

Anthropogenic ^{129}I in seawaters along the north-central part of the English Channel: Levels and tracer applications

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Abstract

The English Channel (the Channel) represents a major sink and transport pathway of anthropogenic radioactive ^{129}I . Despite this important role, data concerning the distribution of ^{129}I in seawater of the Channel are scarce, and most of existing data are restricted to the eastern part of the Channel. The advection and dispersion of ^{129}I from the French coast toward the central and further the English coast, especially in the Channel west of Cap de La Hague, are not fully investigated. We present results of iodine isotopes (^{127}I and ^{129}I) analyses of surface water samples collected along the central English Channel in October, 2010. The data show high ^{129}I concentrations between Dover Strait and La Hague, followed by a dramatic drop towards the Celtic Sea and reveal the dispersal of ^{129}I towards central and northern part of the Channel. Our observation also implies that the entire British coast is contaminated by ^{129}I . ^{129}I levels in the westernmost English Channel, close to the English coast, may reflect combined influences from La Hague and Sellafield. Evolution of ^{129}I between 2005 and 2010 suggests a strong link to temporal marine discharges from La Hague plant. The discharges from the nuclear reprocessing facility have continued since 2010 and thus an ecological evaluation of ^{129}I radioactive hazards in the environment of the Channel may be needed.

Key words: ^{129}I , seawater, iodine isotopes, English Channel

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1 Introduction

The major anthropogenic sources of ^{129}I (half-life=15.7×10⁶ a) are the two world's largest nuclear reprocessing facilities (NRFs) at Sellafield (UK) and La Hague (France). These facilities have collectively discharged over 7 000 kg of ^{129}I to the marine environment and the total discharges peaked in the period 1996–2000 and then slightly reduced thereafter (Fig. 1). Although nuclear reprocessing operations at Sellafield were scheduled to be completed in 2020, its impact on eco-environment will persist for a prolonged period. Presently, ^{129}I concentration higher than the natural level of ~10⁵–10⁶ atoms/L has been observed in the marine environment globally, even in regions far away from the

sources such as the Southern Hemisphere and Antarctic (Snyder et al., 2010; Xing et al., 2017). As one of the world's most ^{129}I contaminated area, the English Channel (the Channel) plays a unique role in the world's ^{129}I marine transport and budget.

The Channel is reputed for its strong tidal currents and residual currents that are associated with meteorological forces. This region is a transition zone between oceanic and neritic waters, where it receives large amounts of radionuclide discharges from the La Hague facility. The long-term fate and environmental impact of these radioactive pollutants required attention, and efforts have been made to investigate the distribution and transport of radionuclides (such as ¹³⁷Cs, ¹²⁵Sb, ⁶⁰Co, ⁹⁹Tc, etc.) in the

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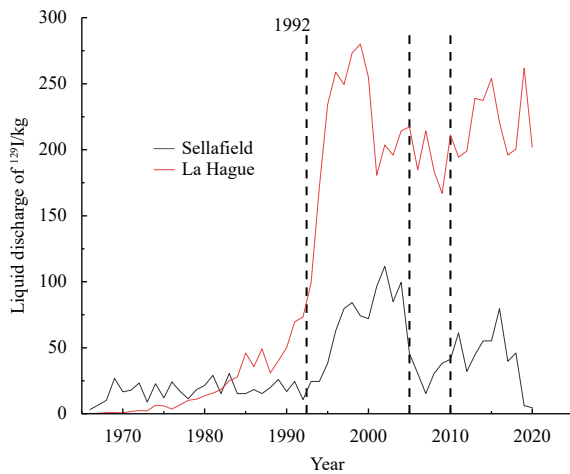


Fig. 1. Annual liquid ^{129}I discharges (1966–2020) from the La Hague and Sellafeld nuclear reprocessing facilities (compiled from He et al. (2014), Sellafeld Ltd. (2021), and Orano Recylage (2020)).

Channel (Dahlgard, 1995; Bailly du Bois and Guéguénat, 1999; Villa et al., 2015). The investigations of ^{129}I have shown that major La Hague-bearing ^{129}I transport pathways extend along the eastern English Channel (continental coast), similar to other conservative radionuclides (e.g., ^{125}Sb and Tritium). In the North Sea, this water mass encounters Sellafeld-labelled ^{129}I carried by the west branch of the North Atlantic Current (Fig. 2). The combined ^{129}I plume predominantly merges into the Norwegian

Coastal Current and mostly ends up in the Arctic Ocean, with some return currents back to the Nordic seas and the Labrador Sea (Alfimov et al., 2004; Smith et al., 2005; Castrillejo et al., 2018).

Although the Channel represents a major reservoir of ^{129}I , the spatial and temporal coverage of concentration data, their distribution pattern and transport mechanisms within the Channel are limited. Previous investigations focused on ^{129}I in algae samples along the French coast. For example, seaweeds and other biological samples were used to reconstruct historical La Hague inputs and to monitor the dispersal of ^{129}I in the Channel (Raisbeck et al., 1995; Fréchou et al., 2002; Fiévet et al., 2020). As for seawater, a few studies reported elevated $^{129}\text{I}/^{127}\text{I}$ concentration ratios of up to 6 orders of magnitude above natural seawater values in the Channel at scattered locations (Hou et al., 2007; Daraoui et al., 2016). ^{129}I data that cover the entire English Channel are useful for calibration and validation of hydrodynamic dispersion models, and to improve our understanding of iodine biogeochemical cycling and environmental significance. However, ^{129}I data in the western part, as well as the southern British coast are rare, which hinders accurate assessment of ^{129}I dispersion and the current status of ^{129}I contamination in the north-western coastal areas of the Channel.

In this investigation, iodine isotopes (^{129}I and ^{127}I) were analyzed in surface water samples covering the western and north-central English Channel. The aim of this work is to determine the spatiotemporal variability of iodine isotopes in channel surface waters. The status of ^{129}I in the western and northern parts of the Channel is of particular concern for the current study. Our results will help to assess the radiological impact of ^{129}I on the marine ecosystem and identify possible hazards to local residents.

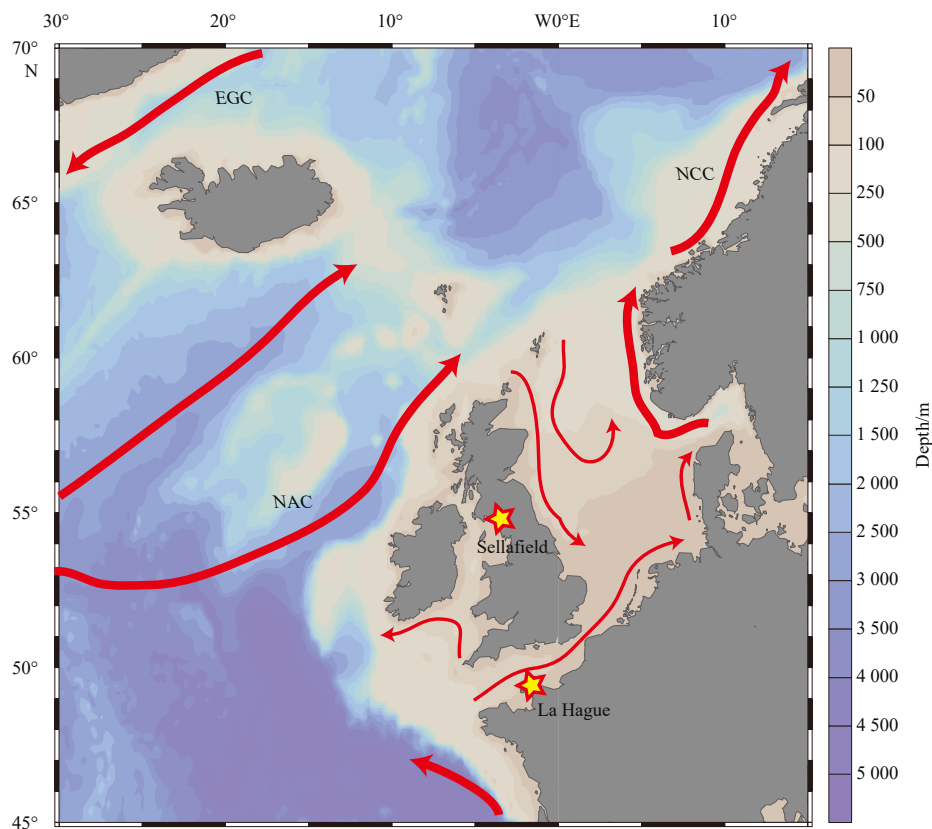


Fig. 2. Map showing the main surface currents in the North Atlantic Ocean and the Nordic seas. EGC, NCC and NAC refer to the East Greenland Current, the Norwegian Coastal Current and the North Atlantic Current, respectively. The stars show the locations of La Hague and Sellafeld.

2 Materials and methods

Surface seawater samples were collected in the Channel (Fig. 3) in October 29–31, 2010 as part of the 2010/2011 Antarctica two-ship expedition jointly funded by the Swedish Polar Research Secretariat and the US National Science Foundation. All water samples were pumped through Teflon pipes from a submerged inlet below the ship and were immediately filtered on-board through a 0.45 μm membrane filter (Sartorius AG, Germany), and stored in clean polyethylene containers under cold and dark conditions until analysis. Loss of ^{129}I during storage has been shown to be insignificant (Hou et al., 2001). A standard CTD sampler was used during sampling to measure water temperature and salinity along the transect. Meteorological parameters, such as wind velocity and humidity, were also measured at the same time.

For ^{129}I analysis, a well-established ^{129}I standard (NIST-SRM-4949c), carrier-free ^{125}I (Amersham Pharmacia Biotech), ^{127}I carrier (Woodward iodine; MICAL Specialty Chemicals) along with analytical grade chemical reagents and deionized water (18.2 M Ω -cm) were used in experiment. An amount of 1.0 mL ^{127}I carrier (Woodward iodine, 2 mg/mL) and 0.1 mL ^{125}I tracer (250 Bq) were added to filtered seawater before iodine extraction. The extraction of iodine from the marine waters adopted the procedure described in Hou et al. (2001). The procedure in summary includes reduction of iodine to iodide with $\text{Na}_2\text{S}_2\text{O}_5$ followed by separation of iodine (as I_2) dissolved in CHCl_3 , and iodine was further extracted into water phase (as I^-) and precipitated as AgI using AgNO_3 . The extract was then dried. The chemical yield of iodine according to the ^{125}I tracer during the separation procedure was 77%–99%. The dried AgI precipitate was mixed with niobium powder and the mixture was packed into a copper cathode for accelerator mass spectrometry (AMS) analysis at a terminal voltage of 3.5 MV, with a relative standard error of less than 3% at the Tandem Laboratory, Uppsala University. Blanks were prepared using the same procedure as the samples. The back-

ground of the AMS system for $^{129}\text{I}/^{127}\text{I}$ concentration ratio was 4×10^{-14} with blanks values below 10^{-13} and samples values above 10^{-11} . Measurement of ^{127}I was performed using X-Series II inductively coupled plasma mass spectrometry (ICP-MS). The detection limit for ^{127}I , calculated as 3 standard deviations of blanks, was 0.02 ng/mL. The overall analytical uncertainties were <7% for ^{127}I and <10% for ^{129}I .

Intensity and direction of depth-mean currents on the day of sampling (October 29, 2010; daily-mean) in Dover Strait areas were generated using MathworksTM MATLAB (R, 2020b). The metadata were provided by E.U. Copernicus Marine Service Information, with a spatial resolution of $0.111^\circ \times 0.067^\circ$, which is available at <https://resources.marine.copernicus.eu/products> (Accessed: September 1, 2021). Ocean bathymetry, coastlines and land topography in the studied area were generated using Ocean Data View (version 5.5.0).

3 Results and discussion

3.1 Levels of ^{129}I in the north-central English Channel

Concentrations of iodine isotopes (^{127}I and ^{129}I) and $^{129}\text{I}/^{127}\text{I}$ concentration ratios in late 2010 are presented in Fig. 4. The results show a wide variation of ^{129}I concentrations in the sampled region, ranging from 8×10^9 atoms/L to 467×10^9 atoms/L. These values are 4–6 orders of magnitude higher than the estimated ^{129}I value prior to the nuclear era (since the 1940s), and at least two orders of magnitude higher than the value due to aboveground nuclear weapons tests. Even the lowest value observed here is approximately a factor of 10 higher than those reported in other ocean waters (He et al., 2016). Previous studies reported ^{129}I levels of 10^7 – 10^{11} atoms/L in marginal seas (e.g., North Sea, Baltic Sea and Labrador Sea) and other sites in the Northern Hemisphere (Smith et al., 2005; Cooper et al., 2001; Schnabel et al., 2007). These water masses were suggested to be influenced by NRF discharges transported by sea currents. To our best know-

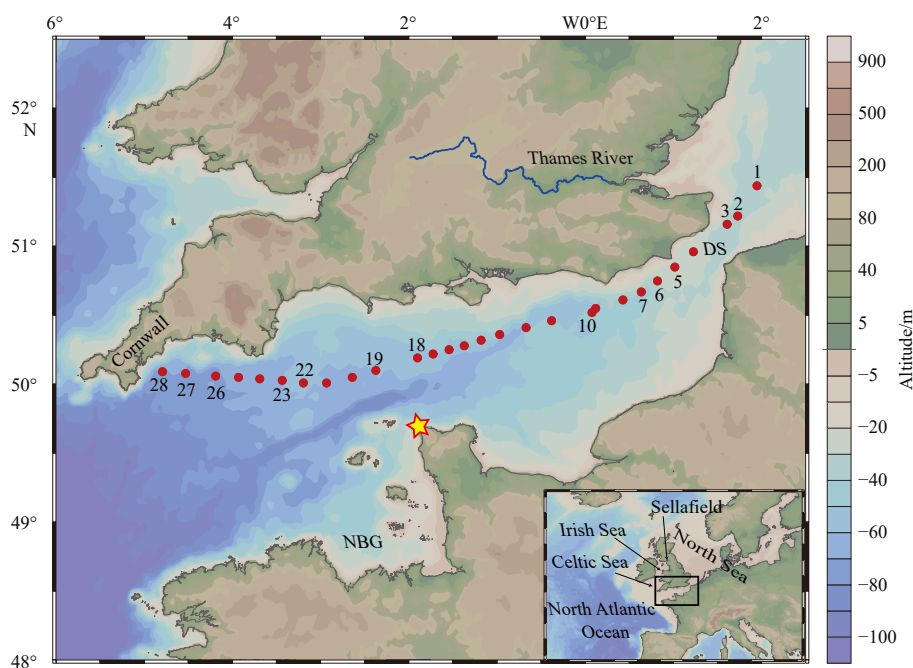


Fig. 3. Location of the water sampling sites collected in this investigation in 2010. Relative locations of the investigated area to the North Atlantic Ocean, Irish Sea, Celtic Sea and the Sellafeld site are indicated in the insert. DS refers to the Dover Strait; NBG, the Normand-Breton Gulf. The star refers to the La Hague site.

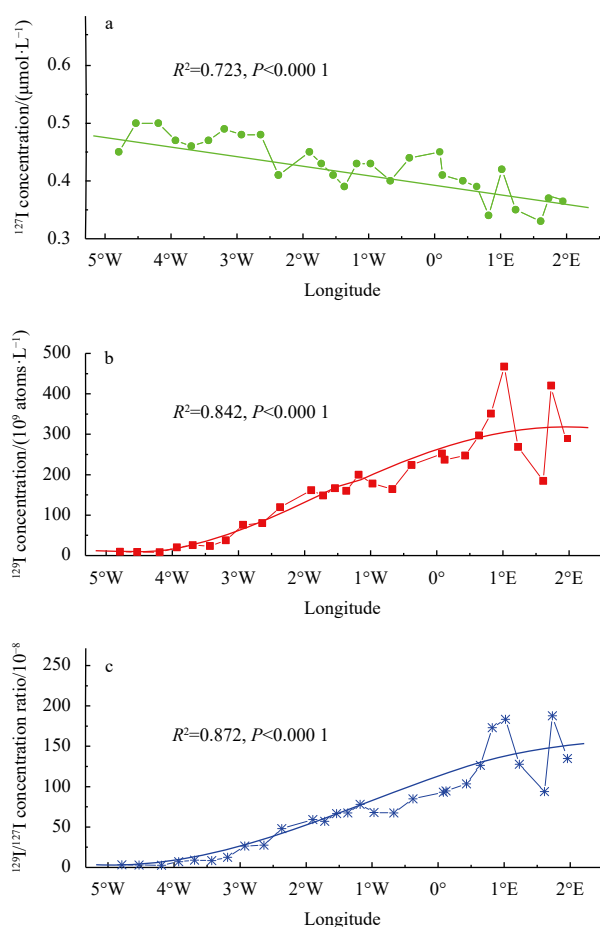


Fig. 4. Longitudinal distribution and correlation coefficient of ^{127}I concentration (a), ^{129}I concentration (b) and $^{129}\text{I}/^{127}\text{I}$ concentration ratio (c) in the English Channel surface water.

ledge, the highest ^{129}I concentration ever reported in the seawater was $1\,280 \times 10^9$ atoms/L, which was measured in a sample close to the Sellafield NRF (Atarashi-Andoh et al., 2007). The highest ^{129}I concentration (467×10^9 atoms/L) found in the samples analyzed here is Site 5 (Fig. 3), and ^{129}I concentration and $^{129}\text{I}/^{127}\text{I}$ concentration ratio show decreasing trends moving southwestward, away from the North Sea. There is also a sharp south-north gradient between the French and UK coasts, east of the Cotentin Peninsula.

The concentration of ^{127}I in the analyzed samples of the Channel ranges from $0.33 \mu\text{mol/L}$ to $0.50 \mu\text{mol/L}$, with an average of $0.43 \mu\text{mol/L}$ (Fig. 4a). The average value is comparable to iodine concentration in seawater (approximately $0.45 \mu\text{mol/L}$). Longitude- and salinity-dependent variations of ^{127}I were observed along the transect (Figs 4a and 5a). These features reflect dilution of iodine-rich Atlantic water by continental fresh water input in the Channel. High ^{127}I concentrations ($>0.45 \mu\text{mol/L}$) occur in the western part of the Channel, followed by a steady decrease to around $0.35 \mu\text{mol/L}$ in the northeast. The iodine isotopic ratios follow an increasing trend comparable to that of the ^{129}I trend (Figs 4b, c). $^{129}\text{I}/^{127}\text{I}$ concentration ratio variations are even larger than ^{129}I concentration, as the ratio increases from west to east by nearly two orders of magnitude with an average of 70×10^{-8} . Sample of Site 18 (Fig. 3) was collected at the station which is most close to the La Hague plant. However, this station is located near the La Hague plume fringe, thus did not exhibit highest

^{129}I level. The highest isotopic ratios are observed close to the Dover Strait, and are comparable with earlier studies in the vicinity (i.e., the southern Bight) (Hou et al., 2007). However, our cruise has not captured the highest ^{129}I level in the Channel because most of the La Hague discharge flows along the near-shore continental region. Previous study suggests that soluble radionuclide concentrations close to the French coast could be 10 times higher than in the central part of the Channel (Bailly du Bois and Dumas, 2005). Accordingly, the lowest $^{129}\text{I}/^{127}\text{I}$ concentration ratios are observed in the most western part of the Channel, which may be attributed to a lesser impact from La Hague discharges. However, the lowest isotopic ratio measured here is still four orders of magnitude higher than the pre-nuclear “natural ratio” of $\sim 1.5 \times 10^{-12}$ (Snyder et al., 2010).

3.2 Environmental conditions revealed by ^{129}I

Our data reveal about 30% decrease of ^{127}I from west to east in the Channel. Off the southwestern coast of England, relatively cold and saline surface water reflects the Atlantic water front that enters the Channel as illustrated by Sites 27 and 28. Apart from these samples, ^{127}I concentrations change linearly with salinity, and show eastward dilution ($R^2=0.69$, $P<0.0001$; Fig. 5a), while ^{129}I dilutes in the opposite direction. The process of ^{129}I fixation onto living organisms has relatively little influence compared to the huge amount of ^{129}I discharges in the Channel. Therefore, large-scale transport of ^{129}I in the Channel is mainly attributed to complicate mixing of water currents. Earlier investigations reported an overall non-conservative behaviour of iodine in brackish water and offshore regions (Truesdale et al., 2001; Truesdale and Upstill-Goddard, 2003). Compared to the open sea, the relatively fast conversion between iodide and iodate and migration of iodine into seaweed are mainly found in estuarine and littoral zones. Therefore, ^{129}I should behave more conservatively in the oxygenated waters of the central English Channel. This conservative behaviour was further proved by its dispersion pattern across the Normand-Breton Gulf (NBG, Fiévet et al., 2020). Hence, as an oceanographic tracer, accurate estimate of bio-mediated ^{129}I consumption effects do not appear to be problematic in the open water sites studied here.

Despite the distinct behavior of ^{127}I and ^{129}I in the Channel water, the plot of ^{129}I concentrations versus the $^{129}\text{I}/^{127}\text{I}$ concentration ratios (Fig. 5b) indicates a highly positive correlation ($R^2=0.996$, $P<0.0001$). The longitudinal distribution pattern of $^{129}\text{I}/^{127}\text{I}$ concentration ratio in the eastern English Channel also coincides with other radionuclides (Bailly du Bois and Dumaset al., 2005). This feature may demonstrate that the $^{129}\text{I}/^{127}\text{I}$ concentration ratio variations have little to do with ^{127}I concentration changes and are primarily determined by ^{129}I concentration. Our data show a strong longitudinal dependence for both ^{129}I and ^{127}I ($R^2 = 0.84$ and $R^2 = 0.87$, respectively), and $^{129}\text{I}/^{127}\text{I}$ concentration ratio and ^{129}I concentration exhibit similar trends; with increases from west to east, as ^{127}I decreases (Fig. 4). This pattern coincides with the main direction of water mass movement in the Channel.

However, water samples collected near the Dover Strait show both ^{129}I concentration and $^{129}\text{I}/^{127}\text{I}$ concentration ratio fluctuations that deviate from the trend line (Figs 4b, c). The ^{129}I concentration variations in this region also agree with the behavior of other radionuclides (e.g., ^{137}Cs , ^{125}Sb and ^{99}Tc) (Herrmann et al., 1995). Possible explanations are: (1) high ^{129}I plume moves along the French coast encounters less-contaminated water that flows along the British coast in Dover Strait region, and (2) strong winds (on average of 13.6 m/s during this campaign) from south

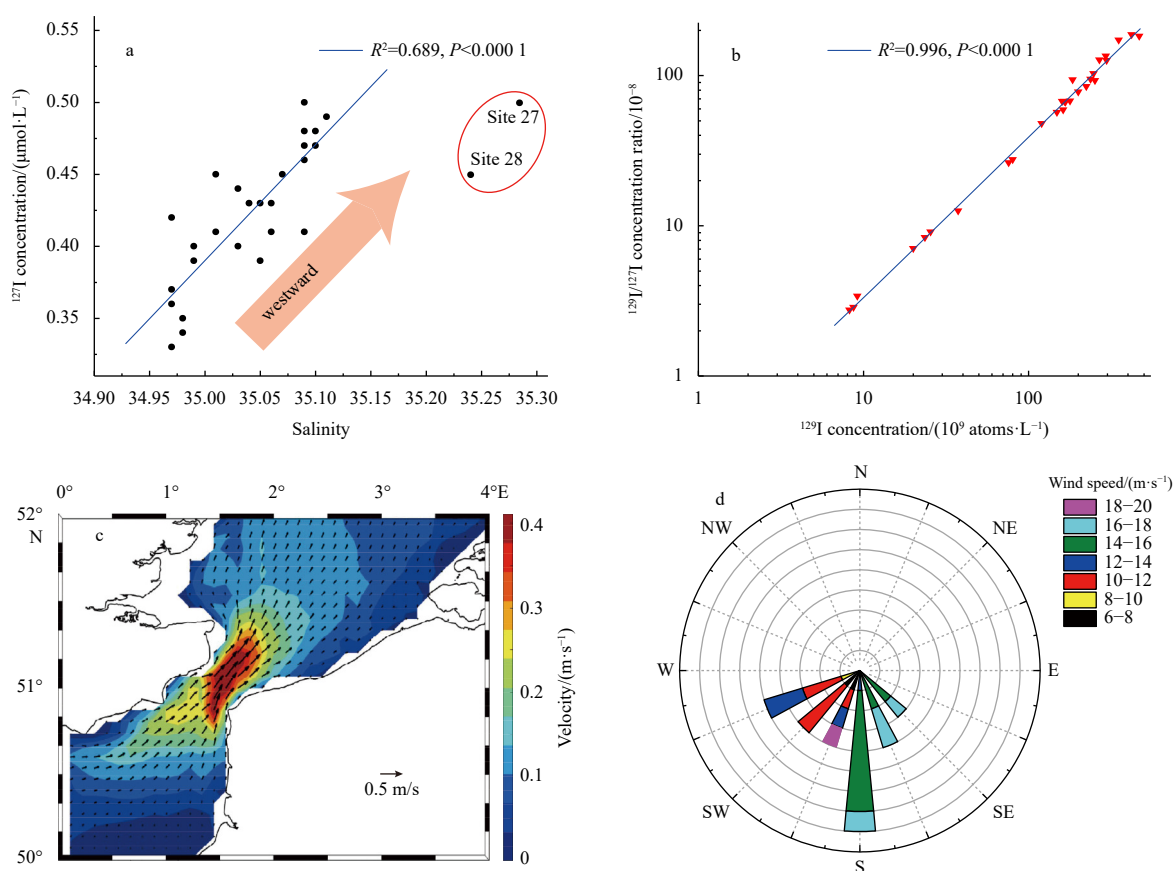


Fig. 5. Distribution of ^{127}I concentration vs. salinity (a), the arrow represents ^{129}I dilution direction; and the relationship between $^{129}\text{I}/^{127}\text{I}$ concentration ratio and ^{129}I concentration (b); depth-mean water circulation velocity (daily mean) in the Dover Strait area (c) and wind conditions during 2010 campaign (d). The data of c obtained from website (<https://resources.marine.copernicus.eu/>) on October 29, 2010.

and southwest that speed up surface water current in this area, which promote rapid mixing of channel water with the North Sea water (Figs 5c, d). The released ^{129}I is mainly transported northward in a narrow vein close to the continental coast, as suggested by a long-term hydrodynamic model (Villa et al., 2015). We note that the wind direction shifted southeasterly in the vicinity of the Dover Strait during our sampling period. This may have enhanced the surface seawater transport of ^{129}I towards the English coast, as suggested by elevated ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ concentration ratios in Sites 2 and 5. The $^{129}\text{I}/^{127}\text{I}$ concentration ratio in Site 3 is half of that in Site 2, which suggests the influence of a relatively iodine-poor water source, possibly river water (e.g., Thames River) input that can dilute ^{127}I concentration and $^{129}\text{I}/^{127}\text{I}$ concentration ratio when it mixes with seawater.

Moving further west, the $^{129}\text{I}/^{127}\text{I}$ concentration ratio in Site 7 and thereafter along the transect decreases from 126×10^{-8} to 2.8×10^{-8} without apparent fluctuations. Previous studies suggest that specific regional hydrography, as well as long-term residual tidal currents and wind conditions regulate the dispersion and advection of the ^{129}I plume, which is confined in a few kilometers off the French coast (Bailly du Bois and Dumas, 2005; Tappin and Millward, 2015). Our data show that ^{129}I concentrations in excess of $100\times 10^9 \text{ atoms/L}$ in the central channel occur east of the Cotentin Peninsula (La Hague). These sampling sites are generally $\sim 100 \text{ km}$ from the French coast and closer to the British coast. Obviously, the northward dispersal of ^{129}I from La Hague may be expected to contaminate the English coast.

Long-term water mass transport simulated by models illustrates a persistent southbound pathway driven by counterclockwise gyres in the NBG (Bailly du Bois et al., 2012; Fiévet et al., 2020). Thus, part of the released conservative radionuclides from the La Hague will advect to the southwest and can reside there for more than 18 months before moving further south, due to the presence of an anticyclonic gyre in the region (Breton and Salomon, 1995; Salomon et al., 1995). Along the transect, a steep gradient, with one order of magnitude decline for both ^{129}I concentration and $^{129}\text{I}/^{127}\text{I}$ concentration ratio, can be observed within a short distance in the central western part samples of the Channel. However, the presence of detectable ^{129}I in the central and the English coast of the western English Channel cannot be ignored, since the ^{129}I concentrations in this area were still at least three orders of magnitudes higher than other non-contaminated oceans. This feature indicates that the entire southern English coast was already influenced by La Hague.

Considering the fact that the total ^{129}I discharged from Sellafield is slightly over a fifth of that of La Hague during 2005–2010 (Fig. 1), $^{129}\text{I}/^{127}\text{I}$ concentration ratio in seawater along the transect in the north-central portion of the western channel suggests that La Hague releases are proportionally larger, especially for Sites 19–22. Sellafield ^{129}I releases generally move northward, so a southward-moving Sellafield ^{129}I plume will become diluted as it approaches the Celtic Sea. However, ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ concentration ratio in the west part of the Channel are comparable to those measured off the west coast of Scotland, and

the east coast of Ireland (Schnabel et al., 2007; Keogh et al., 2007). These observations further support the occurrence of a La Hague ^{129}I signal in the west English Channel, as described by Hou et al. (2007), and this source contributes directly to ^{129}I concentrations in the Celtic Sea and the Atlantic Ocean.

The lowest $^{129}\text{I}/^{127}\text{I}$ concentration ratios ($<1 \times 10^{-9}$) along our transect occur in Sites 23–28 which were collected close to the south coast of the Cornwall area (UK). This region forms the westernmost part of the Channel and is exposed to the Celtic Sea and the Atlantic Ocean. Sources of ^{129}I in this area should be carefully evaluated because influence of Sellafield may involve. Previous investigations confirm a fraction of southward Irish seawater (Bailly du Bois and Guéguéniat, 1999) and this Sellafield-labeled water may occur in the sampled seawater at Sites 23–28. A relatively high ^{129}I concentration at 100 m depth, compared to the surface in the westernmost part of the Channel, was reported by Michel et al. (2012). This feature may be attributed to sinking channel surface waters in this region and a contribution from Sellafield as suggested by He et al. (2014). Due to the lack of iodine isotope data, we cannot conclude that Sellafield releases dominates ^{129}I concentrations off the westernmost UK coast. Nevertheless, the influence of Sellafield NRF in the westernmost part of the English coast at the sampling time should not be ignored.

3.3 Comparison with earlier measurements

The first ^{129}I distribution study in the North Sea and its adjacent areas, including the Channel, was conducted in August 2005. Surface water ^{129}I data were reported by two independent groups (Hou et al., 2007; Michel et al., 2012). Another cruise revisited this region in the summer of 2009 to examine the distribution of iodine, uranium and cesium isotopes (Christl et al., 2015; Daraoui et al., 2016). Although the sampling location of the cruises differ somewhat, we believe it is vital to show temporal changes in ^{129}I that may help focus future research on ^{129}I . Thus, we combine the earlier ^{129}I data collected in the Channel and compare them with our results in the aim of exploring ^{129}I concentration changes in the period 2005–2010.

The average ^{129}I concentrations of samples collected during different cruises in the Channel in 2005, 2009 and 2010, show a decrease from about 250×10^9 atoms/L in 2005 to 160×10^9 atoms/L in 2010 (Fig. 6). However, in the west of La Hague (3° – 5°W), the average ^{129}I concentration of three samples collected in 2005

was $\sim 29 \times 10^9$ atoms/L, which is comparable to the 2010 value for the same longitude ($\sim 26 \times 10^9$ atoms/L). The small number of samples collected in 2005 does not allow us to conclude that ^{129}I concentrations experienced a 5-year decrease from 2005 to 2010. However, in the 2009 expedition, seawater was generally sampled east of Cap de La Hague, and shared seven sampling stations with the 2005 cruise. Sites 903 and 901 of previous studies are close to our Sites 10 and 1, respectively, which provide a view for a comparison (Fig. 7). Within the Channel, three shared sites (904, 903 and 902) are close to the British coast, while the other three shared sites, namely Sites 909, 910 and 911, are closer to the French coast. For all shared stations, ^{129}I concentrations show a 10%–80% decrease from 2005 to 2009, and the concentration differences between the two campaigns in stations off the continental coast, decreased from the source to the Dover Strait (Fig. 7). The total amount of ^{129}I annually discharged from La Hague and Sellafield decreased from 1.7 TBq to 1.3 TBq during 2005–2009, and the ^{129}I released from La Hague decreased substantially. If meteorological conditions were typical during the two sampling events, ^{129}I variations between 2005 and 2009 in Site 909 reflect discharge variations from La Hague, as the transit time from La Hague to Site 909 was estimated to be only one month (Salomon et al., 1995). Nevertheless, it should be noted that this variation was smoothed out with the dilution and dispersal of the ^{129}I plume. In 2010, samples collected near Sites 903 and 901 show an increase in ^{129}I concentration compared to 2009 (Fig. 7). This suggests that the temporal evolution of ^{129}I is positively correlated with the La Hague release functions, at least in the eastern channel. In fact, ^{129}I concentrations in most of the 2010 samples are higher than their nearest 2009 sampling stations in east part of the Channel.

Temporal ^{129}I changes depend on transit times, or transfer functions, for the radioactive plume from La Hague to different parts of the Channel under particular climatic conditions. The transit time decreases significantly from the continental shelf to about 100 km off the coast, where a faster vein (corresponding to a minimum transit time) has been well documented (Salomon et al., 1995). An estimate of 1 year has been reported for the La Hague plume to reach the central North Sea (Dahlgard et al., 1995). A much shorter transit time of 6 months was estimated for the plume to be transported to the Rhine River Estuary (Herrmann et al., 1995). Note that the transit time should not represent a precise number, but rather a temporal relationship between the dis-

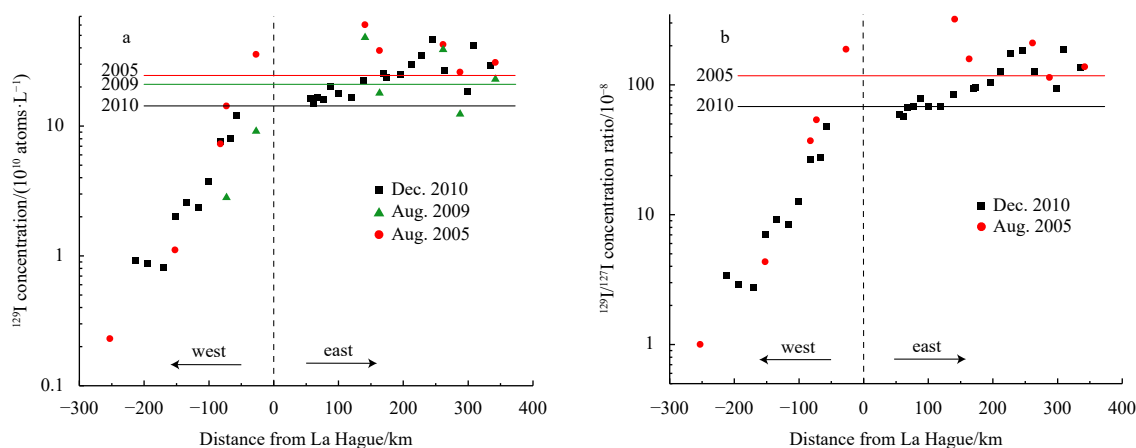


Fig. 6. Distribution of ^{129}I concentration (a) and $^{129}\text{I}/^{127}\text{I}$ concentration ratio (b) from the 2005, 2009 and 2010 sampling campaigns with distance from La Hague (Michel et al., 2012; Christl et al., 2015). The horizontal lines represent average values in 2005, 2009 and 2010.

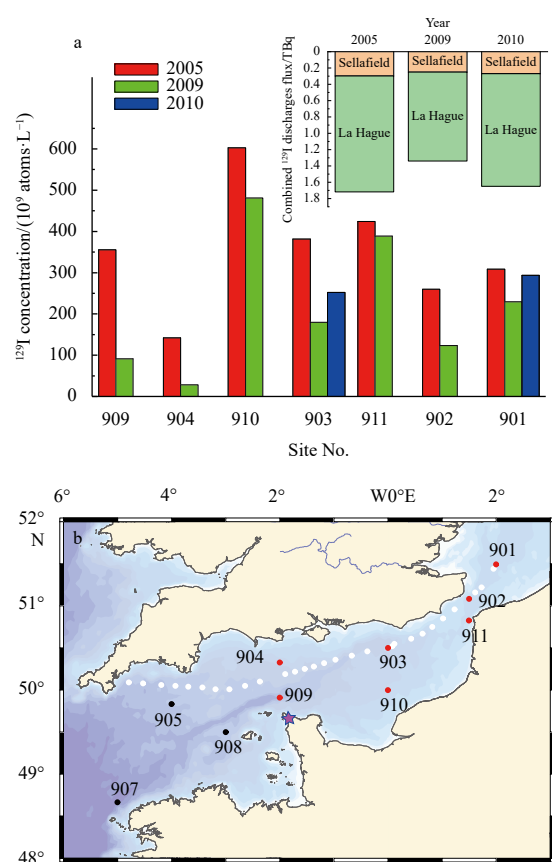


Fig. 7. Comparison of ^{129}I concentrations from the sampling campaigns 2005, 2009 and 2010. Sites 1 and 10 (this work) are used because they are close to Sites 903 and 901, respectively (a). b. the location of the ^{129}I seawater sampling sites in 2010 (white dots) and 2005 (blank dots). Red dots represent sampling sites shared by both the 2005 and 2009 cruises. The star shows the location of the La Hague site.

charge and transport functions of a radionuclide. Transit times for ^{129}I discharged from La Hague to the extreme northeastern sampling site (Site 1) vary from 2 to 6 months, depending on the distance of the water vein from the French coast (Guéguénat et al., 1995; Salomon et al., 1995). Therefore, our samples collected in the central channel, with lower transit time compared to a pathway along the continental coast, seems more sensitive to discharge intensity changes from La Hague. Unfortunately, monthly ^{129}I discharge data are not available and thus the variability is integrated over annual discharge data which do not accurately predict the magnitude of change as given by seawater datasets (2005–2010) collected on individual days. However, ^{129}I discharges from La Hague maintain a relatively high level of approximately 200–250 kg/a after 2010 (Fig. 1). Therefore, ^{129}I concentration in the Channel, at least in the eastern part, is expected to continue to rise.

High biochemical mobility and long half-life provide conditions for easy concentration of ^{129}I in ocean organisms. Seawaters containing high ^{129}I can also pollute ambient terrestrial environment through sea-spray and oceanic emission-precipitation mechanism, which may eventually end up in human thyroid (Fréchou and Calmet, 2003). Nevertheless, the highest ^{129}I level measured in this campaign was still 3 orders of magnitudes lower than the World Health Organization (WHO) guideline for drink-

ing water (Snyder et al., 2010). Considering the relatively low ^{129}I level in fresh water ($\sim 10^{10}$ atoms/L in the English Lake District; Atarashi-Andoh et al., 2007), and that the ^{129}I transfer from sea-food to human thyroid is not straightforward, this radionuclide currently does not pose a radioactive risk in the Channel region. However, it should be monitored regularly, as there is no indication that the discharge of ^{129}I from La Hague will be substantially reduced soon. The results presented here, form a basis to better understand the biogeochemical behavior of ^{129}I and identify potential ecological hazards in the Channel area.

4 Conclusions

The ^{129}I distributions in surface waters of the central English Channel indicate high values between the Dover Strait and La Hague, followed by a dramatic drop towards the west. The ^{129}I concentrations represent some of the highest values in the marine environment globally. Levels of ^{129}I concentration and $^{129}\text{I}/^{127}\text{I}$ concentration ratio clearly reflect water mass circulation and mixing within the Channel, which were generally in accord with the output of long-term published hydrodynamic models. Our data reveal ^{129}I dispersal in the central part of the western channel. These observations indicate that the entire channel region, from south to north and east to west, is highly influenced by anthropogenic ^{129}I . ^{129}I in central and west part of the Channel is mainly related to La Hague releases, whereas a Sellafield source may contribute measurable ^{129}I to waters in the westernmost channel. Time-dependent ^{129}I variations between 2005 and 2010 are linked to the La Hague marine discharge pattern. ^{129}I concentrations are not currently environmentally harmful but require regular monitoring. More data on the depth distribution of iodine isotopes in the Channel waters, and ^{129}I concentrations in the food web, and human thyroid pathways are needed in future to provide a comprehensive understanding of possible ecological hazards.

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References

- Alfimov V, Aldahan A, Possnert G, et al. 2004. Anthropogenic iodine-129 in seawater along a transect from the Norwegian coastal current to the North Pole. *Marine Pollution Bulletin*, 49(11–12): 1097–1104, doi: [10.1016/j.marpolbul.2004.08.019](https://doi.org/10.1016/j.marpolbul.2004.08.019)
- Atarashi-Andoh M, Schnabel C, Cook G, et al. 2007. $^{129}\text{I}/^{127}\text{I}$ ratios in surface waters of the English Lake District. *Applied Geochemistry*, 22(3): 628–636, doi: [10.1016/j.apgeochem.2006.12.008](https://doi.org/10.1016/j.apgeochem.2006.12.008)
- Bailly du Bois P, Dumas F. 2005. Fast hydrodynamic model for medium- and long-term dispersion in seawater in the English Channel and southern North Sea, qualitative and quantitative validation by radionuclide tracers. *Ocean Modelling*, 9(2): 169–210, doi: [10.1016/j.ocemod.2004.07.004](https://doi.org/10.1016/j.ocemod.2004.07.004)
- Bailly du Bois P, Dumas F, Solier L, et al. 2012. *In-situ* database toolbox for short-term dispersion model validation in macro-tidal seas, application for 2D-model. *Continental Shelf Research*, 36: 63–82, doi: [10.1016/j.csr.2012.01.011](https://doi.org/10.1016/j.csr.2012.01.011)
- Bailly du Bois P, Guéguénat P. 1999. Quantitative assessment of dissolved radiotracers in the English Channel: Sources, average impact of La Hague reprocessing plant and conservative behaviour (1983, 1986, 1988, 1994). *Continental Shelf Research*,

- 19(15–16): 1977–2002, doi: [10.1016/S0278-4343\(99\)00049-7](https://doi.org/10.1016/S0278-4343(99)00049-7)
- Breton M, Salomon J C. 1995. A 2D long term advection-dispersion model for the Channel and Southern North Sea Part A: Validation through comparison with artificial radionuclides. *Journal of Marine Systems*, 6(5–6): 495–513, doi: [10.1016/0924-7963\(95\)00020-P](https://doi.org/10.1016/0924-7963(95)00020-P)
- Castrillejo M, Casacuberta N, Christl M, et al. 2018. Tracing water masses with ^{129}I and ^{236}U in the subpolar North Atlantic along the GEOTRACES GA01 section. *Biogeosciences*, 15(18): 5545–5564, doi: [10.5194/bg-15-5545-2018](https://doi.org/10.5194/bg-15-5545-2018)
- Christl M, Casacuberta N, Lachner J, et al. 2015. Status of ^{236}U analyses at ETH Zurich and the distribution of ^{236}U and ^{129}I in the North Sea in 2009. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 361: 510–516, doi: [10.1016/j.nimb.2015.01.005](https://doi.org/10.1016/j.nimb.2015.01.005)
- Cooper L W, Hong G H, Beasley T M, et al. 2001. Iodine-129 concentrations in marginal seas of the north Pacific and Pacific-influenced waters of the Arctic Ocean. *Marine Pollution Bulletin*, 42(12): 1347–1356, doi: [10.1016/S0025-326X\(01\)00151-5](https://doi.org/10.1016/S0025-326X(01)00151-5)
- Dahlgaard H. 1995. Transfer of European coastal pollution to the Arctic: radioactive tracers. *Marine Pollution Bulletin*, 31(1–3): 3–7, doi: [10.1016/0025-326X\(95\)00003-6](https://doi.org/10.1016/0025-326X(95)00003-6)
- Dahlgaard H, Herrmann J, Salomon J C. 1995. A tracer study of the transport of coastal water from the English Channel through the German Bight to the Kattegat. *Journal of Marine Systems*, 6(5–6): 415–425, doi: [10.1016/0924-7963\(95\)00017-J](https://doi.org/10.1016/0924-7963(95)00017-J)
- Daraoui A, Tosch L, Gorny M, et al. 2016. Iodine-129, Iodine-127 and Cesium-137 in seawater from the North Sea and the Baltic Sea. *Journal of Environmental Radioactivity*, 162–163: 289–299, doi: [10.1016/j.jenvrad.2016.06.006](https://doi.org/10.1016/j.jenvrad.2016.06.006)
- Fiévet B, Bailly du Bois P, Voiseux C, et al. 2020. A comprehensive assessment of two-decade radioactivity monitoring around the Channel Islands. *Journal of Environmental Radioactivity*, 223–224: 106381, doi: [10.1016/j.jenvrad.2020.106381](https://doi.org/10.1016/j.jenvrad.2020.106381)
- Fréchou C, Calmet D. 2003. ^{129}I in the environment of the La Hague nuclear fuel reprocessing plant—from sea to land. *Journal of Environmental Radioactivity*, 70(1–2): 43–59, doi: [10.1016/S0265-931X\(03\)00127-9](https://doi.org/10.1016/S0265-931X(03)00127-9)
- Fréchou C, Calmet D, Bertho X, et al. 2002. $^{129}\text{I}/^{127}\text{I}$ ratio measurements in bovine thyroids from the North Cotentin area (France). *Science of the Total Environment*, 293(1–3): 59–67, doi: [10.1016/S0048-9697\(01\)01145-7](https://doi.org/10.1016/S0048-9697(01)01145-7)
- Guéguéniat P, Bailly du Bois P, Salomon J C, et al. 1995. FLUX-MANCHE radiotracers measurements: A contribution to the dynamics of the English Channel and North Sea. *Journal of Marine Systems*, 6(5–6): 483–494, doi: [10.1016/0924-7963\(95\)00019-L](https://doi.org/10.1016/0924-7963(95)00019-L)
- He Peng, Aldahan A, Hou Xiaolin, et al. 2016. Tracing variability in the iodine isotopes and species along surface water transect from the North Sea to the Canary Islands. *Journal of Radioanalytical and Nuclear Chemistry*, 307(2): 1407–1412, doi: [10.1007/s10967-015-4449-y](https://doi.org/10.1007/s10967-015-4449-y)
- He Peng, Hou Xiaolin, Aldahan A, et al. 2014. Radioactive ^{129}I in surface water of the Celtic Sea. *Journal of Radioanalytical and Nuclear Chemistry*, 299(1): 249–253, doi: [10.1007/s10967-013-2792-4](https://doi.org/10.1007/s10967-013-2792-4)
- Herrmann J, Kershaw P J, Bailly du Bois P, et al. 1995. The distribution of artificial radionuclides in the English Channel, southern North Sea, Skagerrak and Kattegat, 1990–1993. *Journal of Marine Systems*, 6(5–6): 427–456, doi: [10.1016/0924-7963\(95\)00026-L](https://doi.org/10.1016/0924-7963(95)00026-L)
- Hou Xiaolin, Aldahan A, Nielsen S P, et al. 2007. Speciation of ^{129}I and ^{127}I in seawater and implications for sources and transport pathways in the North Sea. *Environmental Science & Technology*, 41(17): 5993–5999, doi: [10.1021/es070575x](https://doi.org/10.1021/es070575x)
- Hou Xiaolin, Dahlgaard H, Nielsen S P. 2001. Chemical speciation analysis of ^{129}I in seawater and a preliminary investigation to use it as a tracer for geochemical cycle study of stable iodine. *Marine Chemistry*, 74(2–3): 145–155, doi: [10.1016/S0304-4203\(01\)00010-X](https://doi.org/10.1016/S0304-4203(01)00010-X)
- Keogh S M, Aldahan A, Possnert G, et al. 2007. Trends in the spatial and temporal distribution of ^{129}I and ^{99}Tc in coastal waters surrounding Ireland using *Fucus vesiculosus* as a bio-indicator. *Journal of Environmental Radioactivity*, 95(1): 23–328, doi: [10.1016/j.jenvrad.2007.01.009](https://doi.org/10.1016/j.jenvrad.2007.01.009)
- Michel R, Daraoui A, Gorny M, et al. 2012. Iodine-129 and iodine-127 in European seawaters and in precipitation from Northern Germany. *Science of the Total Environment*, 419: 151–169, doi: [10.1016/j.scitotenv.2012.01.009](https://doi.org/10.1016/j.scitotenv.2012.01.009)
- Orano Recyclage. 2020. Rapport annuel de surveillance de l'environnement Orano la Hague: Edition 2020. Orano la Hague, 1–161. [https://www.orano.group/docs/default-source/orano-doc/groupe/publications-reference/rapport-annuel-surveillance-environnement-la-hague-2020.pdf?sfvrsn=7814fe3c_23\[2021-06-12\]](https://www.orano.group/docs/default-source/orano-doc/groupe/publications-reference/rapport-annuel-surveillance-environnement-la-hague-2020.pdf?sfvrsn=7814fe3c_23[2021-06-12])
- Raisbeck G M, Yiou F, Zhou Zhiqiong, et al. 1995. ^{129}I from nuclear fuel reprocessing facilities at Sellafield (U. K.) and La Hague (France); potential as an oceanographic tracer. *Journal of Marine Systems*, 6(5–6): 561–570, doi: [10.1016/0924-7963\(95\)00024-J](https://doi.org/10.1016/0924-7963(95)00024-J)
- Salomon J C, Breton M, Guéguéniat P. 1995. A 2D long term advection-dispersion model for the Channel and southern North Sea Part B: Transit time and transfer function from Cap de La Hague. *Journal of Marine Systems*, 6(5–6): 515–5227, doi: [10.1016/0924-7963\(95\)00021-G](https://doi.org/10.1016/0924-7963(95)00021-G)
- Schnabel C, Olive V, Atarashi-Andoh M, et al. 2007. $^{129}\text{I}/^{127}\text{I}$ ratios in Scottish coastal surface sea water: geographical and temporal responses to changing emissions. *Applied Geochemistry*, 22(3): 619–627, doi: [10.1016/j.apgeochem.2006.12.007](https://doi.org/10.1016/j.apgeochem.2006.12.007)
- Sellafield Ltd. 2021. Monitoring our environment: discharges and environmental monitoring, Annual Report 2020. Nuclear Decommissioning Authority, 1–97. [https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/1035775/2020_Sellafield_Ltd_Monitoring_Environmental_-_Discharges_and_Monitoring_-_Annual_Report_optimised_.pdf\[2021-07-18\]](https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/1035775/2020_Sellafield_Ltd_Monitoring_Environmental_-_Discharges_and_Monitoring_-_Annual_Report_optimised_.pdf[2021-07-18])
- Smith J N, Jones E P, Moran S B, et al. 2005. Iodine 129/CFC 11 transit times for Denmark Strait Overflow Water in the Labrador and Irminger Seas. *Journal of Geophysical Research: Oceans*, 110(C5): C05006, doi: [10.1029/2004JC002516](https://doi.org/10.1029/2004JC002516)
- Snyder G, Aldahan A, Possnert G. 2010. Global distribution and long-term fate of anthropogenic ^{129}I in marine and surface water reservoirs. *Geochemistry, Geophysics, Geosystems*, 11(4): Q04010, doi: [10.1029/2009GC002910](https://doi.org/10.1029/2009GC002910)
- Tappin A D, Millward G E. 2015. The English Channel: Contamination status of its transitional and coastal waters. *Marine Pollution Bulletin*, 95(2): 529–550, doi: [10.1016/j.marpolbul.2014.12.012](https://doi.org/10.1016/j.marpolbul.2014.12.012)
- Truesdale V M W, Nausch G, Baker A. 2001. The distribution of iodine in the Baltic Sea during summer. *Marine Chemistry*, 74(2–3): 87–98, doi: [10.1016/S0304-4203\(00\)00115-8](https://doi.org/10.1016/S0304-4203(00)00115-8)
- Truesdale V M W, Upstill-Goddard R. 2003. Dissolved iodate and total iodine along the British east coast. *Estuarine, Coastal and Shelf Sciences*, 56(2): 261–270, doi: [10.1016/S0272-7714\(02\)00161-0](https://doi.org/10.1016/S0272-7714(02)00161-0)
- Villa M, López-Gutiérrez J M, Kyung-Sukh K S, et al. 2015. The behaviour of ^{129}I released from nuclear fuel reprocessing factories in the North Atlantic Ocean and transport to the Arctic assessed from numerical modelling. *Marine Pollution Bulletin*, 90(1–2): 15–24, doi: [10.1016/j.marpolbul.2014.11.039](https://doi.org/10.1016/j.marpolbul.2014.11.039)
- Xing Shan, Hou Xiaolin, Aldahan A, et al. 2017. Water circulation and marine environment in the Antarctic traced by speciation of ^{129}I and ^{127}I . *Scientific Reports*, 7: 7726, doi: [10.1038/s41598-017-07765-w](https://doi.org/10.1038/s41598-017-07765-w)