

# Iodine-129 and iodine-127 in seawater of the North Sea and precipitation from northern Germany

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## Abstract

Atmospheric nuclear weapons tests, nuclear accidents, and emissions from reprocessing plants have changed the natural abundances of <sup>129</sup>I (T<sub>1/2</sub>=15.7 Ma) in a sustainable manner. Mainly as a consequence of the <sup>129</sup>I releases from the European reprocessing plants, <sup>129</sup>I is in disequilibrium in all environmental compartments of the Western Europe.

Surface water from the North Sea and the English Channel, which were contaminated by the continuous emission from reprocessing plants (La Hague and Sellafield), were analysed for <sup>129</sup>I by accelerator mass spectrometry (AMS) and for <sup>127</sup>I by inductively coupled plasma mass spectrometry (ICP-MS). Samples of seawater were taken during a cruise of the BSH FS-Pelagia between August 20 and September 09, 2009. This paper reports on the results obtained for <sup>129</sup>I and <sup>127</sup>I in 2009 and compares them with those from 2005.

The concentrations of <sup>129</sup>I in seawater of the North Sea are fairly constant with (44 ± 2) ng/g, with exceptions of coastal areas with high biological activity and of areas influenced by influx from rivers and the Baltic Sea. The results of <sup>129</sup>I/<sup>127</sup>I ratios range between 1.5 × 10<sup>-8</sup> and 2.7 × 10<sup>-6</sup>: at least 4 orders of magnitude higher than the natural equilibrium isotopic ratio in the marine hydrosphere of 1.5 × 10<sup>-12</sup>. The highest isotopic ratios are seen in the English Channel east of La Hague. The emissions from La Hague can be followed through the English Channel and subsequently through the North Sea. The variability of the <sup>129</sup>I/<sup>127</sup>I isotopic ratios is exclusively determined by admixture of anthropogenic <sup>129</sup>I. Together with time series of the iodine isotopes in precipitation in Northern Germany, the results demonstrate the dominating role of the liquid releases for the ongoing atmospheric fallout of <sup>129</sup>I in Western Europe. Comparing these results with our earlier studies shows the disequilibrium of <sup>129</sup>I in the environmental compartments.

Keywords: Iodine-129, iodine, isotopic ratios, environmental pathways

## 1. Introduction

The long-lived radionuclide of iodine <sup>129</sup>I (half-life 15.7 Ma) is produced in nature by spontaneous fission of uranium in the earth-crust and in oceans, and by interaction of galactic cosmic ray particles with xenon in the stratosphere. The natural equilibrium isotopic ratio <sup>129</sup>I/<sup>127</sup>I either calculated or measured is in the order of 10<sup>-13</sup> [Edwards, 1962; Edwards and Rey, 1968; Kohman and Edwards, 1966; Fehn et al., 1986; Schink et al., 1995; Moran et al., 1998].

The anthropogenic <sup>129</sup>I is mostly produced by fission of <sup>235</sup>U and <sup>239</sup>Pu with isobaric yields for fission induced by thermal neutrons of 0.68 % and 1.6 %, respectively; as well as in the operation of nuclear reactors for research and power production. The civil and military nuclear activities produce and release anthropogenic <sup>129</sup>I into the environment. The releases of <sup>129</sup>I from atmospheric nuclear weapons tests were (43 – 150 kg) [Carter and Moghissi 1977, UNSCEAR 1982; Chamberlain 1991, Eisenbud and Gesell 1997], and those from the Chernobyl accident (2 – 6 kg) [Schmidt et al. 1998, Aldahan et al. 2007]. The amount of anthropogenic <sup>129</sup>I in the atmosphere and in surface waters has continued to increase due to releases from nuclear fuel reprocessing facilities, particularly in Western Europe, from the nuclear reprocessing plants La Hague (F) into the English Channel and Sellafield (UK) into the Irish Sea. The releases of <sup>129</sup>I from these two reprocessing plants are shown in Fig. 3.

The atmospheric releases of <sup>129</sup>I from Sellafield stayed with a relative constant rate of 4 kg a<sup>-1</sup> between 1951 and 2005 with maximum 7 kg in 1980, while those from La Hague increased from 1966 (0.01 kg of <sup>129</sup>I) to 1996 (6 kg of <sup>129</sup>I). It can be seen that a similar amount of <sup>129</sup>I has been released to the atmosphere from the two reprocessing plants with total release 263 kg. The total liquid releases of <sup>129</sup>I

from La Hague and Sellafield increased from 1966 ( $> 0.1 \text{ kg a}^{-1}$ ) nearly continuously until 1990 ( $< 50 \text{ kg a}^{-1}$ ), increased significantly to about  $250 \text{ kg y}^{-1}$  for La Hague and  $69 \text{ kg a}^{-1}$  for Sellafield, then levelled off and even slightly decreased since 2000. As a consequence, the  $^{129}\text{I}$  concentration and thus  $^{129}\text{I}/^{127}\text{I}$  ratio in the English Channel, North-, Baltic- and Irish Sea have been increased. In these seawaters, they have elevated to values of  $10^{-8} - 10^{-6}$ . The highest ratio of  $^{129}\text{I}/^{127}\text{I}$  at  $10^{-06} - 10^{-03}$  was found in the samples collected near the reprocessing plants [Kilius et al. 1994, Yiou et al. 1994, Michel et al. 2012].

In this paper, we report on  $^{127}\text{I}$ ,  $^{129}\text{I}$  and  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios in surface waters of the North Sea and the English Channel, and in precipitation in the northern Germany. Iodine isotopes were determined by ICP-MS and AMS after Matrix separation. Our main goal of the present study was to complement and compare the results of the study StSch 4481 [Nies et al. 2010, Michel et al. 2012], to investigate the distribution and transport pathways of  $^{129}\text{I}$  and  $^{127}\text{I}$  in surface waters of the North Sea and the English Channel and to investigate the source of  $^{129}\text{I}$  in samples of precipitation in the northern Germany.

## 2. Experimental

Surface water was collected from 78 stations in the English Channel and the North Sea in August-September 2009 during cruises of the BSH “Pelagia”, under that for first time water samples from site close to the beach. At three stations 39, 40 and 60, samples surface water (15-32 m) as well of deep water from 100 to 512 m depth were collected. Finally, precipitation sampled in Northern Germany were analysed in 2009.

Sample of precipitation was collected from Lower Saxony, Germany. In order to establish a spatial separation, Lower Saxony was divided into four regions (Michel et al. 2012). The region I was the close proximity to North Sea, northern German lowlands distant from the North Sea (region II), the fringe of the Harz Mountains (region III), and an area of the Elbe River in Lower Saxony in the vicinity of Gorleben (region IV). In addition to open-field precipitation sampled in each region II, through-falling precipitation was collected; through-falling mean here rain that falls down on forest and interacts with the canopy of the trees.

The samples water were filled into clean polyethylene bottles and added two pills of NaOH to hydrolyze organic species, to stabilize the iodine and to transform the iodine species to iodate until analysis [Michel et al. 2012]. We analysed 1-10 ml of filtered waters (through  $0.45 \mu\text{m}$  filter paper) for their  $^{127}\text{I}$  concentration by ICP-MS (Thermo X7, Thermo Electron Corp) at the institute of inorganic chemistry, Leibniz University of Hanover, after 2-10 fold dilution in 25% TMAH. The detection limit was  $0.1 \mu\text{g kg}^{-1}$  and relative standard deviations (RSD) were 1 – 5 %. For the AMS measurement of  $^{129}\text{I}$  iodine matrix separation was necessary in the aqueous sample and used ion exchange resin. About 150 ml of original sample water were used for extraction iodine as AgI. All iodine species were first oxidized with 5 ml of  $\text{Ca}(\text{ClO})_2$  to iodate and then reduced with 20 ml  $[\text{NH}_2\text{OH}]\text{HCl}$  and 10 ml  $\text{NaSO}_3$  to iodide. After the separation with using a DOWEX<sup>®</sup> 1x8 analytical grade ion exchange resin, about 3.0 mg of stable iodine (as NaI prepared from Woodward iodine) added as a carrier. The iodine was precipitated as AgI. Finally, the dried AgI precipitates were mixed with silver powder (AgI:I 1:4 by weight) and pressed into titan targets for AMS measurement.

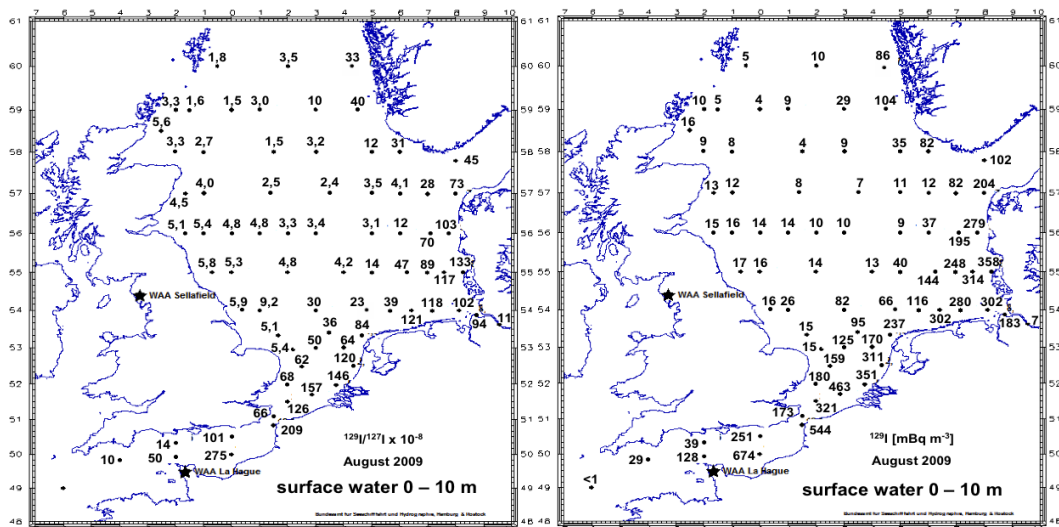
The  $^{129}\text{I}/^{127}\text{I}$  ratio was determined by 0.5 MV “Tandy” accelerator of the PSI / ETH Zürich AMS facility for the selection of charge state +3. The  $^{129}\text{I}/^{127}\text{I}$  machine background was  $(3 - 5) \times 10^{-13}$ . The  $^{129}\text{I}$  blank value of the total analyses were determined with Woodward iodine as trace catcher and were  $1.5 \times 10^{-15} \text{ g}$  and RSD 1 – 18 %.

## 3. Results and Discussion

### 1. $^{127}\text{I}$ , $^{129}\text{I}$ and salinity in the North Sea

The concentration of  $^{127}\text{I}$ , the activity concentration of  $^{129}\text{I}$ , the ratios of  $^{129}\text{I}/^{127}\text{I}$  and salinity in North Sea are listed in Table 1. It can be seen that the stable iodine concentration was justly constant with value of  $44 \text{ ng g}^{-1}$ . Only in coastal areas have low variation ( $36\text{-}42 \text{ ng g}^{-1}$ ).

The activity concentrations of  $^{129}\text{I}$  show a fairly large spread between ( $4\text{-}674 \text{ mBq m}^{-3}$ ) (Fig. 1, left). So there are up to 100 times difference between the low (station 47: West North of North Sea) and high (station 910: near of La Hague) values. The lowest activity concentrations are a factor of more than 40 higher than those found in the seawater from Hawaii ( $50 \text{ } \mu\text{Bq m}^{-3}$ ) and from the Indian Ocean ( $90 \text{ } \mu\text{Bq m}^{-3}$ ) [Michel et al. 2012]. It has been shown that the highest activity concentrations of  $^{129}\text{I}$  are exhibited in the English Channel and along the coasts of France, Belgium, the Netherland, Germany, and until Denmark with values reaching ( $82\text{-}674 \text{ mBq m}^{-3}$ ). Our values show good agreement with ( $2\text{-}890 \text{ mBq m}^{-3}$ ) in the North Sea and the English Channel in year 2005 [Michel et al. 2012] and ( $280\text{-}420 \text{ mBq m}^{-3}$ ) in the northern part of the English Channel and in the south eastern North Sea [Hou et al. 2007].



**Fig. 1:**  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios (right) and  $^{129}\text{I}$  activity concentrations in  $\text{mBq m}^{-3}$  (left) in the North Sea and the English Channel.

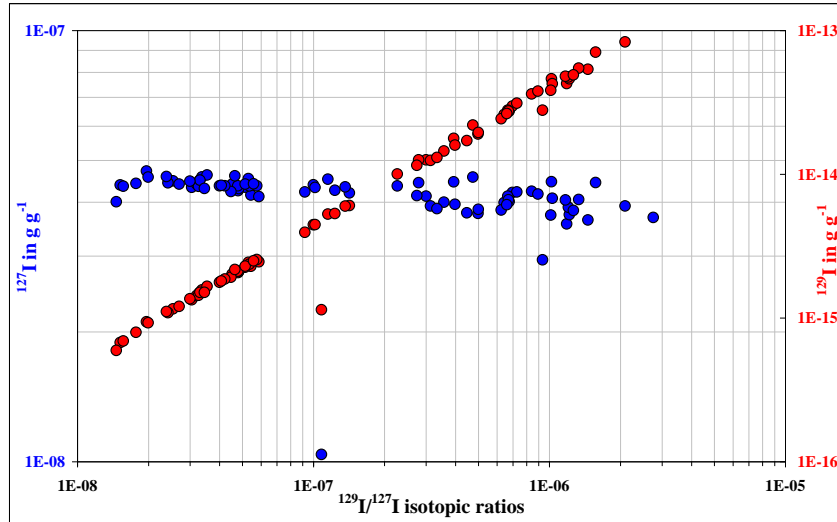
Here we clearly see the high influence of La Hague on the North Sea and English Channel. But the influence of Sellafeld has been showed in eastern and northern parts of North Sea. This can be confirmed with Help the other radionuclides as  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{238,239}\text{Pu}$  and  $^{99}\text{Tc}$ , for more details see the studies Nies et al. 2010 and Michel et al. 2012. The complicated water currents in the North Sea can explain the pattern distribution of  $^{129}\text{I}$ .

The  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios show the same distribution of  $^{129}\text{I}$  in the North Sea and English Channel (Fig. 1, right), because the distribution of  $^{127}\text{I}$  is justly constant in the North Sea.

Fig. 2 shows positive correlations between  $^{129}\text{I}/^{127}\text{I}$  ratios and  $^{129}\text{I}$ .  $^{127}\text{I}$  concentrations are not influence the  $^{129}\text{I}/^{127}\text{I}$  ratios. Also it exhibits the disequilibrium of iodine isotopes in the North Sea. Only the station Stade in the Elbe River and the station Medem in the mouth of the Elbe Estuary show  $^{127}\text{I}$ - and  $^{129}\text{I}$ -concentrations below. Their low  $^{127}\text{I}$  and  $^{129}\text{I}$  concentration of ( $10\text{-}29 \text{ ng g}^{-1}$ ) and ( $1\text{-}28 \text{ fg g}^{-1}$ ), respectively, are explained by the influx of continental surface water from the Elbe River. The highest  $^{129}\text{I}/^{127}\text{I}$  ratios  $2.7 \times 10^{-6}$  were observed in the English Channel east of La Hague (Station 910) and he decrease along the coasts of France, Belgium, the Netherland, Germany and until Denmark (Fig. 1). The lowest  $^{129}\text{I}/^{127}\text{I}$  ratios of  $1.5 \times 10^{-8}$  is least 4 orders of magnitude higher than the natural equilibrium isotopic ratio in the marine hydrosphere of  $1.5 \times 10^{-12}$ . In this study, no relationships between  $^{127}\text{I}$ ,  $^{129}\text{I}$  and salinity are seen

**Table 1:** Salinity,  $^{127}\text{I}$ ,  $^{129}\text{I}$ , and  $^{129}\text{I}/^{127}\text{I}$  ratios isotopic in the North Sea and the English Channel.

Station	Nummer	Latitude (°N) ; Longitude (°E)	Salinity (PSU)	$^{129}\text{I}$ (fg g <sup>-1</sup> )	$^{127}\text{I}$ (mBq m <sup>-3</sup> )	$^{127}\text{I}$ (ng g <sup>-1</sup> )	$^{129}\text{I}/^{127}\text{I}$ (10 <sup>-8</sup> )
STADE	1	53.370 ; 9.329	0.540	1.1±0.1	7.5±0.3	10.4±0.6	10.8±0.8
MEDEM-Reede	2	53.528 ; 8.431	10.70	27.9±1.1	182.5±7.7	29.4±1.5	93.5±5.9
Elbe 1	3	54.001 ; 8.066	30.97	46.2±1.9	301.9±12.7	44.6±1.9	101.7±6.1
	3A	54.009 ; 7.101	33.11	42.8±1.8	279.7±11.7	35.6±1.1	118.3±6.1
Borkumriffgrund	7	53.600 ; 6.250	33.17	46.1±1.9	301.5±12.7	37.4±1.1	121.4±6.0
	7A	54.002 ; 5.405	34.51	17.8±0.8	116.3±4.9	44.6±1.9	39.3±2.3
VTG Friesland	8	54.000 ; 4.501	34.52	10.0±0.4	65.6±2.8	43.6±1.4	22.7±1.2
Botney Cut	9	54.000 ; 3.001	34.60	12.6±0.5	82.3±3.5	41.3±2.1	30.0±1.9
	9A	53.251 ; 3.299	34.72	14.5±0.6	94.8±4.0	40.0±1.4	35.7±1.9
	9B	53.200 ; 4.391	34.66	36.2±1.5	236.7±9.9	42.4±1.6	84.1±4.6
	10	53.000 ; 4.000	35.14	26.1±1.1	170.4±7.2	39.9±1.4	64.3±3.4
Ijmuiden Appr.	11	52.300 ; 4.199	33.05	47.6±2.0	310.9±13.1	38.9±1.2	120.3±6.1
Maas Approach	12	51.600 ; 3.440	34.23	53.8±2.3	351.4±14.8	36.4±1.2	145.5±7.5
Bligh Bank	13	51.422 ; 2.514	34.86	70.8±3.0	462.8±19.6	44.4±1.4	157.0±7.0
	14	52.000 ; 2.003	35.03	27.5±1.2	179.8±7.6	40.1±1.4	67.5±3.6
Noord Hinder	15	52.289 ; 2.293	34.86	24.3±1.0	158.9±6.7	38.4±1.2	62.4±3.1
	15A	53.000 ; 3.000	34.72	19.1±0.8	124.5±5.3	37.7±1.3	49.8±2.6
	15B	52.572 ; 2.121	34.38	2.3±0.1	14.9±0.6	41.5±1.1	5.4±0.3
Dowsings	16	53.200 ; 1.400	34.52	2.3±0.1	14.8±0.6	43.3±1.1	5.1±0.2
Neptune Gas Field	17	54.000 ; 1.000	34.65	4.0±0.2	25.8±1.1	42.3±1.3	9.2±0.5
	17A	53.600 ; 0.227	34.41	2.5±0.1	16.1±0.7	41.2±1.1	5.9±0.3
Barmada Bank	18	54.599 ; 0.000	34.62	2.4±0.1	15.9±0.8	45.4±1.5	5.3±0.3
	18A	54.599 ; -0.397	34.45	2.6±0.1	16.7±0.7	43.7±1.7	5.8±0.3
Dogger Bank	19	54.599 ; 2.002	34.54	2.1±0.1	13.5±0.6	42.5±2.0	4.8±0.3
Elbow Spit	20	54.598 ; 3.599	34.95	2.0±0.1	13.2±0.6	44.0±1.5	4.5±0.2
Nordschillgrund	21	54.600 ; 5.002	34.68	6.1±0.3	39.6±1.7	42.0±1.7	14.2±0.8
Weisse Bank	22	55.000 ; 6.150	34.46	22.0±0.9	143.8±6.1	45.7±1.4	47.4±2.4
	22A	54.599 ; 7.001	33.93	37.9±1.6	247.9±10.5	41.8±1.2	89.4±4.4
Sylt 2	23	55.000 ; 7.351	32.53	48.1±2.0	314.1±13.2	40.5±1.1	116.9±5.6
Lister Tief	25	55.000 ; 8.150	30.58	54.8±2.3	358.2±15.2	40.6±2.0	133.0±8.4
W of Ringkø	26	56.000 ; 7.480	33.06	42.6±1.8	278.7±11.8	40.8±1.3	102.8±5.2
	26A	56.001 ; 7.018	34.07	29.8±1.3	194.8±8.2	42.1±1.1	69.7±3.3
	27	56.000 ; 6.001	34.93	5.3±0.2	34.6±1.5	45.2±1.4	11.5±0.6
N of Doggerbank	29	56.000 ; 3.000	35.04	1.6±0.1	10.2±0.4	45.8±1.3	3.4±0.2
N of Doggerbank	30	56.001 ; 2.000	35.10	1.5±0.1	9.8±0.4	44.9±1.1	3.3±0.2
N of East Bank	31	56.000 ; 1.000	34.89	2.1±0.1	13.8±0.6	43.0±1.1	4.8±0.2
Swallow Hole	32	56.001 ; 0.013	34.80	2.1±0.1	13.9±0.6	43.65±1.1	4.8±0.2
Berwick Bank	33A	55.600 ; -1.000	34.50	2.4±0.1	15.5±0.7	43.6±1.1	5.4±0.3
	33	55.598 ; -1.385	34.64	2.3±0.1	15.0±0.6	44.0±1.2	5.1±0.3
	34	56.600 ; -1.000	34.89	1.8±0.1	11.60±0.5	43.7±1.9	4.0±0.2
	34A	56.597 ; -1.391	34.83	1.9±0.1	12.6±0.5	42.3±1.7	4.5±0.3
	34	56.597 ; -1.391	34.83	1.9±0.1	12.2±0.5	43.6±1.2	4.2±0.2
	36	57.000 ; 3.300	35.10	1.1±0.1	7.1±0.3	44.3±1.6	2.4±0.1
	37	56.599 ; 5.003	35.05	1.7±0.1	10.9±0.5	46.2±1.9	3.5±0.2
	38	57.000 ; 5.600	35.01	1.8±0.1	11.8±0.5	43.7±1.2	4.1±0.2
	38A	57.001 ; 6.597	34.64	12.6±0.5	82.3±3.4	44.4±1.9	27.9±1.7
Hanstholm	39 (0 m)	57.000 ; 7.599	34.13	31.2±1.3	204.0±8.6	42.2±1.7	72.8±4.2
	39 (15 m)	57.000 ; 7.599	34.13	27.9±1.2	182.7±7.7	41.3±1.4	66.6±3.6
Skagerrak	40 (0 m)	57.480 ; 7.599	30.90	17.1±0.7	112.0±4.7	37.8±1.2	44.7±2.3
	40 (249 m)	57.480 ; 7.599	35.20	0.9±0.1	6.2±0.3	47.2±1.7	2.0±0.1
	40 (509)	57.480 ; 7.599	35.16	1.1±0.1	7.3±0.3	45.9±0.9	2.38±0.11
	40	57.480 ; 7.599	32.00	11.5±0.5	75.5±3.2	41.4±1.0	27.43±1.29
SW-lich Listafjord	41	58.000 ; 5.599	32.26	12.5±0.5	81.6±3.4	39.2±1.4	31.36±1.69
Eigersundbanken	42	58.000 ; 4.599	34.37	5.3±0.2	34.9±1.5	42.6±1.5	12.32±0.65
Midtbanken	43	58.000 ; 2.598	34.97	1.4±0.1	9.4±0.4	44.1±1.7	3.20±0.18
Andrew Oil Filed	44	57.600 ; 1.300	35.21	0.7±0.1	4.4±0.2	43.9±2.0	1.52±0.09
Bosies Bank	45	57.600 ; -0.599	35.12	1.2±0.1	7.9±0.3	44.0±1.3	2.70±0.13
	45A	58.001 ; -2.001	34.99	1.4±0.1	9.4±0.4	43.5±1.1	3.26±0.15
	45	58.303 ; -2.302	34.75	2.5±0.1	16.3±0.7	44.1±1.3	5.58±0.28
	46	58.600 ; -1.299	35.22	0.7±0.1	4.5±0.2	43.6±1.7	1.56±0.09
E-lich Orkneys	47	59.000 ; 0.000	35.13	0.6±0.1	3.9±0.2	40.08±1.0	1.5±0.1
	48	59.000 ; 1.003	35.05	1.4±0.1	8.9±0.4	44.8±1.4	3.0±0.1
Utsiragrunnen	49	58.600 ; 3.000	34.17	4.5±0.2	29.1±1.2	43.9±2.4	10.0±0.7
w-lich Boknafjord	50 (0 m)	59.000 ; 4.295	32.58	16.0±0.7	104.4±4.4	39.5±1.1	39.8±1.9
w-lich Boknafjord	50 (101 m)	59.000 ; 4.295	35.05	2.2±0.1	14.2±0.6	46.1±1.7	4.65±0.3
	50 (254 m)	59.000 ; 4.295	35.20	0.9±0.1	6.05±0.3	45.7±1.2	1.99±0.1
E-lich Bergenbank	51	60.000 ; 4.301	32.35	13.1±0.6	85.5±3.6	38.6±1.2	33.39±1.7
Bergenbanken	52	60.000 ; 1.599	34.79	1.5±0.1	9.85±0.4	43.02±1.1	3.45±0.2
E-lich Shetlands	53	59.600 ; -0.300	35.29	0.8±0.1	5.20±0.2	44.2±1.2	1.8±0.1
östl. Falls Gap	901	51.300 ; 2.000	35.04	49.2±2.1	321.0±13.0	38.27±1.1	126.4±6.2
Str. von Dover	902	51.050 ; 1.301	35.09	26.5±1.1	172.9±7.3	39.5±1.3	66.0±3.4
O-Meridian	903	50.301 ; 0.002	35.07	38.5±1.6	251.0±10.0	37.3±0.9	101.3±4.8
NE Casquettes	904	50.200 ; -2.000	35.22	6.0±0.3	39.3±1.7	43.4±1.1	13.6±0.6
SSW-lich Start Point	905	49.500 ; -3.599	35.23	4.5±0.2	29.2±1.2	43.3±1.2	10.2±0.5
NW-lich Ouessant	906	49.001 ; -5.600	35.21	-	-	44.6±0.9	-
Cap de la Hague	909	49.550 ; -2.000	35.16	19.5±0.8	127.6±5.4	38.5±1.2	49.9±2.5
N-lich Le Havre	910	50.000 ; 0.000	34.85	103.0±4.4	673.9±28.6	36.9±1.4	275.4±15.3
Cap Griz Nez	911	50.499 ; 1.299	34.68	83.3±3.5	544.3±23.0	39.2±1.2	209.1±10.6



**Fig. 2:**  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios versus  $^{127}\text{I}$ - and  $^{129}\text{I}$ -concentrations in the North Sea and in the English.

## 2. Depth dependence of $^{127}\text{I}$ and $^{129}\text{I}$ in the North Sea

The depth profiles analysed add further information on the transport and distribution of the  $^{129}\text{I}$  from the reprocessing plants to the North Sea. Here we compare only 3 same stations (39, 40 and 50) (Tab. 2). At station 39 (Hanstholm, northwestern Denmark) until 30 m depth the concentration of  $^{127}\text{I}$  and activity concentration of  $^{129}\text{I}$  are fairly constant with depth and represent 33 % in the depth. But at the same station Michel et al. 2012 found, particularly for  $^{129}\text{I}$ , more  $^{129}\text{I}$  (80 %) under 15 m as surface water (0 m). At station 40 (Skagerrak) and 50 (Boknafjord) the situation is other. At station 40 the concentration of  $^{127}\text{I}$  (38  $\text{ng g}^{-1}$ ) at surface is relatively low, while at 200 m and 512 m depth it was 46  $\text{ng g}^{-1}$ . The concentration of  $^{129}\text{I}$  at the surface (211  $\text{mBq m}^{-3}$ ) is 30 times higher than at 200 and 512 m depth. By Michel et al. 2012 the activity concentration of  $^{129}\text{I}$  at the surface is only 5 times higher than at 200 and 512 m depth. The same is observed at station 50. Generally  $^{129}\text{I}$  and thus  $^{129}\text{I}/^{127}\text{I}$  ratios are higher at the surface than at depth.

**Tab. 2:**  $^{127}\text{I}$ ,  $^{129}\text{I}$  and  $^{129}\text{I}/^{127}\text{I}$  ratios isotopic in sea water depth from the North Sea during the August 2009 cruise.

Station	Depth [m]	$^{127}\text{I}$ [ $\text{ng g}^{-1}$ ]	$^{129}\text{I}$ [ $\text{mBq m}^{-3}$ ]	$^{129}\text{I}/^{127}\text{I}$ ratio [ $10^{-8}$ ]	% $^{127}\text{I}$	% $^{129}\text{I}$
39	0	42	204	73	34	36
	15	41	183	67	33	32
	32	41	180	67	33	32
40	0	38	211	45	29	94
	200	47	6	2	36	3
	512	46	7	2	35	3
50	0	40	104	40	30	84
	100	46	14	5	35	11
	230	46	6	2	35	5

### 3. $^{127}\text{I}$ and $^{129}\text{I}$ in precipitation and their fall-out in Northern Germany

The mean concentrations of  $^{127}\text{I}$  in the precipitation from Lower Saxony, North Germany are in 2009 between 2.4 and 5.1 ng g<sup>-1</sup>, the mean activity concentration of  $^{129}\text{I}$  and  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios are between 0.4 and 2.3 fg g<sup>-1</sup> and between  $1.1 \times 10^{-7}$  and  $5.7 \times 10^{-7}$ , respectively (Tab. 3). The highest values of  $^{129}\text{I}$  activity concentrations as well the  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios were in precipitation from Norderney, hier we see the direct influence of the North Sea via sea-spray.

The concentration of  $^{127}\text{I}$  as well the activity concentration of  $^{129}\text{I}$  in through-falling precipitation from Buer-Osterwald are the two to five times higher than in open-field precipitation, but are justly two time for the  $^{129}\text{I}/^{127}\text{I}$  ratios. We can explain that with the dry precipitation of iodine isotopes on the leaves of the trees for the though fall Sample. So we haven't observed grand difference between  $^{129}\text{I}/^{127}\text{I}$  ratios from the dry and wet precipitation. Generally the  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios are in the region I justly doppelt than in the other regions, but are the same values in the region II and IV.

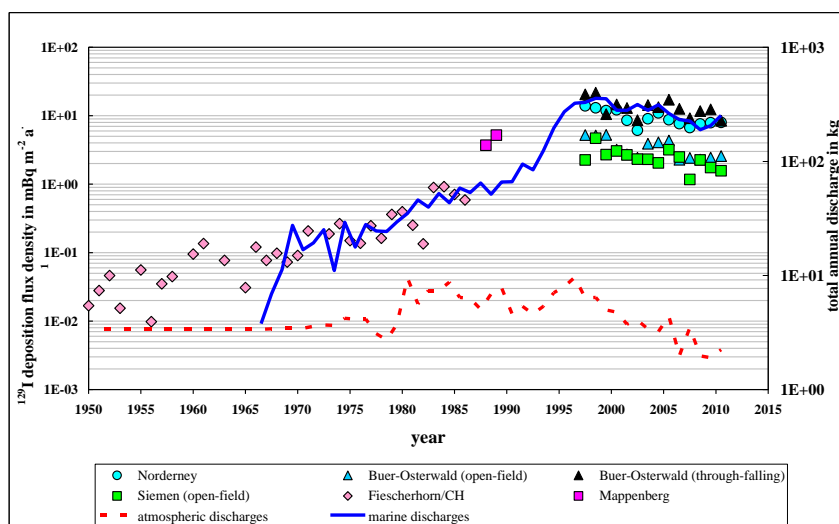
**Tab. 3:**  $^{127}\text{I}$ ,  $^{129}\text{I}$  and  $^{129}\text{I}/^{127}\text{I}$  isotopic ratio in 2009 at different sampling Precipitation in Lower Saxony, Germany

sampling	Precipitation art	$^{127}\text{I}$ [ng g <sup>-1</sup> ]	$^{129}\text{I}$ [fg g <sup>-1</sup> ]	$^{129}\text{I}/^{127}\text{I}$ ratios [10 <sup>-7</sup> ]
Norderney	Open-field	4.2±1.2	2.3±0.6	5.7±1.5
Buer Osterwald	Open-field	5.1±3.0	0.5±0.3	1.1±0.5
Buer Osterwald	through-falling	9.9±4.3	2.4±0.9	2.5±0.8
Siemen	Open-field	2.4±0.8	0.4±0.1	1.7±0.3

Today, the North Sea appears as the dominant source of air-borne iodine in coastal regions of Northern Germany due to the liquid emissions of the European reprocessing plants. Tab. 4 presents the annual precipitation of  $^{127}\text{I}$  and  $^{129}\text{I}$ . We see the different deposition rates of  $^{127}\text{I}$  as well  $^{129}\text{I}$  in open-field and through-falling precipitation and it is in through-fall four times high than in open-field, which are interpreted by us as being differently influenced by wet and wet-plus-dry fallout (Szidat et al. 2000b, Ernst 2003, Michel et al. 2004-2012). During the years 1997 - 2009, the geometric mean annual  $^{129}\text{I}$  deposition rates in regions II and III of Lower Saxony were 3.1 mBq m<sup>-2</sup> a<sup>-1</sup> for open-field precipitation and 12.2 mBq m<sup>-2</sup> a<sup>-1</sup> for through-falling precipitation. At the Norderney in the region I near the North Sea the annual  $^{129}\text{I}$  deposition rates was 10.4 mBq m<sup>-2</sup> a<sup>-1</sup> and this is higher than thus of the other regions, which due to the influence of sea-spray at the coast.

**Tab. 4:** The annual  $^{127}\text{I}$  and  $^{129}\text{I}$  deposition rates during 1997 – 2009 at different sampling in Lower Saxony, Germany.

Sampling precipitation	precipitation in mm a <sup>-1</sup>	annual $^{127}\text{I}$ deposition rate in mg m <sup>-2</sup>	annual $^{129}\text{I}$ deposition rate in mBq m <sup>-2</sup>
Norderney Open-field	742	2.7 x 1.4 <sup>±1</sup>	10.4 x 1.4 <sup>±1</sup>
Buer-Osterwald Open-field	775	1.6 x 1.7 <sup>±1</sup>	3.1 x 2.0 <sup>±1</sup>
Buer-Osterwald through-falling	775	5.4 x 1.5 <sup>±1</sup>	12.8 x 1.5 <sup>±1</sup>
Sessen Open-field	775	1.2 x 1.4 <sup>±1</sup>	3.0 x 1.6 <sup>±1</sup>
Sessen through-falling	775	4.0 x 1.2 <sup>±1</sup>	11.4 x 1.3 <sup>±1</sup>
Siemen Open-field	675	1.4 x 1.5 <sup>±1</sup>	2.2 x 1.7 <sup>±1</sup>



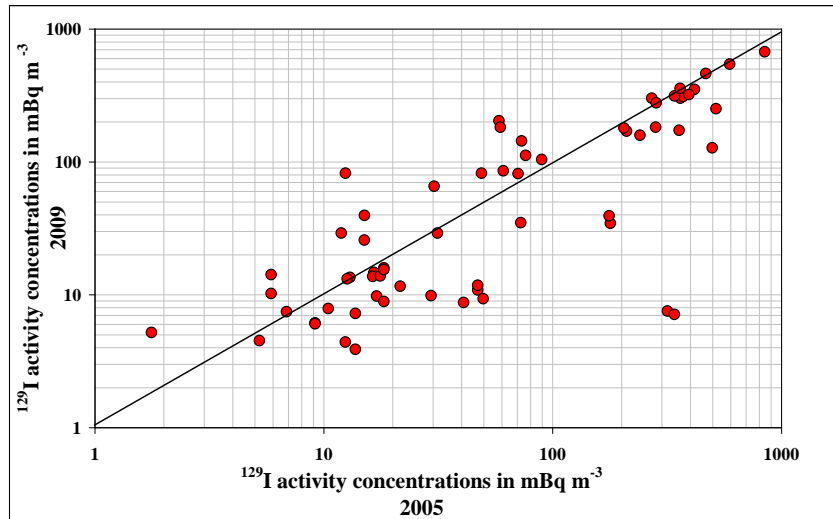
**Fig. 3:** Time series of annual  $^{129}\text{I}$  deposition rates in  $\text{mBq m}^{-2}$  at Norderney (region I), Buer-Osterwald (region III) and Siemen (region IV) in Northern Germany. In addition, the annual sums of the Sellafield and La Hague gaseous and liquid releases in kg are given.

Fig. 3 shows the total liquid and gaseous releases from Sellafield and La Hague and the annual  $^{129}\text{I}$  deposition rates in Northern Germany. After 1996, the total liquid releases from Sellafield and La Hague is nearly constant with slightly decreases. Between 1996 and 2010 the total releases gaseous decreases rapidly than the total liquid releases. On the other side we see the same pattern the annual  $^{129}\text{I}$  deposition rates the precipitation and the total liquid releases from Sellafield and La Hague, and different pattern with the total gaseous releases.

#### 4. Comparison the results of $^{129}\text{I}$ and $^{129}\text{I}/^{127}\text{I}$ isotopic from 2005 and 2009

In this study, we have the possibility to compare the results of iodine in the North Sea of 2009 with those of 2005. The concentrations of  $^{127}\text{I}$  in the North Sea in 2005 are  $(30\text{-}57) \text{ ng g}^{-1}$  with mean of  $40.1 \text{ ng g}^{-1}$  and 13.9 % as relative standard derivation (RSD). In comparison with in 2009, the concentrations of  $^{127}\text{I}$  are between  $(38\text{-}47) \text{ ng g}^{-1}$  with mean of  $43.1 \text{ ng g}^{-1}$  and 4.6 % as RSD. Despite the different concentration of  $^{127}\text{I}$  in 2005 and 2009 shows the mean value of the ocean ( $45 - 60 \text{ ng g}^{-1}$ ).

The first observation in Fig. 4 is seen to be the higher activity of  $^{129}\text{I}$  of 2005 than of 2009 in the near from La Hague and in the English Channel. General shows the compared of  $^{129}\text{I}$ -activity and  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios the same pattern.



**Fig 4:**  $^{129}\text{I}$  activity concentrations in  $\text{mBq m}^{-3}$  in 2005 and 2009 in the North Sea and in the English Channel.

## 5. Conclusion

- The results of the investigation in 2009 reveal high  $^{129}\text{I}$  contaminations in the North Sea and in the English Channel.
- $^{129}\text{I}$  from La Hague is transported through the English Channel and to the North Sea.
- Due to a higher dilution and a more complex current system, the  $^{129}\text{I}$  signal from Sellafield is not so clear-cut in the North Sea.
- The  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios with a maximum of  $2.7 \times 10^{-6}$  are at least four orders of magnitude higher than the natural isotopic ratio of  $1.5 \times 10^{-12}$ .
- The variability of the  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios is exclusively determined by admixture of anthropogenic  $^{129}\text{I}$ .
- Time series of the iodine isotopes in precipitation in Northern Germany demonstrate the decisive role of the marine discharges for the on-going  $^{129}\text{I}$  atmospheric fallout in Western Europe.
- $^{129}\text{I}$  is presently not of radiological significance, but it can be regarded as an indicator of the human long-term impact on the environment:  $^{129}\text{I}$  can be used as tracer.

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