



# Influence of releases of $^{129}\text{I}$ and $^{137}\text{Cs}$ from European reprocessing facilities in *Fucus vesiculosus* and seawater from the Kattegat and Skagerrak areas



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## HIGHLIGHTS

- $^{129}\text{I}$  content was measured in seaweed collected in 2007 in the west Swedish coast.
- $^{129}\text{I}$  contents were found to be highest in the Skagerrak area.
- Concentration factor (CF) for  $^{129}\text{I}$  was found to be  $(10563 \pm 1038)$ .

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## ABSTRACT

$^{129}\text{I}$  is a very long-lived radionuclide ( $T_{1/2} = 15.7 \times 10^6$  years) that is present in the environment because of natural and anthropogenic sources. Compared to the pre-nuclear era, large amounts of  $^{129}\text{I}$  have been released to the marine environment, especially as liquid and gaseous discharges from two European reprocessing facilities located at Sellafield (England) and La Hague (France). The marine environment, i.e., the oceans, is the major source of iodine. Brown seaweed accumulates iodine at high levels up to 1.0% of dry weight, and therefore they are ideal bioindicators for studying levels of  $^{129}\text{I}$ . In this work,  $^{129}\text{I}$  concentrations have been determined in seaweed *Fucus vesiculosus* and seawater collected in the Kattegat and Skagerrak areas in July 2007. The resulting data were evaluated in terms of  $^{129}\text{I}$  concentrations and  $^{129}\text{I}/^{137}\text{Cs}$  ratios.  $^{129}\text{I}$  concentrations were found to be in the order of  $(44\text{--}575) \times 10^9$  atoms  $\text{g}^{-1}$  in seaweed and  $(5.4\text{--}51) \times 10^9$  atoms  $\text{g}^{-1}$  in seawater, with an enhancement in the Skagerrak area in comparison to the Kattegat area. Iodine-129 concentrations in both seaweed and seawater were used to determine the concentration factor of iodine in brown seaweed *F. vesiculosus*.

The high levels of  $^{129}\text{I}$  and  $^{129}\text{I}/^{137}\text{Cs}$  ratios in the Skagerrak area and their gradually decreasing trend to the Kattegat indicates that the most important contribution to the  $^{129}\text{I}$  inventory in those areas comes from Sellafield and La Hague reprocessing plants.

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## 1. Introduction

The only natural stable isotope of iodine is  $^{127}\text{I}$ . The marine environment is the major source of iodine present on the Earth's surface, with an average concentration of  $\sim 60$   $\mu\text{g}$  iodine  $\text{L}^{-1}$  in

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seawater. Iodine is released by volatilization from the marine environment to the atmosphere mainly as organic iodine (mostly iodomethane). Living marine organisms such as marine algae, phytoplankton and aerobic bacteria play the major roles in transformation of iodine to volatile organic forms (Bell et al., 2002).

$^{129}\text{I}$  is the only naturally-occurring radioactive isotope of iodine having a very long half-life for which the natural abundances have been altered in a significant way by human actions. Iodine-129 ( $T_{1/2} = 15.7 \times 10^6$  years) is produced in nature by cosmic rays interactions with Xe in the atmosphere, by spontaneous fission of  $^{238}\text{U}$

and  $^{235}\text{U}$  in the lithosphere (Eisenbud and Gesell, 1997) and, to a minor extent, by neutron-induced reactions on  $^{128}\text{Te}$  and  $^{130}\text{Te}$  in the Earth crust (NCRP, 1983). These processes lead to typical isotopic ratios  $^{129}\text{I}/^{127}\text{I}$  between  $10^{-12}$  (Kilius et al., 1992) and  $6 \times 10^{-13}$  (Fabryka-Martin et al., 1985). The natural inventory of  $^{129}\text{I}$  has been estimated to be  $\sim 230$  kg, most of which resides in the deep

oceans (Rao and Fehn, 1999).  $^{129}\text{I}$  has also been released to the environment from anthropogenic sources during the past 60 years during fission processes associated with civil and military nuclear activities and accidents, such as that of Chernobyl (April 26th, 1986). An estimated 50–150 kg of  $^{129}\text{I}$  was released to the atmosphere during the main period (1945–1964) of nuclear weapons

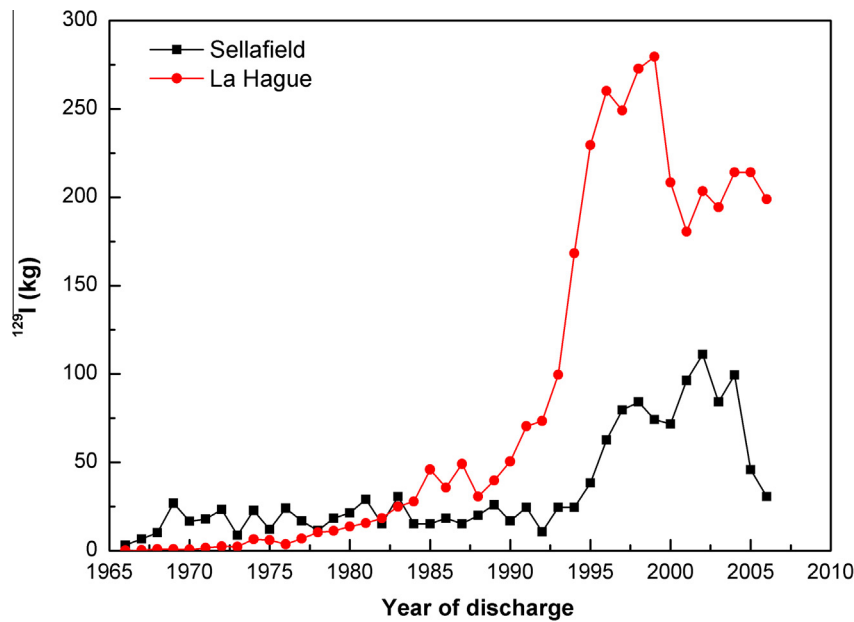


Fig. 1. Annual  $^{129}\text{I}$  release to the sea from the reprocessing facilities in Sellafeld and La Hague. Compilation until 2004 from López-Gutiérrez et al. (2004) and the rest by Monitoring our Environment (2006) and Areva Rapport Environnement (2006).

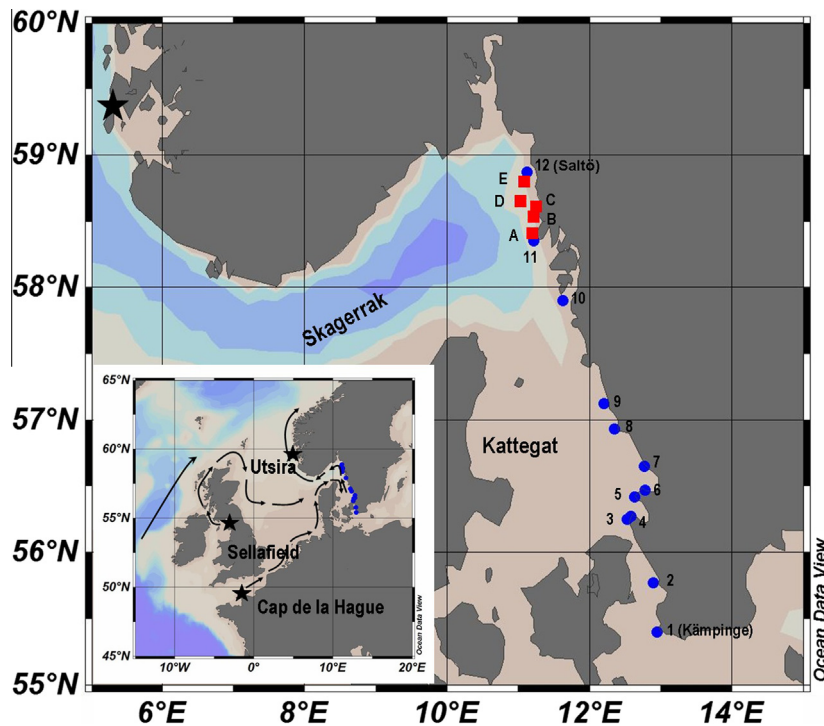


Fig. 2. Sampling sites (blue dots) of *Fucus vesiculosus* and seawater extracted from Lindahl et al. (2003). Red squares represent seawater sampling sites from Aldahan et al. (2007b). The small map in the lower left corner shows the location of Sellafeld and La Hague reprocessing facilities with respect to the sampling sites and major transport currents in the North Sea for radioactive contaminants. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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  $^{129}\text{I}$  are routine discharges from nuclear fuel reprocessing plants at La Hague, France, and Sellafield, England (Fig. 1). It has been estimated that, by 2004, 1371 kg of  $^{129}\text{I}$  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  $^{129}\text{I}$  had been released from La Hague plant as liquid discharges into the English Channel, with an additional 68 kg released in gaseous form (López-Gutiérrez et al., 2004; Aldahan et al., 2007a). The relatively long residence time (30 kyr) of iodine in the marine environment (Chai and Zhu, 1994), as well as the continuous releases from nuclear fuel reprocessing facilities, makes this isotope a suitable transient tracer for the study of marine biogeochemical cycle of stable iodine. 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). As part of this transport/dispersion, there is an inflow of saline seawater from the North Sea that enters the Skagerrak, bringing anthropogenic pollutants from the European reprocessing facilities to this area (Fig. 2).  $^{129}\text{I}$  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  $\text{CH}_3\text{I}$  (Aldahan et al., 2006), and iodate (Lehto et al., 2012).

Fallout following the Chernobyl accident did not increased significantly  $^{129}\text{I}$  concentrations in the Baltic Sea (Gómez-Guzmán et al., 2013a), but it strongly increased  $^{137}\text{Cs}$  concentrations, regulated by the outflow of Baltic waters to the Swedish west coast (Lindahl et al., 2003).

Seaweed is an excellent biological indicator for monitoring radionuclides and stable elements in the marine environment (Druehl et al., 1988; Cooper et al., 1998; Hou et al., 2000a), as they are capable to concentrate these with concentration factors (CF = biota activity concentration/seawater activity concentration) ranging from 1 to about  $10^5$  dependent on element, species and chemical form (Hou and Yan, 1998). The brown seaweed *Fucus vesiculosus* has a particularly high uptake of iodine in seawater, with reported average CF of 10000 in open ocean waters (IAEA, 2004).

The purpose of this study was to examine the concentrations of  $^{129}\text{I}$  and  $^{137}\text{Cs}$  in the marine environment (seaweed and seawater) along the Swedish west coast, contaminated by anthropogenic radionuclides from western European nuclear reprocessing plants and Chernobyl accident debris.  $^{129}\text{I}/^{137}\text{Cs}$  ratios were also evaluated. Iodine-129 concentrations in both seaweed and seawater were used to determine the concentration factor of iodine in brown seaweed *F. vesiculosus*.

## 2. Material and methods

### 2.1. Sampling

Seaweed and seawater samples were collected in July 2007 along the Swedish west coast (Fig. 2) comprising the Kattegat and Skagerrak areas, in order to provide a representative geographical distribution. In the past, these and nearby locations have been widely used for monitoring various radionuclides in seaweed and seawater (Mattsson and Erlandsson, 1991; Carlson and Holm, 1992; Holm, 1995; Lindahl et al., 2003). At each site, approximately

1 kg (dry weight) of the brown seaweed *F. vesiculosus* was collected at low-tide at depths up to 1 m and not more than 3–5 m from the shore and with a mix of whole young and old plants. The samples were first air dried and then dried at 80 °C, grinded and homogenised. Seawater samples were collected in polyethylene bottles, pre-filtered to remove particulate matter, firmly sealed, and stored in a cold dark room. Before storage, all seawater samples were slightly acidified to pH around 5 with  $\text{HNO}_3$  to prevent the formation of algae and the hydrolysis of the cations. As reported by Buraglio et al. (2000), these storage conditions do not introduce significant changes in the  $^{129}\text{I}$  concentration.

The studied area comprises two main and distinct marine areas. Sampling points 3–9 belongs to the relatively shallow Kattegat area which has an estuarine-like circulation. The almost fresh water from the Baltic Sea feeds the outflow which overlays the inverse and more salty water flow from the Skagerrak (coming from the North Sea). The net balance is a mean flow of  $15 \times 10^3 \text{ m}^3 \text{ s}^{-1}$  going out from the Baltic Sea. The estimated residence time of the surface water is a few weeks and that of the bottom water is a few months (OSPAR COMMISSION, 2000). Sampling sites 11 and 12 belongs to the deeper Skagerrak area. Its topography, together with the general anti-clockwise circulation of the North Sea, causes most of the water in the North Sea to pass through this area. This main circulation pattern is complemented with the above described mass exchange with the Kattegat area. The residence time of the surface waters here is about one month, and of several years for the deepest waters (>500 m depth). Sampling site 10 is in the transition zone of those two areas, while sampling sites 1–2 belongs to the Arkona Basin, South of Kattegat, and separated from it by narrow straits.

As all sampling sites (1–12) were within the intertidal area along the coastline, it is expected they will be essentially affected by the less salty surface waters flowing from the Baltic Sea and also supplied by riverine inputs and continental runoff, and, by a lesser extent, by the limited mixing with the deeper and more salty waters flowing from Skagerrak.

### 2.2. Radiochemical procedure

All seaweed samples were digested using an alkaline fusion method (Brown et al., 2005): 0.5 g of the solid material was mixed with 5 mg of  $^{127}\text{I}$  carrier solution (Woodward Iodine with ratio  $^{129}\text{I}/^{127}\text{I} \approx 7 \times 10^{-14}$ , as reported by Roberts et al. (1997)), and 6 mL of a 30% KOH and 3%  $\text{KNO}_3$  solution as a fluxing agent in a crucible. The crucible was then placed in an oven at 90 °C and allowed to evaporate to dryness, after which it was covered and transferred to a muffle furnace preheated to 550 °C. Fusion was completed by heating the sample for 60 min. After this, the crucible was cooled to ambient room temperature. The fused solid sample was then dissolved with water (>18 MΩ cm) produced in a Milli-Q apparatus (Millipore Co., Bedford, MA, USA). The iodine extraction was then made with an organic compound (Gómez-Guzmán et al., 2010): iodine was then extracted three times with 10 mL  $\text{CHCl}_3$  and then back-extracted into aqueous solution by addition of 20 mL 0.1 M  $\text{NaHSO}_3$ . The solution was then stirred and the  $\text{SO}_3^{2-}$  and  $\text{SO}_4^{2-}$  ions were precipitated by addition of 10 mL of saturated  $\text{Ba}(\text{NO}_3)_2$  solution followed by centrifugation. Finally, iodine was precipitated as AgI by adding 10 mL 0.1 M  $\text{AgNO}_3$  solution, washed twice with distilled water and dried at 60–80 °C. The AgI was mixed with about the same volume of Ag powder and loaded in a copper target holder for the measurement of  $^{129}\text{I}$ .

The chemical extraction of iodine from water is usually based on the solubility of  $\text{I}_2$  in  $\text{CCl}_4$ . However, here we preferred to use  $\text{CHCl}_3$  instead of  $\text{CCl}_4$  because of its lower chemical toxicity. The procedure is straightforward: 5 mg of  $^{127}\text{I}$  carrier solution were

added to 400 mL of seawater sample and iodine was then extracted using the same procedure previously described for seaweed samples starting from the extraction with organic compound. Finally, iodine was precipitated as AgI in the same way as previously, and mixed with about the same volume of Ag powder to produce an  $^{129}\text{I}$  sample.

Before the extraction with organic solvent, all iodine compounds (mainly iodate and iodide) must be converted to  $\text{I}_2$ . Iodate was reduced to iodide by using 1 M sodium bisulfite to uniform all iodine chemical species in the sample and then oxidizing iodide to molecular iodine  $\text{I}_2$  by sodium nitrite at  $\text{pH} < 2$ .

### 2.3. Determination of $^{129}\text{I}$ and $^{137}\text{Cs}$

Determination of  $^{129}\text{I}/^{127}\text{I}$  ratios in seaweed and seawater samples was conducted at the CNA AMS (Accelerator Mass Spectrometry) tandem accelerator (1MV Tandetron, High Voltage Engineering Europe, Holland). Details about the measurement and the facility have been previously described (Klein et al., 2006, 2007; Gómez-Guzmán et al., 2012), so only a short description will be given here. A beam of negative iodine ions is extracted from the AgI+Ag target using the  $\text{Cs}^+$  high-intensity sputter source at 35 keV. At the terminal of the tandem (1MV) negative iodine ions are changed to positive iodine ions by the stripper (pressure at  $6 \times 10^{-3}$  mbar of argon, which corresponds to a mass thickness of about  $0.15 \mu\text{g cm}^{-2}$ ) and they are then accelerated. Following acceleration, selected positive ions (charge state +3) are analyzed by mass spectrometer. The  $^{127}\text{I}$  current beam is measured in a Faraday cup, and  $^{129}\text{I}^{3+}$  ions are counted in a gas ionization chamber. AMS measurements were performed relative to a standard with known  $^{129}\text{I}/^{127}\text{I}$  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  $^{129}\text{I}/^{127}\text{I} \sim 10^{-9} - 10^{-11}$ . On the other hand, chemical blanks to control contamination in the sample preparation process were prepared exactly in the same way as normal samples. The AMS  $^{129}\text{I}/^{127}\text{I}$  ratios for the blank were around  $3 - 4 \times 10^{-13}$  being, in the worst case, around 5% of the isotopic  $^{129}\text{I}/^{127}\text{I}$  ratio measured in the real samples.

Before the measurement of  $^{129}\text{I}$ , caesium-137 was determined in seaweed samples. Analysis of seaweed is not different to other matrixes: there is a density correction factor that has to be taken into account, although it is not very high for  $^{137}\text{Cs}$  (662 keV gamma line). The method is nowadays a standardized procedure (Cooper et al., 2003; Hurtado et al., 2006). After being dried at  $80^\circ\text{C}$ , the samples were grinded and packed in calibrated geometries, and

$^{137}\text{Cs}$  activity was measured by HPGe (High Purity Germanium detectors) for 2 d by gamma spectrometry.

## 3. Results and discussion

### 3.1. $^{129}\text{I}$ and $^{137}\text{Cs}$ in seaweed *F. vesiculosus* and $^{129}\text{I}$ in seawater

Concentrations of  $^{129}\text{I}$  in seaweed and seawater samples collected in 2007 around the Western Swedish coast were found to range from  $(44 \pm 2) \times 10^9$  atoms  $\text{g}^{-1}$  and  $(5.4 \pm 0.2) \times 10^9$  atoms  $\text{L}^{-1}$  in position 1 (Kämpinge) respectively, to  $(575 \pm 22) \times 10^9$  atoms  $\text{g}^{-1}$  and  $(51 \pm 3) \times 10^9$  atoms  $\text{L}^{-1}$  in positions 12 (Saltö) and 11 (Smögen) respectively (Table 1, Fig. 3). From Table 1 and Fig. 3 it can be clearly seen that there is a geographical enhancement of  $^{129}\text{I}$  concentrations in samples collected in the Skagerrak area with respect to seaweed samples collected in the Kattegat area. 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, a minor fraction of this contaminated seawater is then transferred through the Kattegat into the Baltic Sea, although most of it returns to the North Sea (Hou et al., 2000b). The same enhancement in the  $^{129}\text{I}$  concentration has been also observed in the seawater samples, as depicted in Fig. 3 (bottom).

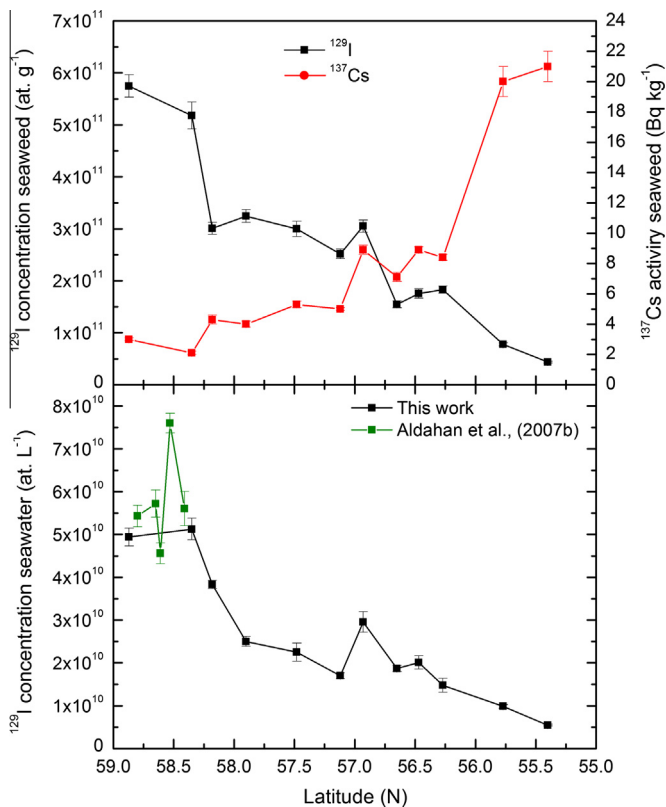
To compare the seaweed  $^{129}\text{I}$  concentrations on a global scale, Table 2 presents a comparison of  $^{129}\text{I}$  levels in seaweed from other locations worldwide. For example, Osterc and Stibilj (2008) reported results obtained in seaweed *Fucus vesiculosus* sampled during the summers of 2005 and 2006 in the coastline of the Adriatic Sea.  $^{129}\text{I}$  concentrations in these samples ranged between  $(1.5 - 3.4) \times 10^9$  atoms  $\text{g}^{-1}$ . Results reported by Fréchu and Calmet (2003) and obtained in seaweed sampled between 1998 and 1999 in Gourey, sited in the vicinity of La Hague nuclear reprocessing plant, show very high  $^{129}\text{I}$  concentrations, ranging between  $(1.4 - 3.2) \times 10^{13}$  atoms  $\text{g}^{-1}$ , between two and three orders of magnitude higher than that found in this study. Hou et al. (1999) reported  $^{129}\text{I}$  concentrations in seaweed sampled in Denmark and Iceland between 1988 and 1992. These values ranged between  $(0.6 - 115) \times 10^9$  atoms  $\text{g}^{-1}$ , and were in the same order of magnitude as found in this study. Our group has previously reported  $^{129}\text{I}$  concentrations in seaweed collected in 1982 and 1986 in some of the sampling sites used in this study (Gómez-Guzmán et al., 2013a), ranging those concentrations between  $(0.82 - 5.89) \times 10^9$  atoms  $\text{g}^{-1}$  in 1982 and  $(1.33 - 38.8) \times 10^9$  atoms  $\text{g}^{-1}$  in 1986. Finally, (Keogh et al., 2007) have measured  $^{129}\text{I}$  concentrations in *Fucus* collected along the Irish coast during three surveys in 1985, 1994 and 2003. The data revealed an enhancement of approximately

**Table 1**

$^{129}\text{I}$  concentration in *Fucus vesiculosus* and in seawater samples, and  $^{137}\text{Cs}$  activities in *Fucus vesiculosus* from the Swedish west coast collected during the summer of 2007. Quoted uncertainties are  $\pm 1\sigma$ . Salinity data interpolated from mean annual data given by Feistel et al. (2010).

No.	Stations	Locations	Salinity (‰)	Seaweed		Seawater
				$^{129}\text{I}$ ( $\times 10^9$ atoms $\text{g}^{-1}$ )	$^{137}\text{Cs}$ (Bq $\text{kg}^{-1}$ )	$^{129}\text{I}$ ( $\times 10^9$ atoms $\text{L}^{-1}$ )
1	Kämpinge	55.40°N, 12.95°E	10.0	44 ± 2	21 ± 1	5.4 ± 0.2
2	Barsebäck	55.77°N, 12.90°E	13.5	78 ± 3	20 ± 1	10 ± 1
3	Lerhamn	56.25°N, 12.53°E	16.6	183 ± 7	8.4 ± 0.2	15 ± 2
4	Arild	56.27°N, 12.58°E	18.0	175 ± 9	8.9 ± 0.2	20 ± 2
5	Torekov	56.42°N, 12.63°E	18.2	155 ± 7	7.1 ± 0.3	19 ± 1
6	Kattvik	56.47°N, 12.78°E	19.5	305 ± 12	8.9 ± 0.3	30 ± 2
7	Görvik	56.65°N, 12.77°E	21.0	252 ± 9	5.0 ± 0.1	17 ± 1
8	Glommen	56.93°N, 12.35°E	24.0	300 ± 15	5.3 ± 0.2	23 ± 2
9	Getterön	57.12°N, 12.20°E	25.0	325 ± 12	4.0 ± 0.2	25 ± 1
10	Marstrand	57.90°N, 11.63°E	25.5	301 ± 11	4.3 ± 0.3	38 ± 1
11	Smögen	58.35°N, 11.22°E	26.0	518 ± 26	2.1 ± 0.1	51 ± 3
12	Saltö	58.87°N, 11.12°E	25.5	575 ± 22	3.0 ± 0.1	49 ± 2





**Fig. 3.** Top:  $^{129}\text{I}$  concentrations (black squares) and  $^{137}\text{Cs}$  activities (red dots) in seaweed *Fucus vesiculosus* sampled around the western Swedish coastline in 2007. Bottom:  $^{129}\text{I}$  concentrations in seawater collected in the same sampling sites (black squares) and reported by Aldahan et al. (2007b) (green triangles). See text for explanation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

two orders of magnitude in the  $^{129}\text{I}$  concentration in seaweed sampled along the northeast coast of Ireland, with respect to concentrations in seaweed taken from the west coast at similar times.  $^{129}\text{I}$  concentrations ranged  $(6\text{--}870) \times 10^9$  atoms  $\text{g}^{-1}$  in the northeast coast sampling sites and  $(0.9\text{--}94) \times 10^9$  atoms  $\text{g}^{-1}$  in the west coast sampling sites.

Activity concentrations of  $^{137}\text{Cs}$  in seaweed samples varied from 2.1 to 3.0  $\text{Bq kg}^{-1}$  at the westernmost localities (11 and 12), to 20  $\text{Bq kg}^{-1}$  at locations close to the Baltic Sea (1 and 2). Thus, the  $^{137}\text{Cs}$  concentration increased by a factor of  $\sim 10$  depending on the geographical distribution of the seaweed samples (Table 1 and Fig. 3 top). The enhancement found in the  $^{137}\text{Cs}$  activity is the opposite found in the  $^{129}\text{I}$  concentrations. In the case of  $^{137}\text{Cs}$ , the most important source of this radionuclide in the Baltic Sea area is the Chernobyl accident, which occurred in April 1986. This accident released a large amount of radioactive material, including about 6 kg of  $^{129}\text{I}$  (Gallagher et al., 2005) and about 4500 TBq of  $^{137}\text{Cs}$  (CEC, 1991). This estimation 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, with the areas surrounding the Bothnian Sea and the eastern Gulf of Finland being the most contaminated. If we extrapolate this behavior in the  $^{137}\text{Cs}$  activity to our sampling area, this would explain the enhancement found in the samples closer to the Baltic Sea. HELCOM (2009) has estimated the total input of  $^{137}\text{Cs}$  to the Baltic Sea originating from Sellafield and La Hague facilities to be 250 TBq, which is much lower than the  $^{137}\text{Cs}$  originated by the Chernobyl accident and even lower than the 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.

The values obtained for  $^{129}\text{I}$  concentration in seawater samples are up to three orders of magnitude higher than values reported for Japan in 2006 (Suzuki et al., 2008), up to two orders of magnitude

**Table 2**  
 $^{129}\text{I}$  concentrations in various seaweeds sampled worldwide.

Seaweed	Sampling zone		$^{129}\text{I}$ ( $\times 10^9$ atoms $\text{g}^{-1}$ )	Reference
	Date	Location		
<i>F. vir.</i>	Sept. 2005	Slovenia	$2.0 \pm 0.6$	Osterc and Stibilj (2008)
<i>F. vir.</i>	June 2006	Italy	$1.5 \pm 1.1$	
<i>F. vir.</i>	Oct. 2006	Croatia	$3.4 \pm 1.1$	
<i>F. ves.</i>	Oct. 1998	Goury, France	$14000 \pm 900$	Fréchou and Calmet (2003)
<i>F. ves.</i>	March 1999	Goury, France	$32000 \pm 2300$	
<i>F. ves.</i>	Dec. 1988	Iceland	$0.6 \pm 0.04$	Hou et al. (1999)
<i>F. ves.</i>	Nov. 1989	Klint, Denmark	$62.1 \pm 4.6$	
<i>F. ves.</i>	April 1992	Klint, Denmark	$115.3 \pm 10.4$	
<i>F. ves.</i>	March 1994	Klint, Denmark	$250.6 \pm 0.9$	Hou et al. (2000b)
<i>F. ves.</i>	March 1996	Klint, Denmark	$391.0 \pm 2.0$	
<i>F. ves.</i>	Jan. 1998	Klint, Denmark	$413.8 \pm 2.4$	
<i>F. ves.</i>	1982	8	$0.82 \pm 0.05$	Gómez-Guzmán et al. (2013a)
<i>F. ves.</i>	1982	28	$5.89 \pm 0.15$	
<i>F. ves.</i>	1986	5	$1.33 \pm 0.06$	
<i>F. ves.</i>	1986	33	$38.8 \pm 2.1$	
<i>F. ves.</i>	1985	Ireland, east coast	29–330	Keogh et al. (2007)
<i>F. ves.</i>	1985	Ireland, west coast	0.9–9.3	
<i>F. ves.</i>	1994	Ireland, east coast	6–386	
<i>F. ves.</i>	1994	Ireland, west coast	3.2–94	
<i>F. ves.</i>	2003	Ireland, east coast	15–870	
<i>F. ves.</i>	2003	Ireland, west coast	1.7–7.1	
<i>F. ves.</i>	2007	1	$44 \pm 2$	This work
<i>F. ves.</i>	2007	12	$575 \pm 22$	

*F. ves.*: *Fucus vesiculosus*.

*F. vir.*: *Fucus viriosoides*.

**Table 3**  
Concentrations of  $^{129}\text{I}$  determined in surface seawater in this study compared to values for seawater from various places.

Sampling zone		$^{129}\text{I}$ ( $\times 10^9$ atoms $\text{L}^{-1}$ )	Reference
Date	Location		
2009	Adriatic Sea	$5.7 \pm 2.9$	Osterc and Stibilj (2012)
1999	Greenland	$0.65 \pm 0.09$	Hou (2004)
2005	English Channel	375	Hou et al. (2007)
2005	North Sea	228	Hou et al. (2007)
2006	Japan (Toyama Bay)	$0.018 \pm 0.005$	Suzuki et al. (2008)
2010	Iceland and Irminger Sea	0.65–3.37	Gómez-Guzmán et al. (2013b)
2001	Kattegat and Skagerrak	4.56–7.60	Aldahan et al. (2007b)
2000	Kattegat and Skagerrak	2.8–268	Hou et al. (2002)
2006–2007	Kattegat and Skagerrak	2.5–168	Yi et al. (2011)
2005	Ireland (east coast)	65–690	Keogh et al. (2007)
	Ireland (west coast)	3.2–4.7	
2007	Kattegat and Skagerrak	5.4–51	This work

higher than values reported for Greenland in 1999 (Hou, 2004) and up to one order of magnitude lower than reported for the English Channel and North Sea (Hou et al., 2007; Keogh et al., 2007) (Table 3), which are under the direct influence of liquid  $^{129}\text{I}$  discharges from European fuel reprocessing plants. Hou et al. (2002) investigated the levels of  $^{129}\text{I}$  in seawater samples from Bornholm, the Belt Sea, Kattegat, Skagerrak and North Sea collected in November 1999 and June 2000. They found  $^{129}\text{I}$  concentrations in the range from 2.8 to  $268 \times 10^9$  atoms  $\text{L}^{-1}$ , with lower values in the area of the Baltic Sea and higher values in the area of the North Sea. Our values are consistent with those previously reported by Hou et al. (2002) and Yi et al. (2011), and show the same trend. They noted that the North Sea is greatly influenced by liquid discharges from the Sellafield and La Hague reprocessing plants, because the radionuclides released are transported by the Atlantic current first to the North Sea, then further to the Norwegian coast and the Arctic by the Norwegian Coastal current. The Baltic Sea is partially isolated from the influence of these currents, so the levels of  $^{129}\text{I}$  in seawater decrease as one moves from the Kattegat to the Skagerrak areas, which connect the Baltic Sea with the North Sea. Aldahan et al. (2007b) reported  $^{129}\text{I}$  concentrations ranging  $(45.6\text{--}76.0) \times 10^9$  atoms  $\text{L}^{-1}$  in surface seawater samples collected at positions located between numbers 11 and 12 of this study (those labeled A to D in Fig. 3 bottom). These concentrations are consistent with our data and, when plotted (Fig. 3 bottom) show a clear trend of decreasing  $^{129}\text{I}$  concentrations as the sampling site becomes closer to the Baltic Sea proper. This trend in the  $^{129}\text{I}$  concentrations supports the idea that the Baltic Sea is partially isolated from the influence of currents coming from the North Sea, greatly influenced by liquid discharges from the Sellafield and La Hague reprocessing plants, and is similar to the trend found in the  $^{129}\text{I}$  concentrations in the seaweed samples.

### 3.2. $^{129}\text{I}/^{137}\text{Cs}$ ratio in seaweed *F. vesiculosus* and in seawater

The ratios of  $^{129}\text{I}/^{137}\text{Cs}$  in both seaweed and seawater have been considered as an index of water mass transport and to distinguish the sources of radionuclides (Smith et al., 1998; Raisbeck and Yiou, 1999; Hou et al., 2000c). Based on the measured  $^{129}\text{I}$  and  $^{137}\text{Cs}$  concentrations in seaweed the ratio  $^{129}\text{I}/^{137}\text{Cs}$  can be directly calculated in those samples. The  $^{129}\text{I}/^{137}\text{Cs}$  isotopic ratios in seaweed samples are mediated by the different concentration factors (those for  $^{129}\text{I}$  being on average, larger by a factor 20 than those for  $^{137}\text{Cs}$ ) and by their different trends of variation with salinity (concentration factors for  $^{129}\text{I}$  slightly increase with increasing salinity, while those for  $^{137}\text{Cs}$  sharply decrease with increasing salinity). Gómez-Guzmán et al. (2013a) reported the following log–log relationship between CF for  $^{137}\text{Cs}$  and salinity for the Baltic Sea area ( $R^2 = 0.79$ , confidence level 99%).

$$\log[\text{CF}_{\text{Cs}}] = 3.55 - 1.03 \cdot \log[S] \quad (1)$$

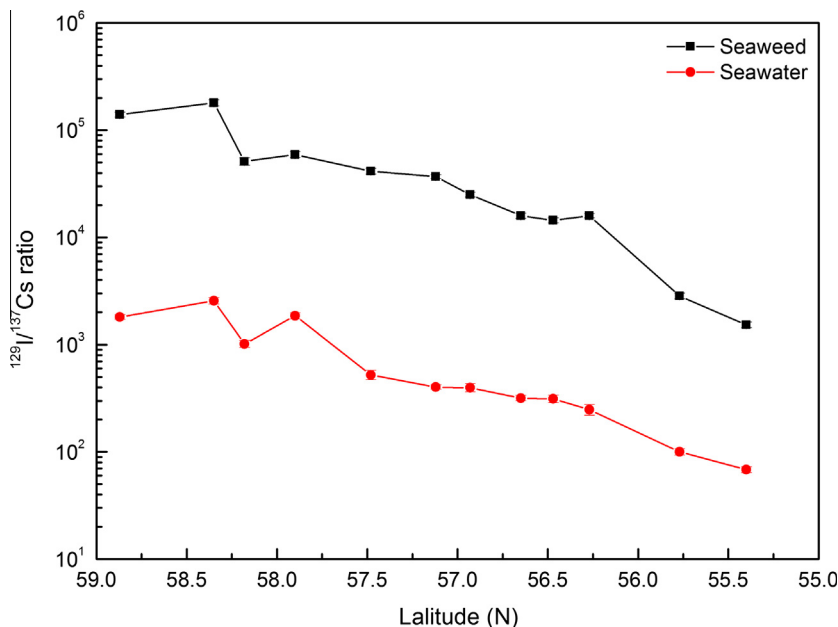
Data on salinity provided in Table 1 can, therefore, be used to infer the concentration factors (CF) for  $^{137}\text{Cs}$  in *F. vesiculosus* in the sampling sites and thus, to calculate the  $^{129}\text{I}/^{137}\text{Cs}$  isotopic ratios in seawater samples. The CF for Cs calculated in this way vary from 144 to  $361 \text{ L kg}^{-1}$  and are in good agreement with those CF reported by HELCOM (2009) for the Baltic Sea area, found to be in the range from  $188 \pm 39 \text{ L kg}^{-1}$  to  $453 \pm 145 \text{ L kg}^{-1}$ , dry weight. Once CF for Cs was calculated in the sampling sites, the  $^{129}\text{I}/^{137}\text{Cs}$  isotopic ratios in seawater samples were straightforward calculated by using Eq. (2),

$$\left[ \frac{^{129}\text{I}}{^{137}\text{Cs}} \right]_{\text{W}} = \frac{[^{129}\text{I}]_{\text{W}} \cdot \text{CF}_{\text{Cs}}}{[^{137}\text{Cs}]_{\text{F}}} \quad (2)$$

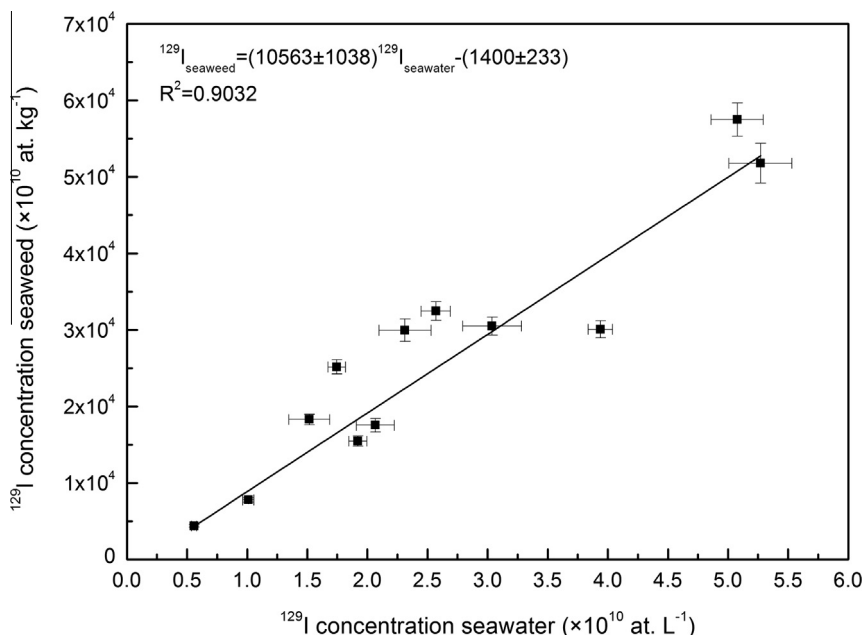
where:  $[^{129}\text{I}/^{137}\text{Cs}]_{\text{W}}$  is the  $^{129}\text{I}/^{137}\text{Cs}$  atom ratio in seawater at the various *Fucus* sampling sites;  $[^{129}\text{I}]_{\text{W}}$  is the  $^{129}\text{I}$  concentration in seawater;  $[^{137}\text{Cs}]_{\text{F}}$  is the  $^{137}\text{Cs}$  concentration in *Fucus* given in Table 1 and  $\text{CF}_{\text{Cs}}$  is the CF previously calculated using Eq. (1) and the salinity data shown in Table 1. The ratios obtained for seaweed and seawater samples are shown in Fig. 4 as a function of the latitude. Maxima and minima values of ratios  $^{129}\text{I}/^{137}\text{Cs}$  in both seawater and seaweed samples are reached in the same sampling locations, with maxima found in location 11 (Smögen), at 180271 and 2563 for seaweed and seawater samples, respectively, and minima found in location 1 (Kämpinge), at 1525 and 68 for seaweed and seawater samples, respectively.

Hou et al. (2000b) reported time-series data for the  $^{129}\text{I}/^{137}\text{Cs}$  atom ratio in seawater collected at Utsira (in the west Norwegian coast and shown in Fig. 1) over the period 1981–1995, with values ranging from 50 in 1981 to 2500 in 1995. Yiou et al. (2002) also reported time-series data for the ratio  $^{129}\text{I}/^{137}\text{Cs}$  over the period 1981–1999 in seawater samples collected at the same location, with values ranging from 70 in 1981 to 10000 in 1999. In both studies, the ratio  $^{129}\text{I}/^{137}\text{Cs}$  in the seawater samples is clearly mediated by the liquid emissions from Cap de la Hague reprocessing facility. In all cases, ratios  $^{129}\text{I}/^{137}\text{Cs}$  are much higher than those attributable to the fall out from nuclear weapon tests (about 1.6) and that in unprocessed fission products ( $\approx 1$ ) (Raisbeck et al., 1995). This indicates that  $^{129}\text{I}$  concentrations are dominated by the discharges from reprocessing plants. For the sampling sites in the inner Kattegat area, the influence of those releases decreases and so  $^{129}\text{I}$  concentrations and ratios  $^{129}\text{I}/^{137}\text{Cs}$  in seawater decrease accordingly, as shown in Figs. 3 and 4, respectively.

The consideration generally accepted is that the main circulation pattern of the Baltic outflow water drifted along the western Norwegian coast into the Barents Sea (Maslowski and Walczowski, 2002) and a large portion of seawater in Utsira has originated from



**Fig. 4.**  $^{129}\text{I}/^{137}\text{Cs}$  atom ratio in seawater (red dots) and in *Fucus vesiculosus* (black squares) at the various sampling sites for 2007. Uncertainties come from the error propagation theory. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

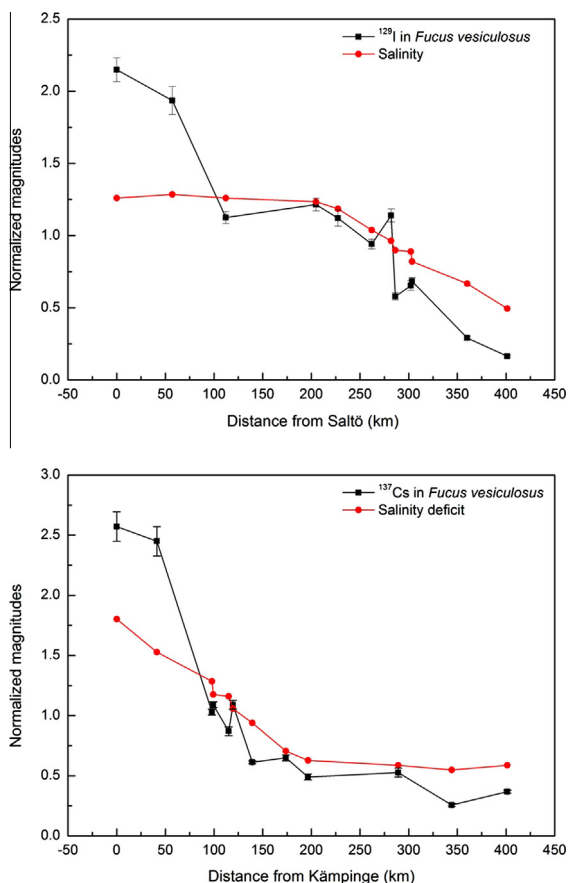


**Fig. 5.**  $^{129}\text{I}$  concentrations in *Fucus vesiculosus* versus  $^{129}\text{I}$  concentrations in seawater for the same sampling sites in 2007.

the Kattegat. This feature can also be confirmed by watching Fig. 2, where a simplified scheme of water masses general circulation in north-western European seas is shown (extracted from Bailly du Bois and Dumas, 2005).

It is interesting to compare the normalized (to the mean value)  $^{129}\text{I}$  distribution in *F. vesiculosus* along the Swedish West coast with the normalized (also to the mean value) salinity distribution shown in Table 1, interpolated from mean annual salinity data given by Feistel et al. (2010). Assuming an Earth's radius of 6370 km it is possible to convert the longitude/latitude data shown in Table 1 for all sampling points to distances measured from the northernmost sampling site (position 12, Saltö). The goal is that salinity, a conservative property, somehow reflects the progressive

mixing of both North-Atlantic and Baltic water masses along the different sampling sites. The behavior shown in Fig. 6 (top) reflects a non-equilibrium situation in which  $^{129}\text{I}$  has a source term in Skagerrak (North Sea waters washing down effluents from Sellafield and La Hague reprocessing plants) with a net transport by advection–diffusion to the Baltic Sea. From here, and following the more salty and denser water flux, it penetrates in the Kattegat area and, finally, in the Arkona basin. The estuarine-like circulation of the Kattegat area, with estimated residence time of the surface water of a few weeks and that of the bottom water of a few months, get a relatively good homogenization in the area, although the Arkona straits create a new natural barrier for the water circulation.



**Fig. 6.** Top: Normalized to the mean  $^{129}\text{I}$  concentrations in seaweed (black squares) and salinity (red dots) around the western Swedish coastline in 2007. Bottom: normalized to the mean  $^{137}\text{Cs}$  concentrations in seaweed (black squares) and salinity deficit  $DS$  (red dots). See text for explanation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

To better understand  $^{137}\text{Cs}$  dynamics we define the *salinity deficit*  $DS$  as the salinity in the sampling sites shown in Table 1 and interpolated from mean annual salinity data, subtracted from the maximum annual salinity (33‰) measured in surface seawaters of the Skagerrak area (Feistel et al., 2010). This index is used to trace the less salty and less dense water masses flowing from the Baltic Sea. As in the case of  $^{129}\text{I}$ , the normalized (to the mean value) *salinity deficit* distribution can be used and compared to the normalized  $^{137}\text{Cs}$  distribution in *F. vesiculosus* along the sampling sites. Distances will be now referred to the southernmost site (position 1, Kämpinge). The result of this comparison is shown in Fig. 6 (bottom), where it can be clearly seen that  $^{137}\text{Cs}$  distribution follows the trend of the *salinity deficit*, indicative of a source term in the Baltic Sea (most likely from the Chernobyl accident) and a relatively attenuated penetration to the North. It is worth to note that part of the  $^{137}\text{Cs}$  is scavenged from the water column by suspended particles and sediments (Charmasson et al., 1998).

### 3.3. Determination of the concentration factor (CF) of iodine in brown seaweed *F. vesiculosus*

The quantity of an element or radionuclide in biological tissue is almost always discussed in terms of concentration, either on a dry or wet weight basis. For modeling purposes, this value is then usually represented in terms of a concentration relative to that of the ambient seawater, traditionally expressed as a concentration factor

(CF). If the seawater concentration is derived in terms of unit volume, the CF is expressed in  $\text{L kg}^{-1}$  (Eq. (3)):

$$\text{CF}(\text{L kg}^{-1}) = \frac{\text{Concentration per unit mass of organism}(\text{atoms kg}^{-1} \text{ or } \text{kg kg}^{-1})}{\text{Concentration per unit volume of seawater}(\text{atoms L}^{-1} \text{ or } \text{kg L}^{-1})} \quad (3)$$

The same concentration factor CF in seaweed is assumed for different iodine isotopes, because it increases only with the atomic radius (Hou and Yan, 1998; Hou et al., 2000c). As a consequence, an element with a larger atomic radius (as iodine) can be easily absorbed by the surface of the seaweed cell and so, seaweed can be highly enriched by this element. The CF (in  $\text{L kg}^{-1}$ ) of iodine in *F. vesiculosus* presented in this study and shown in Fig. 5 were calculated from dry weight  $^{129}\text{I}$  data divided by  $^{129}\text{I}$  concentrations in seawater (Table 1) for year 2007. The resulting CF at  $(10563 \pm 1038)$  is in good agreement with the recommended value (10000) reported by Hou and Yan (1998) and IAEA (2004).

Fréchu and Calmet (2003) calculated stable iodine concentration factors in seaweed samples collected in the vicinity of La Hague using a stable iodine concentration measured in each seaweed sample (dry mass) and assuming the seawater stable iodine concentration to be a constant  $60 \text{ ng L}^{-1}$ . For strands of *Laminaria digitata* they reported a CF value up to 20000 and for *Fucus serratus* 2500. Osterc and Stibilj (2012) has reported averaged iodine CF in *Fucus virsoides* samples collected around the Adriatic Sea in 2009 to be  $40 \pm 10$  for  $^{129}\text{I}$  and  $970 \pm 175$  for  $^{127}\text{I}$ . None of those works reported correlations between CF for iodine in seaweed and salinity of seawater where the samples were collected. This lack of correlation can be also seen when plotting CF for  $^{129}\text{I}$  versus salinity in this work (not shown), with a very weak correlation factor  $R^2 = 0.0179$ .

## 4. Conclusions

$^{129}\text{I}$  in seaweed *F. vesiculosus* and seawater collected in summer 2007 along the Swedish West coast have been measured. The high levels of  $^{129}\text{I}$  in the Skagerrak and their gradually decrease towards the Kattegat are and the Baltic Sea indicate that the  $^{129}\text{I}$  discharged from the reprocessing plants at Sellafield and La Hague has been transported to the Skagerrak from the North Sea and partly further transported to the Kattegat and the Baltic Sea via the Belt Sea. Hence, the most important contribution to the  $^{129}\text{I}$  inventory in those areas comes from the releases of both reprocessing plants. This result is also supported by measured  $^{129}\text{I}/^{137}\text{Cs}$  ratios in *F. vesiculosus* and seawater. This ratio in Fucus was mediated by concentration factors for  $^{137}\text{Cs}$ , which decreased with salinity as a result of the effect of the dominant competitive cations.

Using  $^{129}\text{I}$  concentrations in Fucus and in seawater, concentration factor (CF) of iodine in brown algae *F. vesiculosus* was found to be  $(10563 \pm 1038)$ , in agreement with recommended value (10000) reported by Hou and Yan (1998) and IAEA (2004).

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