Croatia	3.4 ± 1.1	330.7 ± 26.8	
F. Ves.	Oct. 1998	Goury, France	$14,000 \pm 900$	$99,434 \pm 985$	(Fréchou and Calmet,
F. Ves.	March 1999	Goury, France	$32,000 \pm 2300$	$190,992 \pm 17781$	2003)
F. Ves.	July 1967	Kruglaya Levaya		21.4 ± 1.1	(Cooper et al., 1998)
F. Ves.	July 1972	Sosnovetz		71.8 ± 2.0	
F. Ves.	August 1993	Gribovaya Fjord		789 ± 18	
F. Ves.	Dec. 1988	Iceland	0.6 ± 0.04	3.90 ± 0.24	(Hou et al., 1999a)
F. Ves.	Nov. 1989	Klint, Denmark	62.1 ± 4.6	393 ± 6	
F. Ves.	April 1992	Klint, Denmark	115.3 ± 10.4	638 ± 11	
	1982	8	0.82 ± 0.05	22.7 ± 0.16	This work
F. Ves.	1982	28	5.89 ± 0.15	88 ± 10[a]	
F. Ves.	1986	5	1.33 ± 0.06	26.1 ± 1.7[a]	
F. Ves.	1986	33	38.8 ± 2.1	305 ± 24	

[a] Highest $^{129}I/^{127}I$ ratio in 1982 and lowest $^{129}I/^{127}I$ ratio in 1986 were found in positions 23 and 12, respectively.

The natural inventory of ^{129}I has been estimated to be ~ 230 kg, most of which resides in the deep oceans (Rao and Fehn, 1999). It is well established that the ratio $^{129}I/^{127}I$ in the pre-nuclear era had typical values between 10^{-12} (Kilius et al., 1992) and 6×10^{-13} (Fabryka-Martin et al., 1985). These levels, compared with the ones measured in *F. vesiculosus* in this work, account for <0.05% of the total ^{129}I inventory in this region.

An estimated amount of 50–150 kg of ^{129}I were released to the atmosphere during the main period (1945–1964) of nuclear weapons testing (Raisbeck et al., 1995; Wagner et al., 1996), which enhanced the natural $^{129}I/^{127}I$ ratio to 10^{-10}. This level, compared with the isotopic ratios measured in this work, still account for <5% of the total ^{129}I inventory in the Baltic Sea region. According to Salo et al. (1986), the inventory of ^{137}Cs in the Baltic Sea due to this source, based on measured concentrations in water and sediments, is 620 TBq (calculated to 1981).
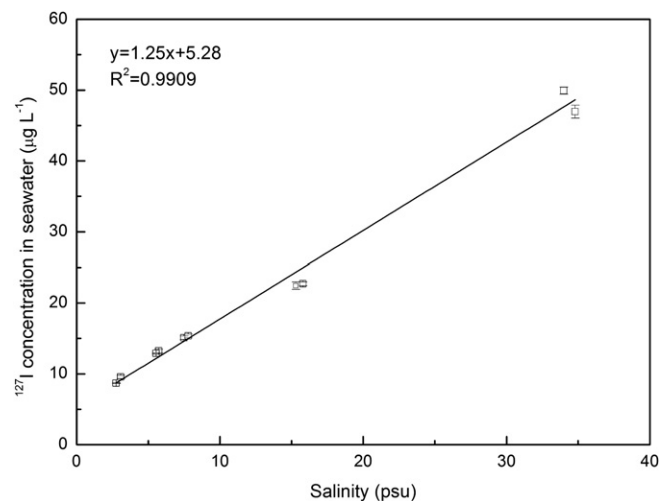
Measured values of ^{127}I concentrations in *F. vesiculosus* sampled around the southern Swedish coastline in 1982 and 1986 along with salinity are shown in Fig. 5 (mean annual values for the period 1900–2005 and for $1° \times 1° \times 10$ m size cells, taken from Feistel et al., 2010). The distribution of ^{127}I concentrations, C, as a function of the sampling point were similar in both campaigns, with an increasing trend towards the Strait of Kattegat that can be linearly correlated with salinity, S, at 99% confidence level ($R^2 = 0.84$, $p < 0.01$; $C = 20.4 \cdot S - 60.4$, in units of $\mu g \; g^{-1}$).

^{127}I concentrations in seawater can be inferred from the salinity data (Fig. 5) and from Fig. 4, which allows the estimation of concentration factors (defined as the ratio between concentrations in *F. vesiculosus* and in seawater). The so obtained values for the two sampling campaigns are compatible with a normal distribution of mean value 6540 and a standard deviation of 400. When excluding the sampling points from the Kattegat area (those with higher salinity, and index >27), the concentration factors follow a normal distribution with mean value 5900 and standard deviation of 355.

5.2. ^{129}I, ^{137}Cs and $^{129}I/^{137}Cs$ ratio

The possible origins of man-made radionuclides in the Baltic Sea can be classified as follows: natural occurrence, fallout from atmospheric nuclear weapon tests, the release from the Chernobyl nuclear power plant accident, authorised discharges of radioactivity into the sea occurring during routine operation of nuclear installations in the Baltic Sea region and discharges from European nuclear fuel reprocessing plants.

Fig. 4. Correlation between ^{127}I concentration and salinity in seawater. ^{127}I and salinity data extracted from Hou et al. (2001) and Lehto et al. (2012).
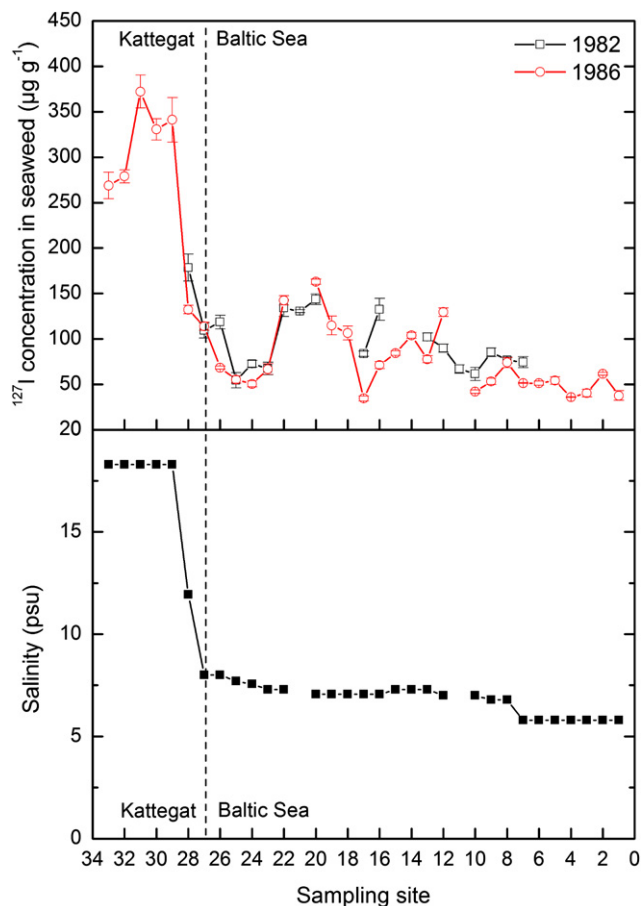
Fig. 5. [127]I concentration (µg g[−1]) in *Fucus vesiculosus* from the Baltic Sea in 1982 (black) and 1986 (red), and salinity (mean annual values for the period 1900–2005 for 1° × 1° × 10 m size cells, taken from Feistel et al. (2010)). [For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.]

Fig. 6. [129]I concentrations in seawater in the different sampling sites inferred by concentration factors for [127]I.

The Chernobyl accident, which happened in April 1986, released a large amount of radioactive material, including about 6 kg of [129]I (Gallagher et al., 2005). The total input of ^{137}Cs from the Chernobyl accident into the Baltic Sea area was estimated at 4500 TBq by CEC (1991). This estimate was later adjusted to 4700 TBq by Nielsen et al. (1999). The deposition fallout from Chernobyl was very unevenly distributed in the drainage area of the Baltic Sea; the most contaminated areas were surrounding the Bothnian Sea and the eastern Gulf of Finland. This behaviour is shown in Table 1, where it can be seen that the highest ^{137}Cs activities in samples from 1986 are in the north-eastern area of the Baltic Sea.

Concentration factors for [127]I can be used to reconstruct the [129]I concentration in seawater from the measured concentrations in *F. vesiculosus* collected in 1982. In the second sampling campaign, samples were collected in July 1986, three months after the Chernobyl accident. The biological half-life of iodine in *F. vesiculosus* has been measured and found to be quite short, between 5 and 10 days (Vives i Batlle et al., 2007; Druehl et al., 1988). Using a CF of 14.7 m[3] kg[−1] and a typical mass of 0.261 kg (Vives i Batlle et al., 2008), a simple two-compartmental model shows that equilibrium with a water body of $\sim 10^{10}$ m[3] should be attained in two months. Although in the real world a steady-state is rarely encountered, under our particular conditions, it seems reasonable to use concentration factors derived from [127]I for estimating the [129]I concentrations in seawater in July 1986. Furthermore, from the results shown in Table 1, ^{137}Cs concentrations in *F. vesiculosus*
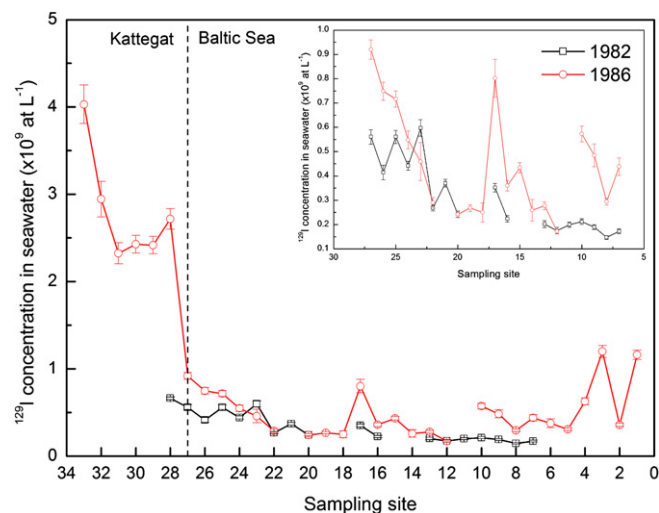
sharply increased in samples collected in July 1986 (it is worth noting that radionuclide discharges from Sellafield and Cap de la Hague continued their decreasing trend in the period between sampling campaigns, and thus, activity concentrations lower than those measured in 1982 could be expected just prior the Chernobyl accident). On the other hand, the application of models with concentration factors constant in time (although varying from site to site) for ^{137}Cs, could reasonably reproduce the observed time series of ^{137}Cs concentrations in *F. vesiculosus* in the Baltic Sea, prior to and after the Chernobyl accident (HELCOM, 2009).

Results are depicted in Fig. 6 (double panel, with the detail for the common area covered by the two sampling campaigns). Concentrations in 1982 show an increasing trend towards the Kattegat Strait, consistent with the expected pattern of [129]I dispersion following liquid discharges from Sellafield and Cap de la Hague (Povinec et al., 2003) — we note that the effect of salinity was already incorporated in the estimation of concentration factors —. Within this common area, [129]I concentrations in 1986 are slightly increased in some locations, as it could be expected of non-uniform atmospheric deposition from the Chernobyl radioactive cloud (HELCOM, 2009), and the limited homogenization of concentrations promoted by water circulation during this elapsed time (Toscano and García-Tenorio, 2004). At the two edges of the expanded area of sampling in 1986, relatively higher concentrations of [129]I were found. To distinguish what extent they are due to localized deposition from the Chernobyl accident or if they are the result of the dispersion pattern from the two reprocessing plants, it is necessary to study the ^{137}Cs distribution and the [129]I/^{137}Cs isotopic ratios.

From Table 1, ^{137}Cs concentrations in *F. vesiculosus* in 1982 were rather uniform, with a mean value and standard deviation of 8.7 and 0.5 Bq kg[−1], respectively. It is well established that the Chernobyl accident was the main source of ^{137}Cs to the Baltic Sea. For example, Carlson and Holm (1992) measured activity ratios [134]Cs/^{137}Cs of (0.45 ± 0.05) in seaweed sampled in these locations, in good agreement with the ratio (0.47–0.48) in July 1986, calculated by IAEA (1986). Thus, ^{137}Cs concentrations in *Fucus* collected in 1986 were very high in samples 1–4 and that can be linked with high local deposition from Chernobyl, and is also consistent with the high concentrations of [129]I found in this area. Isolated peaks in ^{137}Cs concentration also appear in samples 24 and 26. By excluding these six samples, the mean concentration and standard deviation become 35 and 3 Bq kg[−1], respectively, a factor 4 higher than the
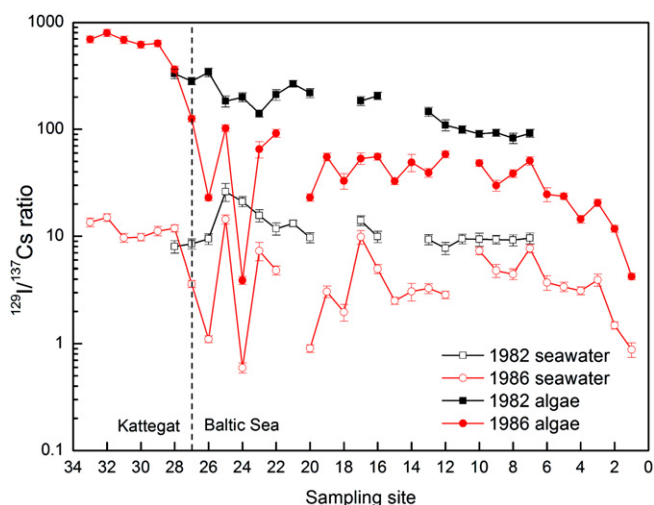
Fig. 7. $^{129}I/^{137}Cs$ atom ratio in seawater (open circles and squares) and in *Fucus vesiculosus* (filled circles and squares) at the various sampling sites for 1982 (black) and 1986 (red). Uncertainties come from the error propagation theory. [For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.]

levels measured in 1982 (and likely higher than the pre-Chernobyl levels, due to the known decreasing trend in ^{137}Cs releases from Sellafield and Cap de la Hague). Finally, ^{137}Cs concentration in *Fucus* in samples 29–33 remained at the level of the previous mean value, which in turn suggests that the high concentrations of ^{129}I found in this area should be related to the effluents from the two reprocessing plants.

The concentration factors for ^{137}Cs in *F. vesiculosus* varies widely from site to site in the Baltic Sea (HELCOM, 2009), most likely linked to changes in salinity. This could mask to some extent the information from isotopic ratios. Thus, ^{137}Cs concentrations in seawater have been estimated from their recommended concentration factors in *F. vesiculosus* in the different regions covered by the sampling campaign (HELCOM, 2009; concentration factors ranging from 188 ± 39 L kg^{-1} dry weight in Kattegat to 453 ± 145 L kg^{-1} in the western Baltic Sea and 622 ± 100 L kg^{-1} in the Bothnian Sea). This allows estimation of the $^{129}I/^{137}Cs$ isotopic ratios in both matrices, *F. vesiculosus* and seawater using, for the last one, the following equation,

$$\left[\frac{^{129}I}{^{137}Cs}\right]_W = \frac{\left[\frac{^{129}I}{^{127}I}\right]_F \times [^{127}I]_W \times CF(^{137}Cs)}{[^{137}Cs]_F}$$

where: $[^{129}I/^{137}Cs]_W$ is the $^{129}I/^{137}Cs$ atom ratio in seawater at the various *Fucus* sampling sites; $[^{129}I/^{127}I]_F$ is the atom ratio in *Fucus*, using ^{129}I and ^{127}I concentrations obtained in this work, and assuming it to be the same as the $[^{129}I/^{127}I]$ ratio in seawater (Hou et al. (2000b)); $[^{127}I]_W$ is the stable iodine concentration in coastal water around Sweden, taken from salinity data (Figs. 3 and 4); $[^{137}Cs]_F$ is the ^{137}Cs concentration in *Fucus* given by Carlson and Holm (1992) and $CF(^{137}Cs)$ is the recommended concentration factor for radiocaesium from seawater to *Fucus* in Swedish coastal waters given by HELCOM (2009). These ratios are shown in Fig. 7. From the estimated composition of the Chernobyl emissions (Hou et al., 2002), the expected $^{129}I/^{137}Cs$ isotopic ratio was 0.18, much lower than the values depicted in Fig. 7. Previous levels of ^{137}Cs concentrations in seawater contribute to decrease this ratio, while the previous existing levels of ^{129}I increase it. If the distribution and deposition of both isotopes did not largely differ, the isotopic ratios

in seawater depicted in Fig. 7 have to be understood in terms of the relatively low contribution of ^{129}I from Chernobyl to enhance previously existing levels (as shown in Fig. 6), what also has been reported elsewhere (Hou et al., 2002; Buraglio, 2000). Furthermore, from Fig. 7 it can be concluded that as the deviation of this isotopic ratio in seawater becomes larger with respect to the 1982 levels, the higher will be the local atmospheric deposition of the Chernobyl effluents. Again we can see that for samples 29–33, the isotopic ratio remains at the same level as that in 1982, which suggests that the origins of ^{129}I in this region are the Sellafield and Cap de La Hague reprocessing plants.

The $^{129}I/^{137}Cs$ isotopic ratios in *F. vesiculosus* are mediated by the different concentration factors (those for ^{129}I being on average, larger by a factor 20 than those for ^{137}Cs) and by their different trends of variation with salinity (concentration factors for ^{129}I slightly increase with increasing salinity, while those for ^{137}Cs sharply decrease with increasing salinity). In fact, these data can be used to infer a relationship for the concentration factor for ^{137}Cs in *F. vesiculosus* as a function of the salinity in the Baltic Sea area, following a log–log relationship ($R^2 = 0.79$, confidence level 99%; data set for 1986):

$$\log [CF_{Cs}] = 3.55 - 1.03\log [S]$$

This relationship can be understood in terms of the effect of competitive ions (Abril and Fraga, 1996). Effectively, ^{137}Cs is usually present in seawater as a monovalent cation at tracer concentrations, in competition with the dominant cations Na^+ and K^+, whose concentrations increase with salinity. It is worth noting in contrast that iodine exists in seawater mainly as dissolved anions iodate and iodide (Hou et al., 2001).

From data shown in Table 1 it can be seen that the ^{137}Cs concentrations in seaweed samples collected in 1986 are much higher than those collected in 1982. This also explains the fact that the atomic ratios $^{129}I/^{137}Cs$ in the Baltic Sea shown in Fig. 7 were higher in 1982 than in 1986. This was because the deposition of additional ^{137}Cs from the Chernobyl accident was much more important than the additional ^{129}I deposition from the same source. The highest ^{137}Cs activity concentration in 1986 was observed at location 1, the northernmost locality, and adjacent to the land area which received the highest fallout deposition in Sweden from the Chernobyl accident (Carlson and Holm, 1992). The unexpected high ^{137}Cs activity concentration found at localities 24 and 26 could be influenced by the nearly presence of two nuclear power plants, located at Barsebäck and Oskarshamn, both releasing ^{137}Cs in the drainage area of the Baltic Sea.

Concerning other possible contributions to ^{129}I and ^{137}Cs in this area, around the Baltic Sea there are nuclear power plants in Sweden, Finland, Russia, Lithuania and Germany (as shown in Fig. 2) that could directly or indirectly release ^{129}I and ^{137}Cs to its drainage area. No data of ^{129}I discharge from these power plants are available, but some authors (Buraglio, 2000; Hou et al., 1999b, 2002) have concluded that their contribution to the ^{129}I inventory in the Baltic Sea is insignificant. HELCOM (2009) has determined the total aquatic ^{137}Cs discharges from these local nuclear facilities into the Baltic Sea to be 2.4 TBq until the end of 2006, compared to the 4700 TBq released from the Chernobyl accident in the same area. So the contribution of these facilities to the total inventory of ^{137}Cs in the Baltic Sea area is also insignificant, although some authors have related ^{137}Cs concentrations measured in *Fucus* collected near Barsebäck nuclear power plant with its released cooling water (Erlandsson and Matsson, 1988; Mattsson et al., 1980).

From our present results, it is clear that the most important contribution to the ^{129}I inventory in the Baltic Sea area comes from the releases of the two reprocessing facilities at Sellafield and La

Hague. HELCOM (2009) has also estimated the total input of ^{137}Cs originating from these two nuclear reprocessing plants to be 250 TBq, which is even less than the ^{137}Cs released as a result of fallout from atmospheric nuclear weapon tests (620 TBq calculated to 1981). Thus, for this radionuclide the most important source to the inventory in the Baltic Sea area is the Chernobyl accident.

6. Conclusions

^{129}I and ^{129}I/^{127}I ratios in seaweed samples *F. vesiculosus* collected in 1982 (during the month of October) and 1986 (during the month of July) have been measured. For both years, ^{129}I/^{127}I ratios are much higher than expected for pre-nuclear era (between 10^{-12} and 6×10^{-13}) and due to atmospheric nuclear weapons testing (10^{-10}). These levels account for <0.05% and <5% of the total ^{129}I inventory in this region respectively.

A linear relationship has been established for ^{127}I concentrations in seawater and salinity in the Baltic Sea, enabling the estimation of concentration factors for ^{127}I in *F. vesiculosus*, ranging from 2500 to 13,300, and showing a slightly decreasing trend with salinity. It has been assumed that these concentration factors equally apply to ^{129}I, enabling the reconstruction its concentrations in seawater and the study of their spatial distribution.

The high levels of ^{129}I and ^{129}I/^{127}I in the Kattegat and their gradually decrease towards the Baltic Sea indicate that the ^{129}I discharged from the reprocessing plants at Sellafield and La Hague has been transported to the Kattegat from the North Sea and partly further transported to the Baltic Sea via the Belt Sea. Hence, the most important contribution to the ^{129}I inventory in the Baltic Sea area comes from the releases of both reprocessing plants. This result was also supported by the ^{129}I/^{137}Cs ratios in *F. vesiculosus* and seawater. This ratio in *Fucus* was mediated by concentration factors for ^{137}Cs, which decreased with salinity as a result of the effect of the dominant competitive cations.

With respect to the Chernobyl accident, ^{129}I concentrations in samples collected in 1986 are not much higher than those expected in less contaminated samples from 1982. This supports the view that the contribution of the Chernobyl accident to ^{129}I in the Baltic region was not significant. In contrast, the ^{137}Cs activity concentrations in samples collected in 1986 are much higher than those in corresponding samples collected in 1982, confirming that the Chernobyl accident was the main source of ^{137}Cs to the Baltic Sea area.

Acknowledgements

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