AMS measurements of ^{129}I in seawater around Iceland and the Irminger SeaJ.M. Gómez-Guzmán^{a,*}, M. Villa^b, F. Le Moigne^c, J.M. López-Gutiérrez^{a,d}, M. García-León^{a,e}^a Centro Nacional de Aceleradores (CNA), Seville, Spain^b Dpto. de Física Aplicada II, ETSIE, University of Seville, Spain^c Department of Ocean Biogeochemistry and Ecosystems, National Oceanography Centre, Southampton, UK^d Dpto. De Física Aplicada I, EUP, University of Seville, Spain^e Dpto. De Física Atómica, Molecular y Nuclear, University of Seville, Spain

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ABSTRACT

Large amounts of iodine-129 were, and still are, released into the environment from nuclear facilities, in particular from two reprocessing facilities located on the east coast of the North Atlantic Ocean (Sellafield and La Hague). The main transport path of the releases from the two facilities is through the North Atlantic Current (NAC) and subsequently the Norwegian Coastal Current (NCC) to the Arctic Ocean. Iceland lies on the Scotland–Greenland ridge, which separates the Atlantic and Arctic oceans. ^{129}I data available in that area are scarce despite their importance in modeling ^{129}I dispersion through the North Atlantic and Arctic Oceans. For this reason, we have determined the $^{129}\text{I}/^{127}\text{I}$ ratio in seawater samples by means of Accelerator Mass Spectrometry (AMS) from three locations at the Iceland and Irminger Basins at different depths (from surface to 1000 m) in order to study the transport pathways of the anthropogenic releases to the waters. The measured $^{129}\text{I}/^{127}\text{I}$ ratios, compared to the pre-anthropogenic $^{129}\text{I}/^{127}\text{I}$ value, show the strong influence of the artificial ^{129}I discharges in the North Atlantic waters.

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1. Introduction

Iodine-129 is a long-lived cosmogenic radionuclide for which the natural abundances have been altered in a significant way by human actions. In nature, it is produced by interactions of cosmic rays with Xe in the atmosphere, by spontaneous fission of ^{238}U and ^{235}U in the lithosphere [1] and, to a minor extent, neutron-induced reactions on ^{128}Te and ^{130}Te in the crust [2]. These processes lead to typical isotopic ratios between 10^{-12} [3] and 6×10^{-13} [4]. However, over the last 60 years, certain artificial nuclear activities, such as bomb tests, the Chernobyl accident, and succeeding activities of nuclear fuel reprocessing plants, especially those of Sellafield and La Hague, have dramatically increased its presence in the environmental compartments, thereby raising the environmental background ratios $^{129}\text{I}/^{127}\text{I}$ to values between 10^{-10} and 10^{-8} [5], with isotope ratio peaks of 10^{-7} or higher in zones directly affected by such nuclear facilities [6].

Due to its biophilic behavior, iodine can enter the food chain via the air as a result of aerial emissions. This means that ^{129}I (15.7×10^6 years), its only long-lived isotope, remains in the food chain, compared to other short-lived isotopes (e.g., ^{131}I). Iodine forms compounds (e.g., I_2 , HI, HOI, CH_3I and KI) that are both

water-soluble and volatile and thus can also easily enter the hydrological cycle. Hence, ^{129}I is an ideal tracer of water masses.

Elevated concentrations of ^{129}I in the surface water of the North Atlantic Ocean [7], the Baltic Sea [8], and the Arctic Ocean [9] were related to the release from the nuclear fuel reprocessing facilities at Sellafield (UK) and La Hague (France) into the Irish Sea and English Channel, respectively. The total annual ^{129}I discharge from both facilities remained below 20 kg year^{-1} ($0.1 \text{ TBq year}^{-1}$) until the beginning of the 1990s. The discharge later increased to 300 kg year^{-1} (2 TBq year^{-1}), 75% of which originated from the La Hague facility [10]. The ^{129}I discharges from these facilities account for >95% of the total inventory in the global ocean up to 2000. This anthropogenic source thus dominated all other sources including natural production, nuclear testing, and other nuclear activities and accidents.

Earlier studies of ^{129}I in the North Atlantic Ocean and Arctic Ocean were aimed not only at evaluating sources of radiological hazards but also at tracing ocean circulation [7,11]. These investigations showed that ^{129}I of the reprocessing facilities reached these waters, and accordingly, this anthropogenic signal was used as a tracer for the rate of circulation of ocean currents, and as an inventory in the North Atlantic and Arctic Oceans.

In this study, an analysis of ^{129}I is presented from three sampling profiles in the North Atlantic Ocean from a depth of up to 1000 m in order to update the map of ^{129}I concentrations in the North Atlantic, thereby providing new insights into the use of this

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radioisotope as a tracer of oceanographic currents. The measured profiles were located in the Iceland Basin and Irminger Basin and sampling was carried out from RRS Discovery as part of the High Latitude North Atlantic cruise in summer 2010.

2. Samples and analysis

^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios were investigated on samples taken at three sites in summer 2010 (02nd July–11th August) on board the RRS Discovery (cruise D354) in the Irminger Basin (Station 8), and in the Iceland Basin (Stations 4 and 6) (Fig. 1 and Table 1). Stations were denoted as ST4 (latitude $61^\circ\text{--}47.5'$, longitude $21^\circ\text{--}04.9'$), ST6 (latitude $59^\circ\text{--}59.01'$, longitude $23^\circ\text{--}37.5'$) and ST8 (latitude $60^\circ\text{--}00.2'$, longitude $34^\circ\text{--}59.5'$). Seawater samples were collected from three depth profiles. All water samples were collected in polyethylene bottles, firmly sealed, and stored in a cold dark room.

Samples from different depths were filtered using a $0.45\ \mu\text{m}$ membrane filter, and 100 ml of the filtered water was then used for the extraction of iodine using a carrier-free method [13]. We first added 4 mg of silver powder (Alfa Aesar, Silver Powder Spherical, -635 mesh, 99.9% metal basis) to 100 ml of seawater in a 200 ml plastic bottle. In order to transform the iodine (which is mostly in the form IO_3^-) in seawater into I_2 , we added 2 ml of nitric acid, and quickly closed the bottle to avoid loss of the volatile I_2 . The silver powder was kept in contact with the acidified seawater for 24–48 h, after which it was centrifuged and the seawater removed. The silver powder was washed with deionised water ($18\ \text{M}\Omega\ \text{cm}$) until the pH was neutral and it was then dried in an

oven at 70°C . The silver powder was then loaded into a copper target holder for the measurement of $^{129}\text{I}/^{127}\text{I}$ ratio at CNA [14–16].

AMS measurements were performed relative to a standard with a known $^{129}\text{I}/^{127}\text{I}$ isotopic ratio. The ^{129}I standard used was made by repeated dilutions from NIST SRM 3230 Iodine Isotopic Standard Level 1, with a certified $^{129}\text{I}/^{127}\text{I}$ ratio of $(4.920 \pm 0.062) \times 10^{-10}$ and its nominal ratio was $(4.66 \pm 0.02) \times 10^{-11}$. $^{127}\text{I}^-$ currents from samples prepared in this way were generally 20–80 nA before the stripping process. Transmission efficiencies, including selection of charge state +3 after the stripping process, were 9–10%, leading to high energy $^{127}\text{I}^{3+}$ currents of 6–25 nA. The isotopic ratios obtained directly by AMS measurement of the samples were typically of the order of $^{129}\text{I}/^{127}\text{I} \sim 10^{-8}$ and 10^{-10} . On the other hand, chemical blanks to control contamination in the sample preparation process were prepared exactly in the same way but deionised water was used and a known amount of Woodward iodine ($^{129}\text{I}/^{127}\text{I} \approx 7 \times 10^{-14}$ [28]) was added in order to have currents of the same magnitude as those from real samples. The AMS $^{129}\text{I}/^{127}\text{I}$ ratios for the blanks were around 5×10^{-13} . These ratios proved to be three to five orders of magnitude lower than those in the unknown samples and, thus, were of relatively minor impact. The total error including statistics and background correction was generally less than 10%.

^{127}I determination was made with a high-sensitivity ICP-MS system located at the Servicio de Radioisótopos (University of Seville, Spain), using technical conditions described elsewhere [12]. 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. Various

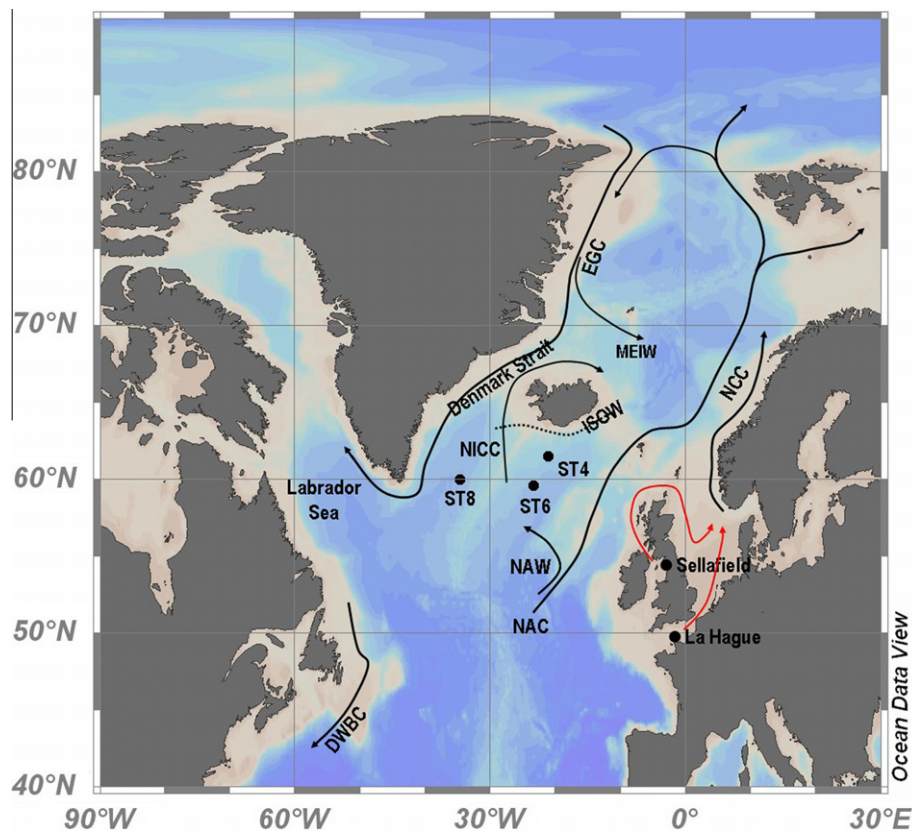


Fig. 1. Map showing location of the profiles used in this study, the pathways of marine discharges from Sellafield and La Hague nuclear reprocessing facilities to the North Sea (red lines) and the major circulation of ocean currents to the Arctic and the North Atlantic Oceans (black line). Solid lines = surface currents, dashed lines = deep currents, LS = Labrador Sea, NCC = Norwegian Coastal Current, EGC = East Greenland Current, ISOW = Iceland–Scotland Overflow Water, DWBC = Deep Western Boundary Current, NAC = North Atlantic Current, NAW = North Atlantic Water, NIIC = North Icelandic Irminger Current, MEIW = Modified East Icelandic Water. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1

Basic data for sampled stations and ^{129}I , $^{129}\text{I}/^{127}\text{I}$ and ^{127}I results. Errors related to ^{127}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios correspond to $\pm 2\sigma$ and $\pm 1\sigma$, respectively. Errors related to ^{129}I concentrations correspond to error propagation theory.

Station	Latitude longitude	Depth (m)	Temperature ($^{\circ}\text{C}$)	Salinity	$^{129}\text{I}/^{127}\text{I}$ ($\times 10^{-10}$) measured	^{127}I (ppb) measured	^{129}I ($\times 10^8$ atom l^{-1}) calculated
ST4	61.47° N 21.05° W	10	12.08	35.12	161.7 ± 6.7	44 ± 3	33.7 ± 2.7
		20	11.31	35.14	112.4 ± 6.0	46 ± 4	24.5 ± 2.5
		30	9.71	35.25	93.3 ± 4.2	46 ± 3	20.3 ± 1.6
		40	9.24	35.26		44 ± 1	
		60	9.13	35.28	80.1 ± 3.8	36 ± 2	13.7 ± 1.0
		100	8.92	35.26	51.4 ± 2.4	47 ± 2	11.5 ± 0.7
		150	8.82	35.28	173.6 ± 15.7	30 ± 4	24.7 ± 4.0
		400	8.49	35.31	64.7 ± 2.7	37 ± 1	11.4 ± 0.6
		600	8.35	35.30	19.4 ± 1.0	40 ± 2	3.7 ± 0.3
		800	7.34	35.20	12.3 ± 0.8	36 ± 6	2.1 ± 0.4
		1000	5.57	35.07	6.1 ± 0.4	41 ± 1	1.2 ± 0.1
		ST6	59.59° N 23.37° W	30	11.06	35.04	66.1 ± 0.4
150	8.57			35.18	42.7 ± 4.8	44 ± 1	8.9 ± 1.0
300	8.21			35.23	25.8 ± 3.0	42 ± 5	5.1 ± 0.9
600	7.09			35.16	19.1 ± 2.0	41 ± 1	3.7 ± 0.4
ST8	60.00° N 34.59° W	20	9.85	34.97	41.3 ± 2.7	33 ± 2	6.5 ± 0.6
		100	6.14	35.07	26.2 ± 2.7	53 ± 3	6.6 ± 0.8
		300	5.50	35.02	34.4 ± 2.9	47 ± 1	7.7 ± 0.7
		600	4.80	34.97	18.4 ± 1.7	52 ± 1	4.5 ± 0.4

data quality tests were used during every instrumental run, such as replicates, matrix matching, memory effects and laboratory reactive blank checking. Five samples were made up as standards for calibration (including a calibration blank) in the ranges 0.1–10 $\mu\text{g l}^{-1}$ of iodine. These standards were made by successive dilutions with deionised water (18 M Ω cm) of an aqueous stock solution at 1000 $\mu\text{g l}^{-1}$ prepared from suprapure potassium iodide (Merck, Suprapure Kaliumiodid Potassium Iodide, 99.995%). All samples were diluted by a factor of 10 with deionised water and the average ^{127}I concentration was 4 $\mu\text{g l}^{-1}$. Chemical blank consisting of deionised water gave a ^{127}I concentration of (0.15 \pm 0.02) $\mu\text{g l}^{-1}$.

3. Results and discussion

Oceanographically, the sampled region is a very mixed region with influences from the subtropical thermocline via the North Atlantic current, and from the Arctic via cold waters moving southwards to the east of Iceland, following the coast of Greenland. Values of temperature and salinity shown in depth profiles (Table 1) range from 4 to 14 $^{\circ}\text{C}$ and from 34.8 to 35.4 respectively for all samples, and are consistent with reported values (between 4 and 12 $^{\circ}\text{C}$ for temperature and between 34.5 and 35.5 for salinity) for samples of a 1200 m-depth profile collected around Iceland in summer 2002 [17]. Temperatures higher than 4.5 $^{\circ}\text{C}$ in all cases indicate that all sampling points are mainly affected by the branch of the North Atlantic Water (NAW) surface current, which turns westward, forming the anticlockwise subpolar gyre and flows around the southwest shelf of Iceland [18,19]. This current flows north around the north of Iceland, and is named as the North Icelandic Irminger Current (NIIC). This current collides with the Modified East Icelandic Water (MEIW), an eastward branch of the East Greenland Current (EGC) [17,20]. The EGC carries a large amount of low-density and cold surface Arctic waters and denser and warmer intermediate Atlantic waters southward along the edge of the Greenland continental shelf [17,21,30]. The influence of the EGC can be seen especially in Station 8, where temperature and salinity drop slightly. According to salinity and temperature values reported previously [21], the T–S values show that the sampling points are not clearly influenced by the Iceland Scotland Overflow Water (ISOW), which is composed of cold dense waters formed in the polar seas north of Iceland, and which flows southwards

through the narrow channel skirting east and south of the Faroe Islands.

The elevated concentrations of ^{129}I in many parts of the North Atlantic Ocean, particularly in the surface layers [7,22] has been linked to several anthropogenic sources including nuclear weapons testing, nuclear accidents, dumping of nuclear waste and discharges from nuclear reprocessing facilities (Sellafield and La Hague). However, the Sellafield and La Hague discharges seem to be dominant compared with the other sources since the ^{129}I discharges from these facilities account for >95% of the total inventory in the global ocean up to 2000 [9].

The influence of the two currents that cover Iceland and Irminger Basins (Arctic waters moving southwards and North Atlantic waters moving northwards) could be the cause of two different effects observed for ^{129}I concentrations: (i) the highest concentrations of ^{129}I are found in surface water samples, and (ii) lower ^{129}I concentrations, but higher than those related to natural production are found in the deepest samples [23]. The inputs that explain the described depth distribution are analyzed below.

3.1. ^{129}I profiles

The ^{129}I concentrations (Table 1 and Fig. 2) display similar patterns in the three studied depth profiles, although certain differences can be found at Station 8, thereby confirming the contribution of EGC in the Irminger Basin. The highest ^{129}I concentrations occur in station ST4 surface samples, values decrease at ST6 and are reduced to half this value at station ST8, where surface values remain fairly constant across the whole depth profile. Measured ^{129}I concentrations range from (1–30) $\times 10^8$ atom l^{-1} in station ST4 and are higher than those found in the Nansen Basin (in the Arctic Ocean) in July 2001 (0.8–12) $\times 10^8$ atom l^{-1} [7,9].

The high ^{129}I concentrations found in Nansen are due to ^{129}I present in the La Hague and Sellafield releases that enter into the Arctic Ocean through the Norwegian Atlantic Current (NWAC) and the North Coastal Current (NCC) [9]. Formation of denser waters in the polar seas north of Iceland and their flow southward would bring ^{129}I back to the North Atlantic [22]. This probably explains the values three orders of magnitude higher than the estimate for the pre-anthropogenic concentration (3 $\times 10^5$ atom l^{-1}) [24] in the intermediate waters collected at ST4, ST6 and ST8. Another potential explanation could be that organic matter, surviving dilution, reaches the deepest sampling zones and releases ^{129}I into

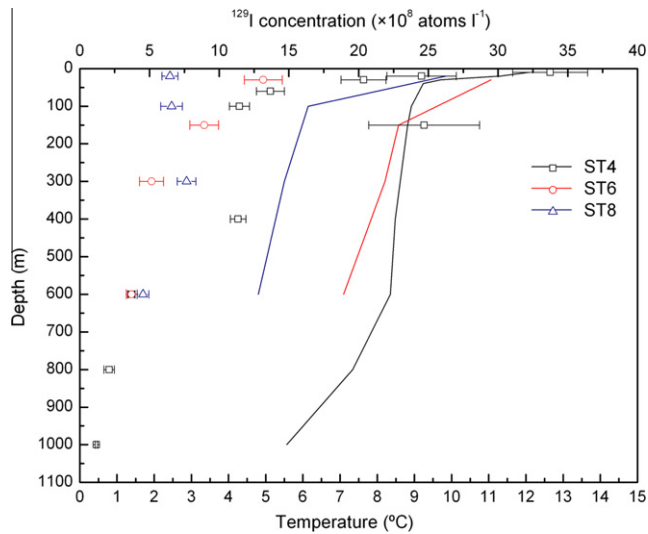


Fig. 2. ^{129}I concentration in the studied depth profiles (squares, circles and triangles). Temperature data have also been included (lines).

the overlying deep water upon anaerobic decomposition in the sediments [25,27].

However, dense waters cannot be responsible for the high ^{129}I concentrations found in surface waters in ST4, ST6 and ST8. Furthermore, those ^{129}I concentrations are not the same order of magnitude as observed in polar waters [7,9]. As commented before, high temperature values found in surface waters indicate that those waters belong to the NAW branch. This is the most probable contribution to ^{129}I concentrations, mostly for Stations 4 and 6, but also 8. According to the literature, the main paths for Sellafield releases are the NWAC and the NCC currents which transport the contamination from the North Sea to the Arctic [31,32]; however, the data presented here suggest that part of the contamination is also transported westbound to the Icelandic waters by means of the westward branch of the NAW. Indeed, it is possible that outbreaks from the boundary currents into the interior might carry the iodine from the Sellafield releases to the South of Iceland and subsequently to the Irminger Basin.

The decrease of the ^{129}I concentration at ST8 located in Irminger Basin confirms this hypothesis, since a stronger contribution of polar waters to the area would dilute the ^{129}I concentrations. Alifimov et al. (2003) [33] show that in the Denmark Strait, ^{129}I could be distinctively measured at $12.5\text{--}0.5 \times 10^8 \text{ atom l}^{-1}$ in 2003. The very low values (<0.5) found from 25.5° W to 26.5° W prove that most of the southward contribution of ^{129}I is due to the EGC flows which are close to the coast of Greenland and to the Labrador Strait. The EGC current may affect the ^{129}I concentrations at Station 8, where slightly lower temperature and salinity values confirm the contribution of polar waters; however, its contribution to waters at Stations 4 and 6 is doubtful, since the westward NAW is predominant in the Iceland Basin, as evidenced by the high temperature values measured [17,21].

The profiles in Fig. 2 support this interpretation, displaying a relationship between temperature and ^{129}I concentration, i.e. NWA- ^{129}I concentration. Higher temperature values and ^{129}I concentrations correspond to Station 4, and lower temperatures and lower ^{129}I concentrations correspond to Station 8, less affected by the NAW warm current.

Finally, ST4 presents a clear increase in ^{129}I concentrations in relation to year 1999, where concentrations measured nearby (latitude 59.5° , longitude 21°) were $(0.30 \pm 16) \times 10^8 \text{ atom l}^{-1}$ [8]. This probably reflects the increase of liquid discharges from European reprocessing fuel facilities in recent years ([29] and references

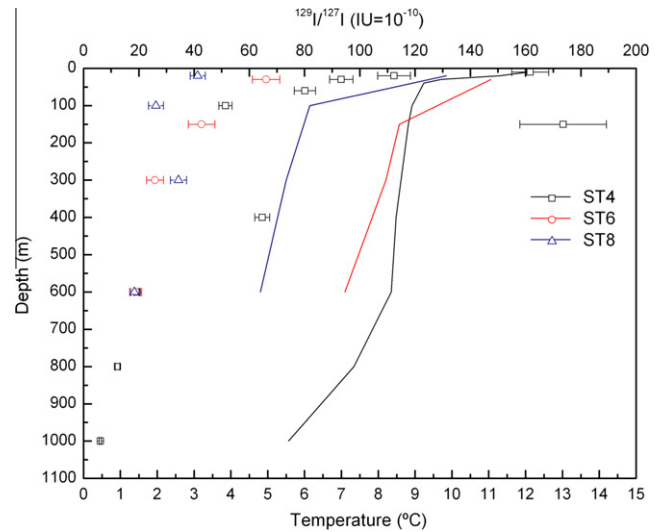


Fig. 3. $^{129}\text{I}/^{127}\text{I}$ ratio in the studied depth profiles (squares, circles and triangles). Temperature data have also been included (lines).

therein). However, the precise path that the ^{129}I follows from the NWA branch to the South of Iceland is uncertain and this path is relevant in the determination of the ^{129}I transient time from Sellafield to Iceland after the increase in releases. ^{129}I from the polar mixed layer and Atlantic layer of the Arctic Ocean which is ventilated by the EGC into the Nordic Seas and North Atlantic Ocean constitute the main pathways of ^{129}I back into the North Atlantic Ocean. The direct contribution of the NAW westward branch has never been considered. For these two reasons more studies are recommended in order to obtain transient times and to reinforce this hypothesis. Moreover, further sampling should be conducted in order to obtain additional data in the area for the construction of improved models that describe the dispersion of ^{129}I throughout the North Atlantic and Arctic Oceans.

3.2. $^{129}\text{I}/^{127}\text{I}$ profiles

The $^{129}\text{I}/^{127}\text{I}$ atomic ratios in the three depth profiles studied (Table 1 and Fig. 3) indicate patterns similar to those of ^{129}I concentrations. This would be expected if all of these ratios had the same origin. Measured atomic $^{129}\text{I}/^{127}\text{I}$ ratios range from $(6.1 \pm 0.4) \text{ IU}$ ($1 \text{ IU} = ^{129}\text{I}/^{127}\text{I} = 10^{-10}$) at a depth of 1000 m, to $(173 \pm 16) \text{ IU}$ at 150 m (both at station ST4). These ratios are higher than both the pre- and post-nuclear marine background (0.01 and 1 IU, respectively) [4,26], are one order of magnitude higher than those measured in Denmark Strait in March 1997, and are considerably larger than those seen in the waters of the Greenland Sea. This supports the aforementioned idea that ^{129}I from Sellafield is directly transported around Iceland and the Irminger Sea from the westward branch of the North Atlantic Waters.

4. Conclusion

The results of this study represent a data set covering Iceland and Irminger Basin locations and indicate that the accumulation of anthropogenic ^{129}I in North Atlantic waters is increasing as a result of the increment of the releases from nuclear reprocessing power plants. Samples collected from the sea surface NAW current showed ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ atomic ratios higher than those previously encountered in the Arctic (Nansen Basin) or at the same location.

Similar patterns have been observed in ^{129}I profiles at three stations, with the highest values in the surface layers, and then

decreasing with depth albeit always above the pre- and post-nuclear marine background. Differences can be found in the Irminger Basin station, which presents lower ^{129}I concentrations, probably due to the contribution of EGC waters. The relatively high surface concentrations in superficial warm dense waters confirm that these waters are influenced by surface NAW which transports ^{129}I from the reprocessing plants. ^{129}I concentrations in intermediate samples are lower, but nevertheless remain at orders of magnitude higher than the estimate for the pre-anthropogenic concentration, which would support the idea of polar waters carrying ^{129}I . Similar conclusions can be obtained using $^{129}\text{I}/^{127}\text{I}$ profiles.

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