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**Quantitative Assessment of Dissolved Radiotracers in the English Channel :
sources, average impact of la Hague reprocessing plant and conservative behaviour
(1983, 1986, 1988 and 1994).**

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Abstract :

Knowledge of long-term movements of water-masses in the English Channel has been substantially improved using hydrodynamic modelling coupled with radio-tracers studies ; nevertheless, the precision of results so obtained is still largely dependent on measurement precision.

New tools are now available to make more accurate determinations of radio-tracer distribution :

- Repositioning of station locations at the same tide reference-time, giving a homogeneous spatial data set, coupled with the possibility of interpolating and quantifying the amounts of dissolved radioactivity flowing through the English Channel ;

- The first measurements of tritium (^3H) in seawater on a large scale in the English Channel demonstrate that this fully conservative radionuclide is a clearly identifiable marker of industrial releases ;

- Recent campaigns carried out during the FluxManche II CCE (1994) programme show the general distribution of dissolved radionuclides ^{137}Cs , ^{134}Cs , ^{60}Co , ^{125}Sb , ^{106}Ru and ^3H in the English Channel and the Irish Sea ;

- The re-utilisation of data from previous campaigns (1983, 1986 and 1988) provides indications, at any given location in the English Channel, about the average dilution and distribution of releases derived from the La Hague reprocessing plant.

Excesses and losses of radionuclides are now quantified with respect to known source-terms ; estimates of losses are provided for non-conservative radionuclides, while an excess of ^{137}Cs was observed in the English Channel during the period 1983 - 1994. This excess which has the same order of magnitude as the quantities released from La Hague plant in the English Channel, could be explained by about 1% of Sellafield reprocessing plant releases entering the Channel.

These results confirm and give a more detailed picture of the previously known distribution of water masses in the English Channel. They lead to clear information about transit times and dilution at this scale, and provide directly comparable data for the validation of hydrodynamic models.

1. INTRODUCTION

1.1. Interest of artificial radiotracers for studies of English Channel

The English Channel (Fig. 1), which extends between the north coast of France and the south coast of the United Kingdom, represent a transition zone between the Atlantic ocean to the west and the North Sea to the east. Because of the particular topography of the continental shelf that amplifies the dynamic tidal pulse during its passage from west to east, this sea is affected by strong periodic tidal currents. These tides, combined with the influence of an average south-westerly wind, give rise to a residual current running from west to east ; waters entering the English Channel come in from the Atlantic via the Western Approaches, and leave to enter the North Sea through the Straits of Dover.

Even though this general circulation has been known for a long time, it has proved more difficult to know precisely the transport routes and the water fluxes involved. The use of tracers which are present as soluble species with conservative behaviour in seawater has emerged as an ideal tool for such studies.

In this context, the radioactive tracers are of particular interest. Artificial radionuclides occurring as dissolved species in seawater are useful tools for long-term current studies. Their source-terms are usually few in number and are well characterised. Some radionuclides have a conservative behaviour during their transport, notably possessing a suitably long half-life, and remaining dissolved in seawater without fixation onto other constituents of the environment such as sediments or living organisms. Furthermore, it is possible to measure extremely low concentrations of such radionuclides in seawater (the detection limit of gamma emitters reaches a level around 40 000th of natural radioactivity).

Since 1960, large-scale studies have chiefly concerned caesium 137, a radionuclide that originates principally from the nuclear fuel reprocessing plant at Sellafield on the Irish Sea (Kautsky, 1988 ; Kershaw and Baxter, 1995 ; Dahlgaard *et al.*, 1986). Atmospheric fallout from nuclear tests and releases from the reprocessing plant at La Hague used to represent a much lower contribution as regards this element, but the accident at the Chernobyl power plant in 1986 provided a new important source-term for the marine environment. Other tracers have also been used to follow the transport of water masses : caesium 134, strontium 90 and technetium 99. Since 1983, the IPSN-Octeville Marine Radioecology Laboratory (LRM, actually LERFA) has undertaken oceanographic campaigns covering the English Channel which have measured radionuclides specific of releases from La Hague : antimony 125, ruthenium 106, and - more recently - tritium.

Otherwise, great progress has occurred in the hydrodynamic modelling of water-mass movement, and recent advances in computing allow the simulation of the long-term physical transport and dispersion of waters on the scale of the English Channel and North Sea. In association with conservative radioactive tracers that enable high-precision calibration of computer simulations, numerical modelling is an increasingly indispensable tool for the study of the marine environment.



Figure 1 : Situation map and location of main sources of artificial radioactivity in the English Channel and Irish Sea

1.2. Aims of the study

New tools were developed to allow a cartographic and quantitative treatment of the results of campaigns for measuring the concentrations of elements transported in seawater. These techniques applied to recent studies make it possible to specify the distribution of dissolved artificial radioactivity in the English Channel and the Irish Sea.

Previous campaigns also benefit from such tools, since they enable the comparative evaluation of labelled areas and the amounts of radioactivity in the English Channel over several years. The circulation pattern and long-term dilution factors of radiolabelled waters can thus be established, and the measured amounts of radioactivity compared with known sources. As a result, it would be possible to estimate the loss rate of non-conservative radionuclides in the Channel, and to quantify potential excesses of radioactivity corresponding to unknown or poorly characterised sources (Chernobyl accident fallout).

The quantitative evaluation carried out in this study provides a basis for estimating the inventories of radioactivity fixed by various other compartments of the marine environment (sediments and living organisms).

2. MATERIALS AND METHODS

2.1. Campaigns

The subject matter of this work originates from campaigns carried out in 1983, 1986, 1988 and 1994 in the English Channel and Irish Sea on board ships of the Institut Français de Recherche pour l'Exploitation de la Mer (IFREMER, Fr.), Centre National de la Recherche Scientifique (CNRS, Fr.), Ministry of Agriculture, Fisheries and Food (MAFF, G.B.), and from measurements performed by the Groupe d'Etudes Atomiques (GEA) of the French Navy and the Institut de Protection et de Sûreté Nucléaire (IPSN-LERFA, Fr). The 1994 and following cruises were realised within the framework of the CCE MAST II FluxManche programme.

Except for the campaigns in 1988 and 1994, the results presented here have already been published and discussed elsewhere (Guéguéniat *et al.*, 1988). In the present work, the original sampling positions were corrected for tidal movements to calculate positions for the sampled water mass at a common reference time. The measures with corrected position were then interpolated on the body of the studied domains. These techniques allow quantitative exploitation of the punctual measurements, and easier comparisons between the different campaigns.

2.2. Sampling and analysis

2.2.1. Measurements of ^{125}Sb , ^{106}Ru , ^{137}Cs , ^{134}Cs , ^{60}Co

Techniques adapted to the dilution conditions observed in the marine environment and the on-board treatment of samples were developed in order to measure artificial activities of the order of 0.3 Bq/m^3 . This compares with the natural radioactivity of seawater, which is of the order of $12\,000 \text{ Bq/m}^3$; the detection of such low activities requires sample volumes varying from 100 litres to more than a cubic metre. The dissolved radionuclides are concentrated by co-precipitation by means of specific adsorbents.

Seawater samples for radioactivity measurements were collected by pumping from a depth of 3 m. The analyses were performed without filtering, that is, the measurements include dissolved as well as particulate activity. This is primarily of importance in the case of ^{106}Ru and ^{60}Co due to their higher tendency to associate with particulate material. The analyses of ^{125}Sb , ^{106}Ru , ^{137}Cs , ^{134}Cs and ^{60}Co were carried out at the GEA at Cherbourg, and the IPSN at Octeville, France. The activities of ^{125}Sb and ^{106}Ru were measured by gamma-ray spectrometry after concentration of the radionuclides from seawater samples of 120-1200 l using manganese dioxide at pH 3.5 (Gandon and Guéguéniat, 1992). Since the concentration of suspended particulate matter is always less than 50 mg/l, all the ^{125}Sb is present in the soluble phase $\text{Sb}(\text{OH})_6^-$ in normally oxygenated seawater (Gandon and Guéguéniat, 1992). Under such conditions, the dissolved radionuclide is adsorbed onto MnO_2 with an extraction yield of 97% at pH 3.5. Because of the relatively high statistical counting errors (15, 10 and 5% for activities of the order of 1.5, 5 and 15 Bq/m^3 , respectively), no yield correction was applied. However, the extraction yields for ^{106}Ru are highly variable, being dependent on the stability of ruthenium complexes; ^{106}Ru coming from reprocessing plants at La Hague is present in seawater as nitrosylruthenium II complexes, and only the hydrolysis products of such species are adsorbed onto MnO_2 (Gandon *et al.*, 1993), with the consequence that this method produces random results when used to concentrate ruthenium. When ^{60}Co is present in seawater as Co^{2+} , it is adsorbed with antimony onto MnO_2 , nevertheless ^{60}Co is present in La Hague effluent in other chemical forms which are not fixed on this adsorbent and extraction yields are highly variable.

^{137}Cs and ^{134}Cs (half-lives of 30 years and 2.2 years, respectively) were extracted from the same seawater samples by adding Co-K ferrocyanide powder (at a concentration of 8.10^{-2} g/l) to the

manganese dioxide precipitate, thus bringing about ion exchange with the caesium in solution. In seawater, caesium is present only as the soluble species Cs^+ and extraction yield is always 100%.

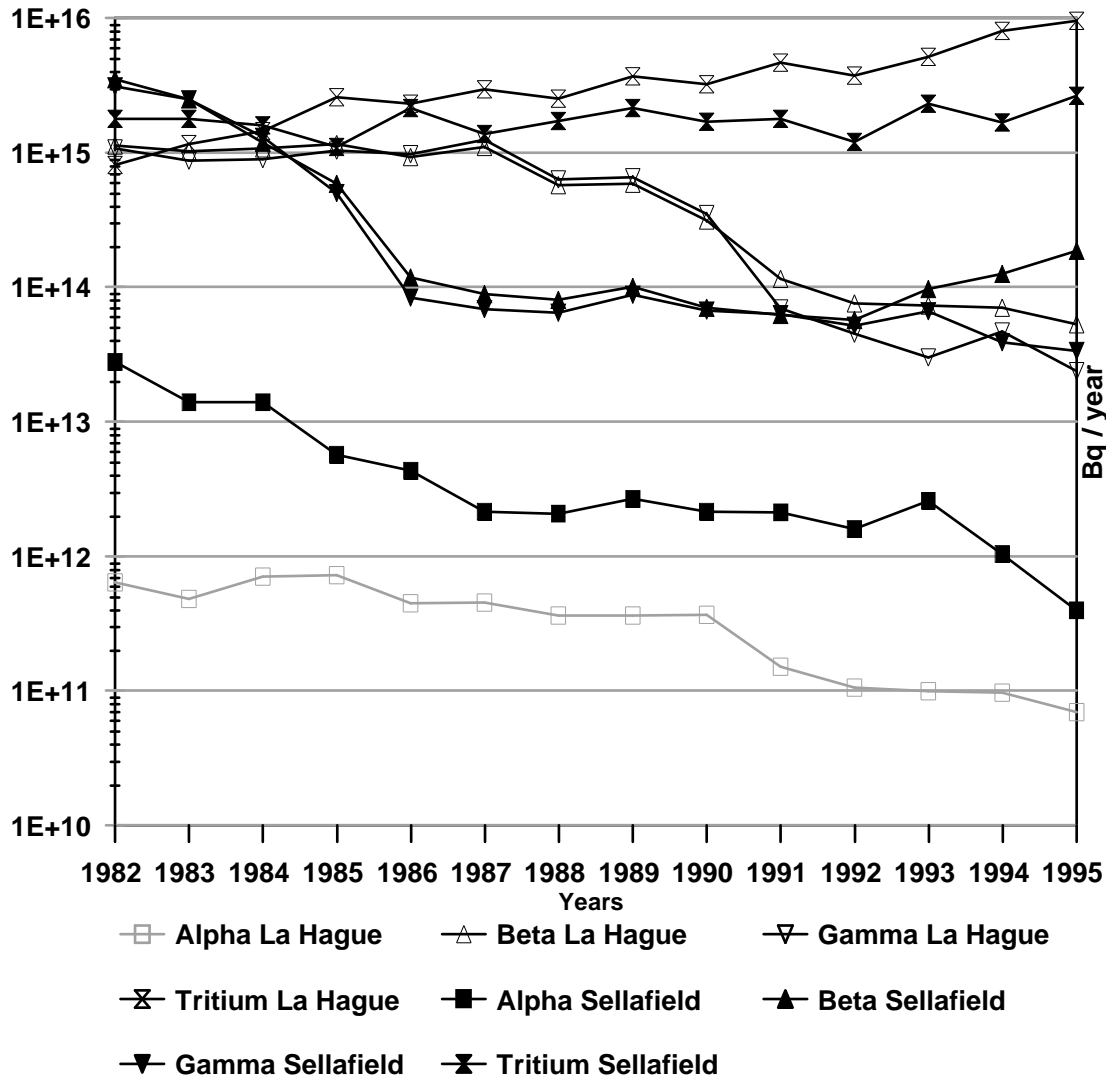


Figure 2 : Annual liquid discharges from La Hague and Sellafield

From 1991 onwards, a new extraction method was used that involves associating a precipitate of copper ferrocyanide with ferric hydroxide (Gandon and Guéguéniat, 1992 ; Gandon *et al.*, 1993). This enables the coprecipitation of ^{125}Sb , ^{106}Ru and radiocaesium at a constant yield evaluated at 100% in the case of wastes released from the reprocessing plant at La Hague.

2.2.2. Tritium : a new fully conservative tracer in English Channel waters

In 1994, tritium (3H) was measured for the first time on a large scale in English Channel waters ; the uniform increase of releases of this element from the La Hague plant leads us to suppose that it could be a good tracer of its dispersion in the Channel in comparison to the seawater background. In fact, since this element is present in the effluent as tritiated water and cannot be concentrated or eliminated, the releases in the liquid effluents are proportional to the quantities of nuclear fuel retreated, in contrast to the other radionuclides that have diminished by a factor of about ten over the last ten years (Fig. 2).

The direct measurement of 3H by liquid scintillation has a detection limit of the order of 1000 Bq/m³, insufficient to establish the influence of industrial releases in comparison with the seawater background existing at the entrance of the English Channel, which is of the order of 300 Bq/m³. It is

possible to enrich the ^3H content of the sample by a factor 10 or more by means of electrolysis, but such a technique is difficult to set up in practice. Because of this, the present authors approached the SMSRB laboratory (a division of the French Commissariat à l'Energie Atomique - CEA), which makes routine use of this technique under favourable operational conditions (detection limit between 150 and 230 Bq/m³).

2.3. Correction of sample positions using tide data

2.3.1. Advantage of repositioning

In a sea with strong tides such as the English Channel, periodic movements of seawater can have a large amplitude (more than 30 km from the cape of La Hague); a sample taken from seawater at a given hour and place, could correspond to a water mass situated a long distance away some hours later. This leads to an important uncertainty on the position of the water samples (or other substances transported by seawater : dissolved elements, suspended particles, plankton). By way of example, two samples collected at some hours interval, one to the north of Guernsey, the other to the south of this island, may under certain conditions sample exactly the same water volume. By the same token, two samples taken at the same position at an interval of six hours could represent waters that, in reality, are tens of kilometres apart (**Fig. 3a**).

In order to represent in a homogenous and precise manner the distribution of tracers associated with seawater in the Channel, it is essential to correct the sample position. This position, which is given in terms of the reference frame of the solid Earth, has to be corrected in comparison with the dynamic reference frame of moving water masses. The relative positioning of the stations with respect to each other can be significantly modified since sampling is generally carried out at any moment of the tide. After correction of the positions, provided the number and distribution of samples permit, interpolations and quantitative inventories can be performed on a set of homogeneous measurements.

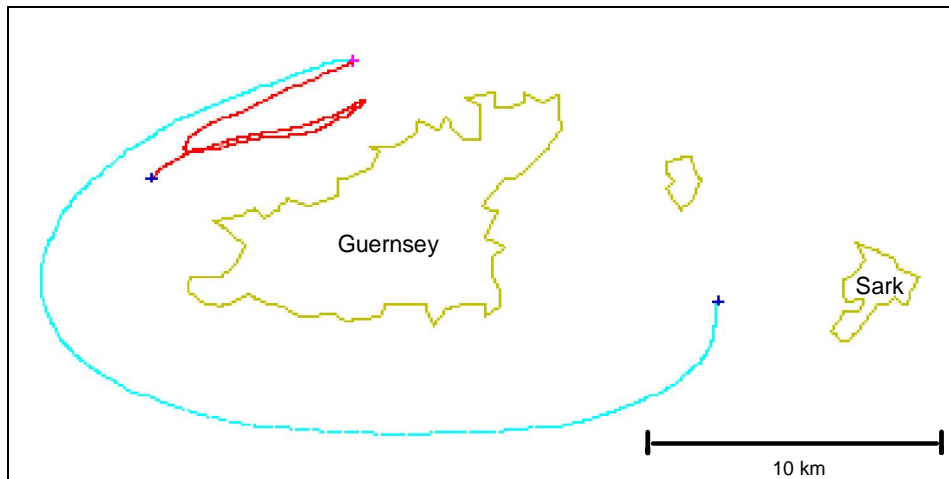


Figure 3 a : 12 hours movements of the seawater after two stations made at the same location (north of Guernsey island)

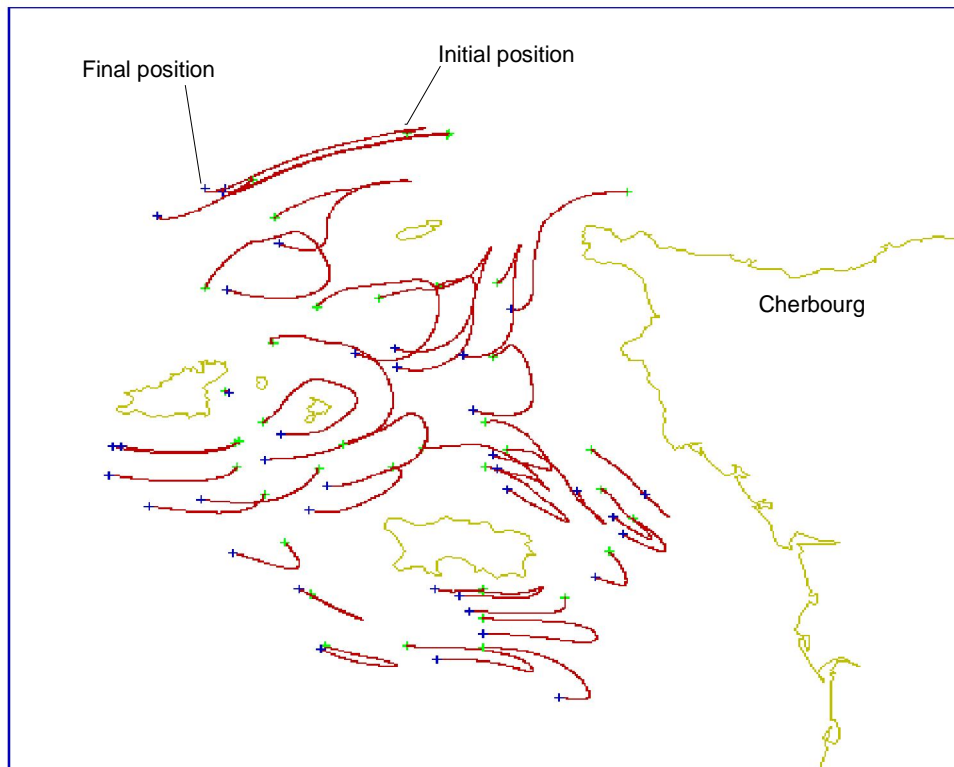


Figure 3 b : Corrections of stations locations at the same tide-reference time (full tide at Brest). Campaign made in the English Channel in 1995

2.3.2. Methods used

Considering the periodicity of the tide, and knowing the characteristics of associated currents, it is possible to recalculate sample locations to the position that they would have had at a particular reference time of the tidal cycle, carrying out this operation for all samples in the English Channel. The essential preliminary requirement is to have access to a database describing instantaneous currents in the English Channel ; such a database should be comprehensive, reliable and easy to use.

The studies carried out by J.C. Salomon at IFREMER on the numerical modelling of marine hydrodynamic systems (Salomon *et al.*, 1991) establish a model covering the whole Channel with a resolution of a nautical mile (1.85 km). This model is able to generate instantaneous currents with a time-step of twelve minutes, for all conditions of wind and tide.

For the validation of this model, use was made of the measurements of dissolved radiotracers performed by the LERFA since 1986. This approach demonstrates the reliability of the model in

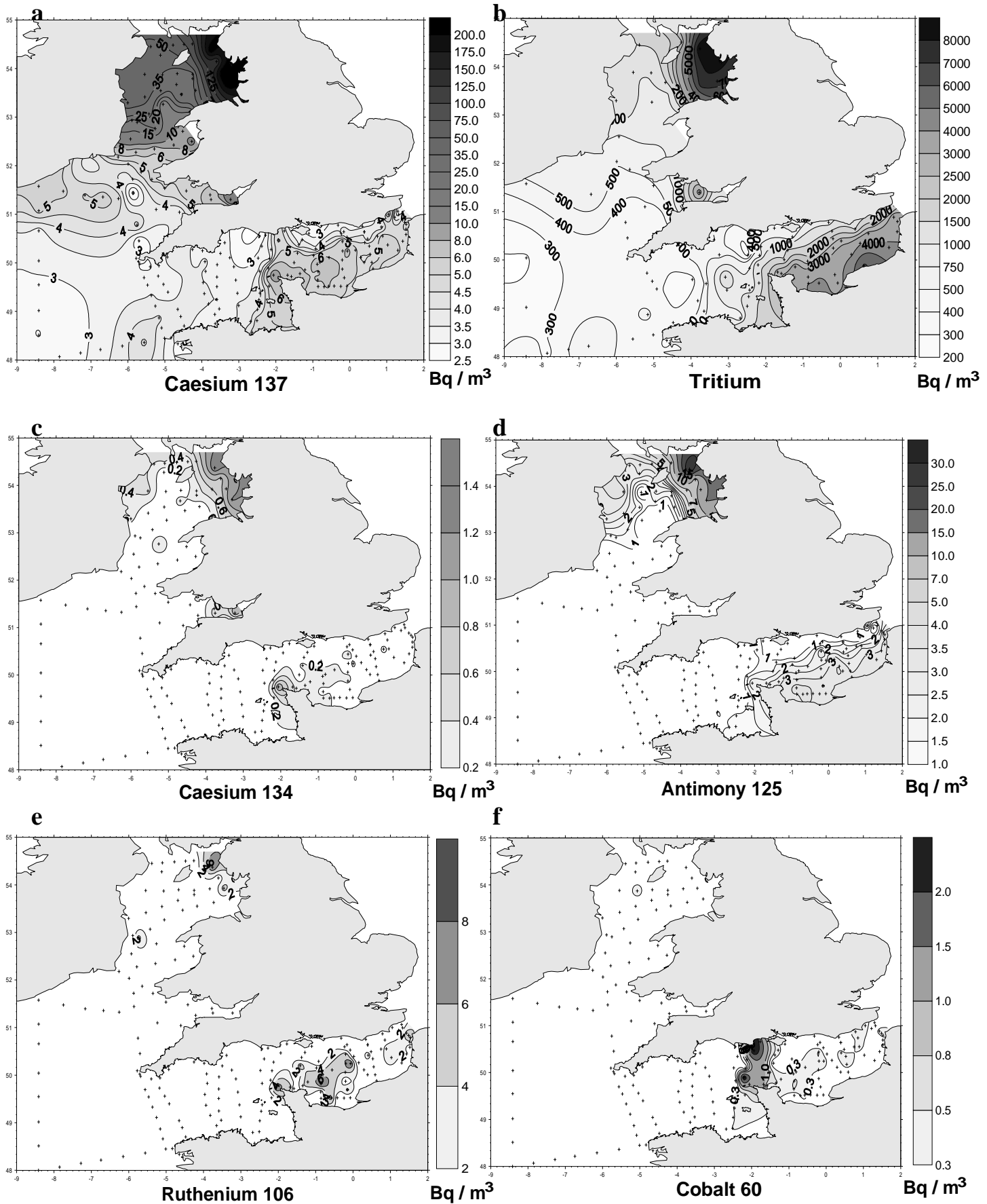
describing the long-term pattern of seawater transport and, as a consequence, the short-term transport as well (the velocity of long-term movements represents only a few percent of the instantaneous fluxes). The values of instantaneous fluxes derived from this model were used to perform the repositioning.

The tables published by the SHOM (Hydrographic and Oceanographic Service of the French Navy) make it possible to find the tidal coefficient for any date, along with the time of high tide at Brest, which is the reference used in this study. The positions of the samples after correction therefore correspond to the position that the waters would have had at the high tide at Brest nearest in time to the hour of sampling (Fig. 3b). In these conditions, the time - correction is always less than six hours, and except very close from a point of release, the effect of dispersion is negligible

The program developed by the LERFA to perform these calculations ("Recalmer" software) is available to laboratories who may request it for research purposes.

2.4. Mapping of radioactive tracer distribution

When campaigns had a sufficient extension, punctual measurements of radioactivity were interpolated over the whole surface of the English Channel in uniform meshes several km wide, the eastern part was only taken into account for campaigns restricted to this area. This treatment allows visualisation and comparison of the results obtained with ^{137}Cs , ^{125}Sb , ^3H , $^{106}\text{Ru}+\text{Rh}$, ^{134}Cs and ^{60}Co (figure 4 a - f). The interpolation method used is the "Krigage" from "SURFER"TM software.



+ Samples locations

Figure 4 : Gedymac campaign, 28/08 - 13/09/1994, radioactivity in seawater a : caesium 137, b : tritium, c : caesium 134, d : antimony 125, e : ruthenium 106, f : cobalt 60.

Figure 4 shows results obtained from the Gedymac campaign, which has the most complete coverage, summarising very well the distribution of radiotracers during the FluxManche II programme. The distributions observed in previous years were quite similar, with higher levels of radioactivity from La Hague and Sellafield corresponding to higher releases from these plants in the past.

3. RESULTS AND DISCUSSION

3.1. General description of radiotracer distribution (figs. 4 a-f)

The raw data gives a good illustration of the circulation of water masses on the Irish Sea and English Channel scale. The different radionuclides measured (^{137}Cs , ^3H , ^{134}Cs , ^{125}Sb , $^{106}\text{Ru}+\text{Rh}$ and ^{60}Co) are consistent, and follow the general distribution pattern recorded previously. The results for ^3H are interesting and confirm that releases from the La Hague and Sellafield plants could be well distinguished from other source terms (background, rivers, rain) of this element, which is strictly conservative in seawater.

The distribution of ^{137}Cs or ^3H (figures 4a - 4b) summarises well the circulation of water masses in the Channel and Irish Sea. Levels representative of Atlantic background increase slightly at the entrance of the Channel (2.9 - 4.1 Bq/m³). The La Hague plant's influence extends from the centre of the Channel, along the French coast of the Eastern Channel, with little influence in the north. The Cherbourg - Wight transect reveals generally the same distribution, with an influence of La Hague up to the middle of the transect. It can be noted that for each campaign, ^{60}Co is measured near the Isle of Wight, which could possibly be attributed to the Winfrith power plant.

In the Irish Sea, the Sellafield impact is much more important around the Isle of Man, with a strong decrease southwards. South of 51°N, levels approach background, but are generally higher than expected in pure Atlantic background surface water (2.5 Bq/m³ for ^{137}Cs , 300 Bq/m³ for ^3H).

3.2. Sources and dispersion of artificial radionuclides in the English Channel, Irish sea and north-east Atlantic

3.2.1. Characteristics of source-terms observed in 1994 - table I

1) Background of Atlantic surface waters (natural, nuclear tests and Chernobyl fallout) : 190 - 300 Bq/m³ of ^3H ; 2.5 - 3 Bq/m³ of ^{137}Cs (Holm et al., 1991 ; Dahlgard *et al.*, 1995 ; Nyffeler *et al.*, 1997 ; Bourlat *et al.*, 1997).

2) Nuclear fuel reprocessing plants at Sellafield on the Irish Sea : ^3H (1000 - 8000 Bq/m³), ^{137}Cs (30 - 220 Bq/m³), ^{125}Sb (2.5 - 31 Bq/m³), ^{134}Cs (0.3 - 1.4 Bq/m³), $^{106}\text{Ru}+\text{Rh}$ (4 - 8 Bq/m³).

3) Nuclear fuel reprocessing plant at La Hague (central Channel, French coast) : ^3H (1000 - 6000 Bq/m³), ^{137}Cs (3 - 27 Bq/m³), ^{125}Sb (1 - 6 Bq/m³), ^{134}Cs (0.2 - 2.4 Bq/m³), $^{106}\text{Ru}+\text{Rh}$ (1 - 13 Bq/m³), ^{60}Co (0.2 - 1.4 Bq/m³).

4) Nuclear power plant at Winfrith : ^{60}Co (0.4 - 2.5 Bq/ m³) and in the Bristol Channel.

Table 1
 Main characteristics and sources of the studied radionuclides

Radionuclide	Source		Background of North Atlantic surface waters in Bq/m ³		Industrials releases in PBq (10 ¹⁵)				Chernobyl fallout
	Symbol	Half-life (years)	Natural	Atmospheric tests of nuclear weapons	Sellafield		La Hague		
					1980 - 1994	1952 - 1996	1980 - 1994	1966 - 1996	
Tritium	³ H	12.35	100	200 *	25.7	48.8	46.4	66.8	
137 Caesium	¹³⁷ Cs	30.15	0	2.5 - 3 *	9.45	41.1	0.27	1	yes
106 Ruthenium +106 Rhodium	¹⁰⁶ Ru + ¹⁰⁶ Rh	1.02	0	0	4.2	55.2	7.9	13.2	
125 Antimony	¹²⁵ Sb	2.76	0	0	0.22	0.28	1.1	1.6	
134 Caesium	¹³⁴ Cs	2.07	0	0	0.71	5.8	0.06	0.1	yes
60 Cobalt	⁶⁰ Co	5.27	0	0	0.005		0.1	0.1	

* From Holm et al., 1991 ; Dahlgard et al., 1995 ; Nyffeler *et al.*, 1997 ; Bourlat *et al.*, 1997.

3.2.2. Fallout from nuclear tests and Atlantic background radioactivity

The atmospheric nuclear tests, that mainly stopped in 1962, although Chinese test 1966 - 1980 gave contributions (French atmospheric tests in the southern hemisphere 1966 - 1974 have only limited effect here), introduced most of their fission products into the upper atmosphere and were then progressively deposited onto the oceans and continents. At present (1994), the North Atlantic ocean showed generally homogenous labelling of the surface waters in ¹³⁷Cs and ³H, with levels of about 2.5 and 300 Bq/m³ respectively (including tritium of natural origin). Because of their half-life, the other radionuclides studied here are not detectable in Atlantic background water.

3.2.3. Controlled industrial releases

Nuclear reprocessing plants and nuclear power plants are authorised to release controlled amounts of radioactive substances into the sea. These releases, made principally in Europe by the La Hague and Sellafield plants have significantly decreased in the last decade (Fig. 2).

3.2.4. Radiolabelling from La Hague

The La Hague plant (Fig. 1) releases radioactive compounds in the centre of the English Channel, in the strong Raz Blanchard current, at the extremity of the Cotentin peninsula. Antimony 125, nearly absent from other sources and conservative in seawater, is the reference marker to identify the water plume labelled by the La Hague plant. It clearly shows the dispersion of these releases in the English Channel and the North Sea (Guéguéniat *et al.*, 1988 ; Guéguéniat *et al.*, 1993 ; Guéguéniat *et al.*, 1994 ; Bailly du Bois *et al.*, 1993 ; Bailly du Bois *et al.*, 1995).

The results presented here (Fig. 4) confirm those already obtained concerning the dispersion of effluents originating from La Hague in the central English Channel (Guéguéniat *et al.*, 1988 ; Guéguéniat *et al.*, 1993). The measured activities of ¹³⁷Cs, ¹³⁴Cs, ¹²⁵Sb, ¹⁰⁶Ru and ⁶⁰Co reflect the strong decrease of releases since 1983, the annual maxima being for ¹²⁵Sb of 200 Bq/m³ in 1983 and 5 Bq/m³ in 1994, with average levels in the east of the Channel in the order of 30 - 70 Bq/m³ in 1983 and 1 - 3 Bq/m³ in 1994. These levels represent about one 10 000th of dissolved natural radioactivity in seawater - mainly due to potassium 40 (12 000 Bq/m³) - and approach the limits of measurable levels.

Ignoring the fluctuations observed during different campaigns - attributed to the variability of releases and meteorological conditions - an overall structure can be pointed out. The dispersion extends globally from west to east, with a decreasing gradient from the French coast to the English coast, these coasts being only weakly labelled by La Hague's plume. The westward extension of the labelling along the French coast generally limits itself to the Gulf of Saint-Malo. The residual current lines revealed by hydrodynamic modelling (Fig. 5 ; Salomon *et al.*, 1991) provides a physical explanation for this dispersion which is discussed elsewhere (Guéguéniat *et al.*, 1988 ; Salomon *et al.*, 1991). Furthermore, releases from La Hague were simulated under realistic conditions and globally showed a very good agreement between measurements and numerical modelling (Salomon *et al.*, 1991 ; Bailly du Bois *et al.*, 1995 ; Breton and Salomon, 1995).

The dispersion of other radionuclides released by La Hague is globally similar, notably for conservative elements (^{137}Cs , ^{134}Cs and ^3H). Tritium measured in 1994 showed that it is a representative tracer of the releases from La Hague in the English Channel, the maximum observed being in the order of 6000 Bq/m^3 along the « Pays de Caux » coasts, in the eastern part of the Channel where the Seine river and nuclear power plant's influence is the stronger one. ^{106}Ru and ^{60}Co , having a greater affinity for particles, their distribution does not extend towards the east, suggesting a progressive disappearance during their transport.

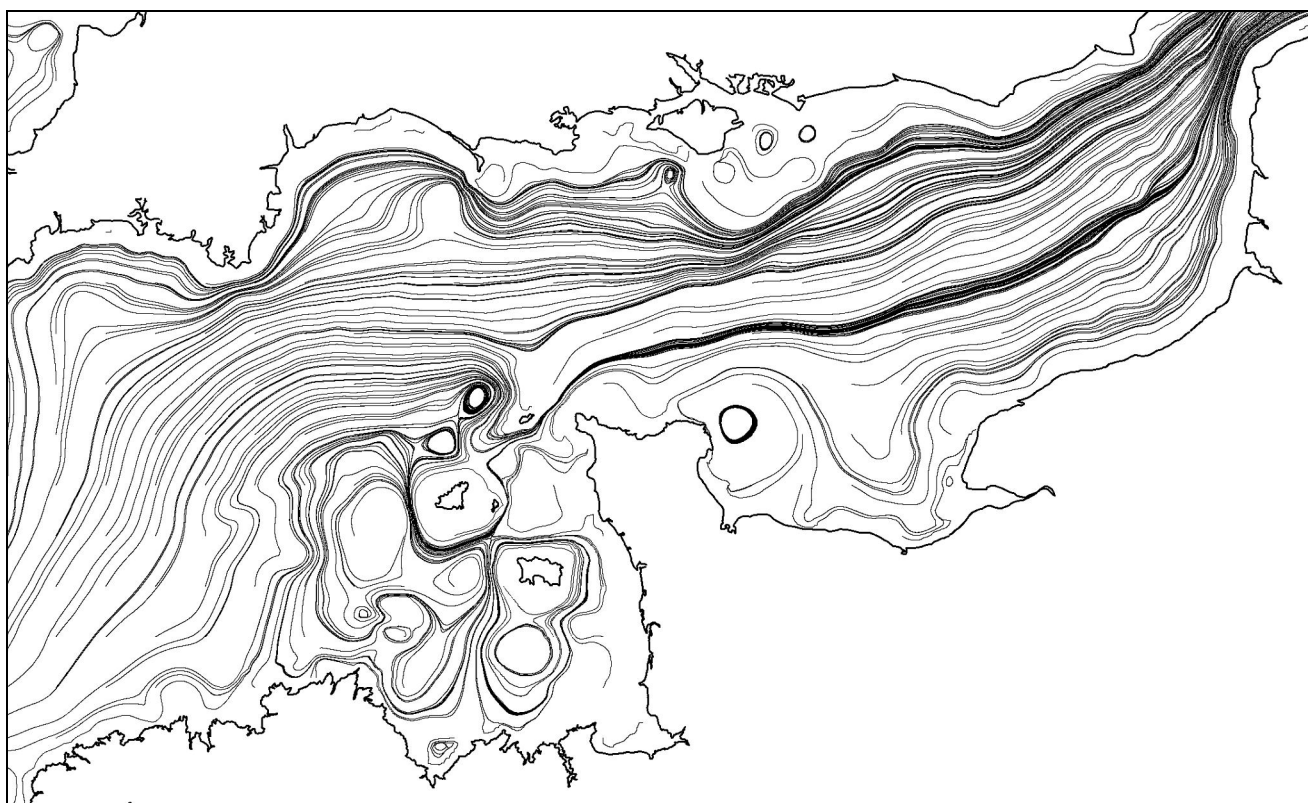


Figure 5 : Numerical simulation of long term trajectories in the English Channel, (average tide, W.S.W. wind ; Salomon J.C., 1991)

3.2.5. Radiolabelling from Sellafield

The Sellafield plant (Fig. 1) releases radionuclides into the Irish Sea which label seawater rather homogeneously north of 53°N (Fig. 4). These releases proceed principally north towards the North Sea, and are the subject of many publications (Kershaw *et al.*, 1987 ; Condren *et al.*, 1996). Because of the amounts released, ^{137}Cs have been a good tracer to identify Sellafield's impact ; releases have strongly decrease since 1986, nevertheless Irish Sea sediments which have fixed great amounts of this radionuclide still represent an important source for ^{137}Cs .

The results obtained in this study (Fig. 4) confirm previous studies, while showing pronounced labelling towards the south to 52° N, the levels of ¹³⁷Cs are 10 to 100 times higher than those measured in the English Channel (50 - 2000 Bq/m³ in 1983, 20 - 150 Bq/m³ in 1994). The labelling observed here is also more pronounced for other radionuclides (¹²⁵Sb, ³H and ¹³⁴Cs see Fig. 4). The strong concentration gradient observed between 52° and 53° N (Saint-George's Channel, Fig. 1) shows the limit in the Irish Sea where hydrodynamics allow a certain homogeneity of waters, which contrasts with a flow of less labelled waters originating from the south. Nevertheless this boundary could be transgressed, since higher levels of ¹³⁷Cs measured in 1983 and 1994, and ³H in 1994 found south of Ireland probably originate from releases at Sellafield. Fluctuations in the Atlantic background water could also possibly be attributed to Sellafield releases. At present, it is not possible to establish with certainty the origin of this labelling, several hypotheses are evoked :

- direct transit from the Irish Sea to the south ;
- passage around the north of Ireland and along the west coast, and return off southern Ireland ;
- return of waters labelled by Sellafield since more than five years ago, after their transit in the North Sea, Norway Sea and Arctic Ocean, returning via Iceland in the North Atlantic surface waters. This last possibility would probably result in a much more diluted and homogenous signal than the first two.

3.2.6. Nuclear power stations

The nuclear power plants generally represent a small fraction of the industrial contribution of artificial radionuclides, and only their local influences can be demonstrated. Thus significant labelling of ⁶⁰Co, observed since 1986 (Fig. 4f) around of the Isle of Wight, can be associated with releases from the nuclear power plant at Winfrith (Fig. 1). Releases from this plant represent a quarter of releases from La Hague for the period 1985 - 1989 (table II). . A higher labelling in ³H was observed in 1994 along the coast of the Pays de Caux (Upper Normandy) ; the influence of releases originating from the nuclear power plants (Fig. 1) of Paluel and Penly (1% of the La Hague releases, table II) may be envisaged, but remains to be confirmed. Finally, labelling was observed at the far end of the Bristol channel (Fig. 1), with different isotopic ratios to those of Sellafield (³H/¹³⁷Cs : 200 - 400 against 15 - 60 ; ¹³⁷Cs/¹³⁴Cs : 10 - 16 against 40 - 150), this is liable to be attributed to the Hinkley and Cardiff plants. The contribution of the Dungeness power plant (Fig. 1), east of the English Channel is also significant (table II), but did not appear clearly at the time of the campaign (09/94).

Table 2

Industrial releases in The English Channel and Irish Sea in 1994 in GBq (10¹² Bq)

Industrial plants :	Sellafield	Other Irish Sea ⁽¹⁾	Bristol Channel ⁽²⁾	La Hague	Winfrith	Dungeness	Palluel + Penly	All EDF power plants ⁽³⁾
Tritium	1680	744.5	872.3	8088	57	407.9	90	202
106 ruthenium + 106 rhodium	13.5	na	na	27.45	na	na	na	na
125 antimony	12.2	na	na	7.19	na	na	na	na
137 caesium	13.8	0.01	1.23	10.45	na	0.185	0.00055	0.00276
60 cobalt	0.11	0.0008	0.001	0.53	0.007	0.0024	0.00261	0.0061

⁽¹⁾ Chapelcross, Barrow, Heysham, Springfields, Capenhurst, Wylfa, Trawsfynydd (MAFF report)

⁽²⁾ Cardiff, Oldbury, Berkeley, Hinkley Point (MAFF report)

⁽³⁾ Palluel, Penly, Flamanville, Nogent (Seine), Gravelines (Pas de Calais) (EDF environmental report)

na : not available (low releases)

3.2.7. Fallout from the Chernobyl accident

Fallout material from the Chernobyl accident extended over all of Europe, and can be recognised by a diffuse labelling of surfaces waters spiked directly by deposition from the atmosphere, or via rivers reaching the sea that received a contribution from rainwashing of soils. The amount of labelling can be calculated taking into account the following factors :

- the isotopic ratio of $^{137}\text{Cs}/^{134}\text{Cs}$ was 1.86 in the fallout at the time of the accident ;
- ^{134}Cs was absent in the background of Atlantic waters before the accident ;
- since the average isotopic ratio of $^{125}\text{Sb}/^{134}\text{Cs}$ in the releases during the period preceding the accident is known, the amount of ^{134}Cs originating from releases of the La Hague plant can be estimated from the activities of ^{125}Sb (N.B. a poor representation of this contribution in the vicinity of La Hague may be seen because of rapid variations of this ratio).

Radiolabelling attributable to the presence of Chernobyl fallout in the English Channel is represented in figure 6. The levels observed in Atlantic waters are probably due to direct atmospheric fallout, while the labelling in the eastern part of the Channel (east of long. 3°W) is linked to the direct fallout combined with fluvial contributions coming from the French and English coasts. The weak labelling in the western Channel is probably the consequence of lack of precipitation in this region in the weeks following the accident.

Globally, the activities of ^{137}Cs originating from the Chernobyl accident and present in English Channel waters can be estimated as 13 TBq at the end of 1986.

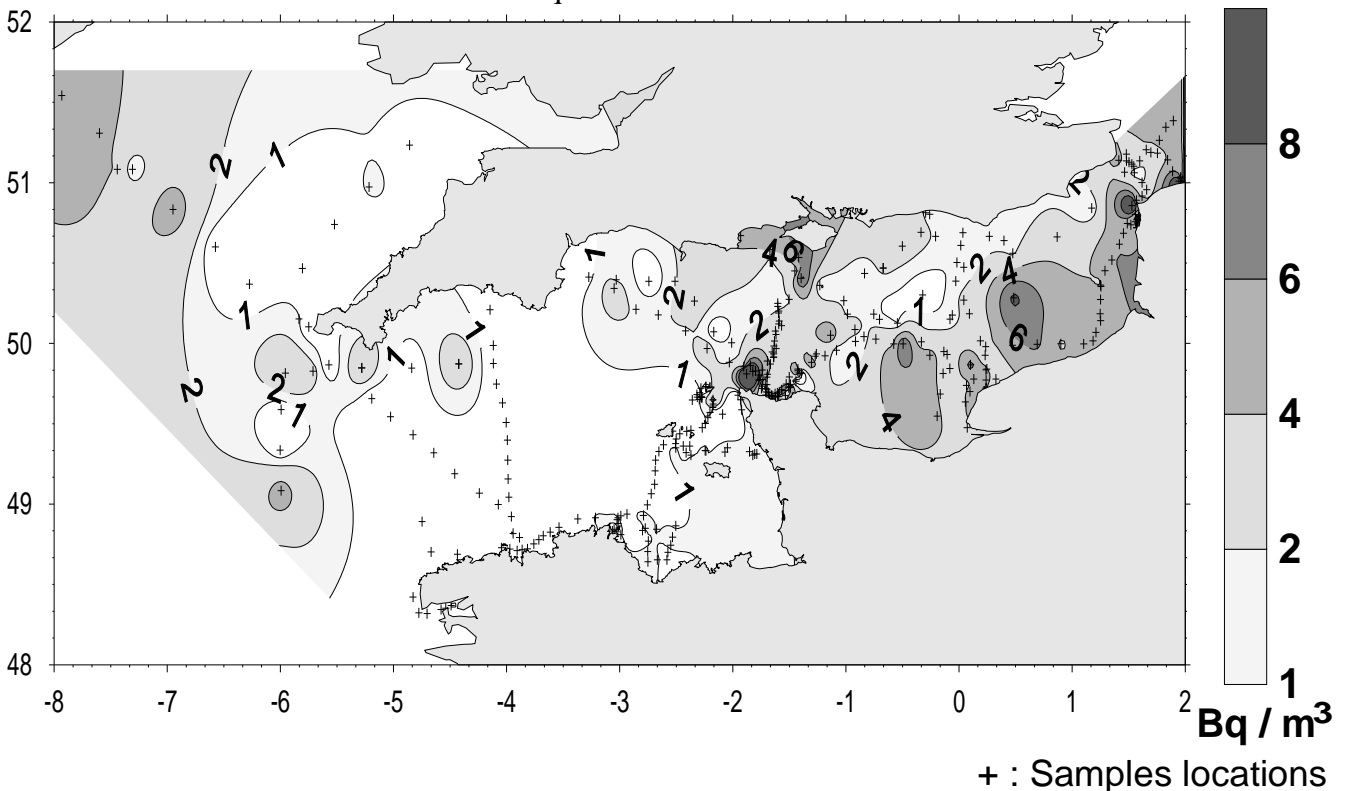


Figure 6 : Caesium 134 coming from Chernobyl accident in the English Channel at the end of 1986

3.2.8. Radioactive waste dumping, tables 3 and 4

Table 3

Average distribution of radionuclides for all dumping sites in the North Atlantic and English Channel from 1942 to 1982 (according to OECD report - AEN 1985 " Review of the continued suitability of the dumping site for radioactive waste in the North-Est. Atlantic ").

Alpha emitters	Beta + gamma emitters
Pu-239 : 41%	tritium : 41%
Pu-240 : 21%	Pu 241 : 23%
Pu-238 : 11%	Co 60 : 3%
Am-241 : 22%	Sr 90 : 1%
	Cs 137 : 2%
other : 5%	other : 30%

Table 4

Total quantities deposited in Atlantic and English Channel

	End of dumping	Mass (tons)	Alpha (TBq)	Beta + Gamma (PBq)	Tritium (PBq)
Central Trench (fosse des Casquets)	1963	16,558	14.5	0.046	
North Atlantic	1982	129,846	671	41.4	15.2

Dumping was stopped in 1982 after the adoption of a moratorium on the immersion of radioactive waste. To this day, it remains impossible to identify leakage from dumped material in the English Channel. Such leaks may eventually contribute to the background of artificial radioactivity in the western approaches of the English Channel, but in the current state of knowledge, it is not possible to differentiate them from other sources. In the same way, even if leaks did take place from the seabed dumping site situated in the Central Trench (Fig. 1, fosse des Casquets in French), the quantities dumped would be insufficient to give rise to detectable amounts of the measured radionuclides in seawater. The background of seawater in the English Channel, as well as the releases from the La Hague plant situated nearby, would mask any eventual labelling. Any leakage would be first detected through the alpha emitting activity that would be fixed onto the sediments or living organisms close to the dumping site.

3.3. Inventory of radioactivity in the English Channel

The shallow depths (less than 200 m) and strong tidal dynamics in the English Channel mean that it can be considered as a poorly stratified water body, so surface measurements of radioactivity can be reasonably extrapolated to the whole water column, principally to the east of long. 4°W (Pingree, 1980). From the individual radioactivity measurements interpolated over the whole surface of the Channel, and taking account of the bathymetry, the total inventory of radioactivity present can be calculated for all the measured radionuclides and all the campaigns. Considering the complexity of the residual circulation pattern in the English Channel (Fig. 5) and the observed variability from one year to another, only two zones were selected to calculate the radioactivity inventories presented here :

- the whole of the English Channel, delimited by a line joining the west of Brittany to Land's End in the west, and the Dover Straits in the east ;
- the eastern English Channel, delimited by a line joining the Cap de La Hague and the Isle of Wight in the west, and the Dover Straits in the east.

Table 5

Radioactivity inventories estimated for the English Channel from 1983 to 1994.

Volume :	Whole English Channel						Eastern English Channel					
	4702 km ³						1576 km ³					
	¹³⁷ Cs	¹³⁴ Cs	¹⁰⁶ Ru	¹²⁵ Sb	⁶⁰ Co	³ H	¹³⁷ Cs	¹³⁴ Cs	¹⁰⁶ Ru	¹²⁵ Sb	⁶⁰ Co	³ H
Year	TBq					PBq	TBq					PBq
1983	44.1	2.13	107.59	126.41	0.9	na	20.05	1.89	53.04	85.63	0.05	na
1986	41.9	7.29	98.7	72.6	1.4	na	23.26	5.29	84.34	57.71	1.16	na
1988	na	na	na	na	na	na	10.89	na	57.30	32.52	na	na
09/1994	18.94	0.27	3.67	3.69	0.75	5.65	7.89	0.21	3.47	3.17	0.58	3.95
11/1994	na	na	na	na	na	na	10.27	0.60	0.65	2.35	0.17	na

na : not available

The quantities obtained (table 5) depend greatly on the meteorological conditions and fluctuations in releases at a given moment, but taking into account the four studied periods, an estimate of the average quantity of radioactivity present in the Channel can be obtained in comparison with releases from La Hague, which is the main source. Using antimony-125 as a specific marker to identify waters labelled by the La Hague plant, the following average estimates are obtained :

- 15 to 30% of the radioactivity originating from La Hague is located to the west of the Cotentin Peninsula, and does not directly participate in the flow of waters from the English Channel towards the North Sea ;
- the English Channel taken as a whole contains the equivalent of seven months of releases ;
- the eastern English Channel contains the equivalent of five to six months of releases.

Assuming that the flow is not laminar (Fig. 5) and that not all zones participate in the same manner to the flow, the average transit time of 4 - 5 months obtained by other methods (Guéguéniat *et al.*, 1993 ; Kershaw *et al.*, 1987 ; Salomon *et al.*, 1993 ; Bailly du Bois *et al.*, 1995) is compatible with this new data.

On the basis of these data obtained with ¹²⁵Sb, it is possible to compare the quantities released from La Hague for the other radionuclides with the correspondent quantities measured in the English Channel, and to evaluate the percentage of losses or excesses of radioactivity measured (table 6). To have a realistic comparison, releases from La Hague have to be corrected with radioactive decay associated to the correspondent duration of release (25 or 32 weeks for the eastern or the whole English Channel). It is also necessary to take account of all known sources in order to derive them from the calculated inventories.

The industrial releases of ¹⁰⁶Ru and ⁶⁰Co are known and these two radionuclides appear to be absent in the background of Atlantic waters. Measurements carried out in 1994 show an excess of ⁶⁰Co that could be associated with the significant labelling observed in the vicinity of Winfrith, so the levels measured cannot be attributed to recent releases from Winfrith or la Hague. In the absence of supplementary data allowing us to explain these observations, the measurements from 1994 were excluded from the calculation.

The following sources have been subtracted :

- Atlantic seawater background originating from the fallout of nuclear atmospheric tests (2.5 Bq/m³ of ¹³⁷Cs and 200 Bq/m³ of ³H) ;
- contribution of ¹³⁷Cs and ¹³⁴Cs from the Chernobyl accident in 1986 as presented above (§3.2.7., Fig. 6).

Table 6

Comparison of measured quantities of radioactivity in the English Channel with correspondent releases from La Hague during 1983 to 1994.

Antimony-125 is used as reference for the calculation ; background of Atlantic surface waters (^{137}Cs , ^3H) and Chernobyl fallout (^{137}Cs , ^{134}Cs) have been deduced.

Volume : Equivalent release duration :	Whole English Channel						Eastern English Channel					
	4702 Km ³ 32 weeks - 7.3 months						1576 Km ³ 25 weeks - 5.7 months					
	^{137}Cs	^{134}Cs	^{106}Ru	^{125}Sb	^{60}Co	^3H	^{137}Cs	^{134}Cs	^{106}Ru	^{125}Sb	^{60}Co	^3H
Number of campaigns	3	2	3	3	3	1	5	3	5	5	2	1
Fraction of the La Hague release	233 %	86 %	26 %	98 %	14 %	103 %	139 %	83 %	19 %	98 %	8 %	121 %

As ^{125}Sb is used as a reference, results are normally equilibrated for this radionuclide ; results for ^3H , which is perfectly conservative, confirms the approach. Concerning ^{60}Co and ^{106}Ru , it appear that 74 - 92 % of these radionuclides leave the water mass, fixed on bottom sediments. Considering the uncertainties of measurements for these radionuclides (cf. §2.2), these fractions represent a maximum. The proportion of losses represent only 14 - 17 % for ^{134}Cs which could be considered as nearly conservative at this scale. For ^{137}Cs , the quantities measured represent more than twice the quantities released from La Hague in the English Channel and shows clearly that other sources have to be accounted to explain this excess. These results will be discussed for each radionuclide with cartographic representation in the following paragraph.

3.4. Average impact of La Hague reprocessing plant (figure 7)

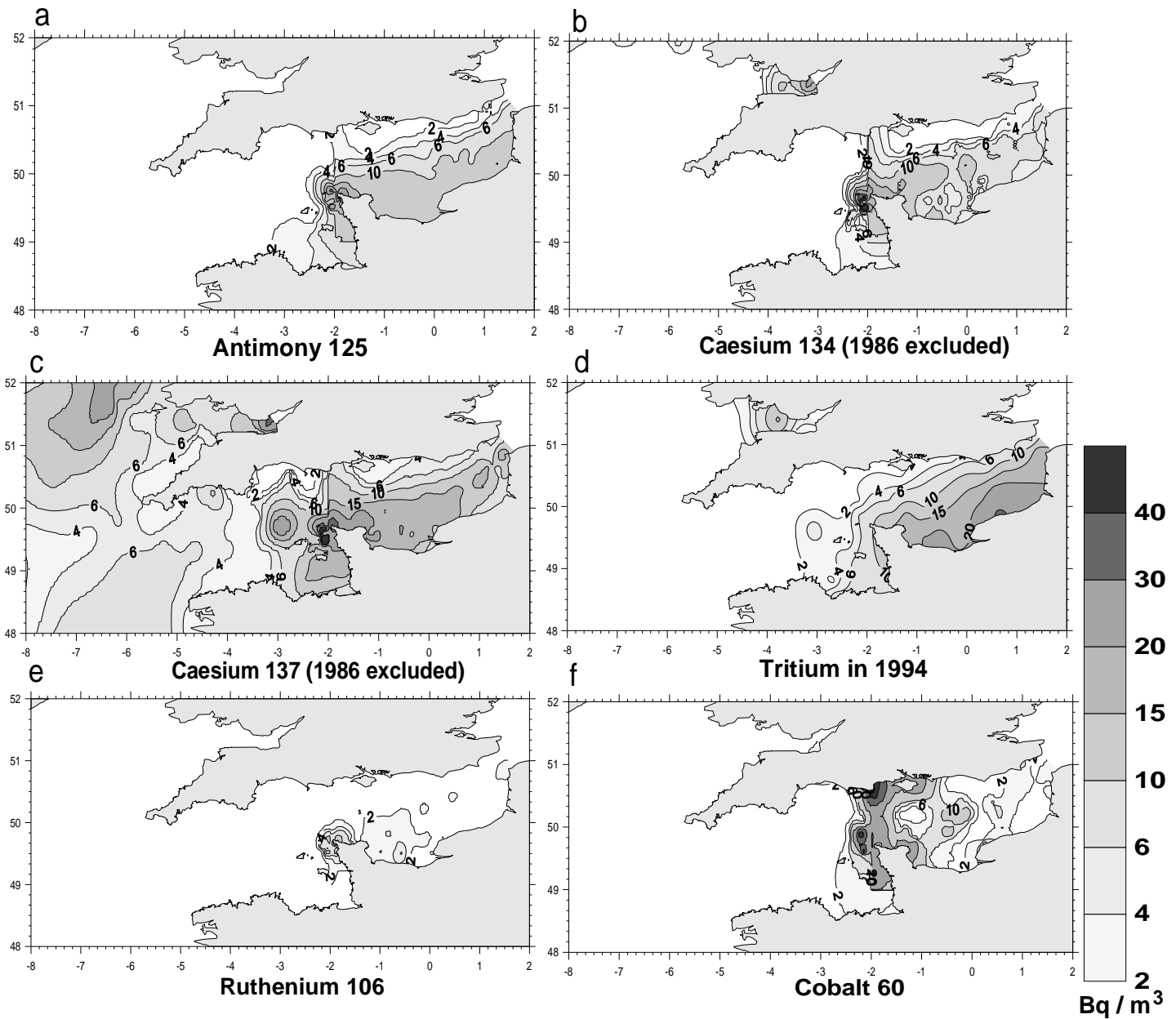


Figure 7 : Average impact of La Hague reprocessing plant in seawater corresponding to a constant release of 1MBq/s ; a : antimony 125, b : caesium 134, c : caesium 137, d : tritium, e : ruthenium 106, f : cobalt 60.

In order to be freed from random factors influencing the dispersion for a given moment (releases and meteorological variability) and to make different campaigns comparable, the distribution of activities measured at the time of a campaign were normalised to the average flow of the radionuclide during the six preceding months of releases from La Hague (this time interval corresponds to the average quantity of radioactivity originating from La Hague contained in the English Channel for a given moment, see above §3.3.). The obtained result, which only takes into account the average of 4 years could be refined in the future with assimilation of results from all the campaigns undertaken by the LERFA. These first results give a good approximation of the impact of the releases from La Hague in an average situation, while indicating the measured dissolved activity in the Channel corresponding to a constant flow of 1 MBq/s.

3.4.1. Average distribution of ^{125}Sb (Fig. 7a, 8a)

^{125}Sb is used as a reference conservative tracer originating exclusively from the La Hague plant. Since only a few years are taken into account, the influence of special conditions remains important, as in the case of the ^{125}Sb -enriched plume to the west of the Isle of Wight, observed only in 1994.

The mapped distribution of radiolabelled waters is closely similar to the pattern obtained by hydrodynamic modelling (Fig. 8b) and exhibit the average impact of la Hague plant in English Channel's waters.

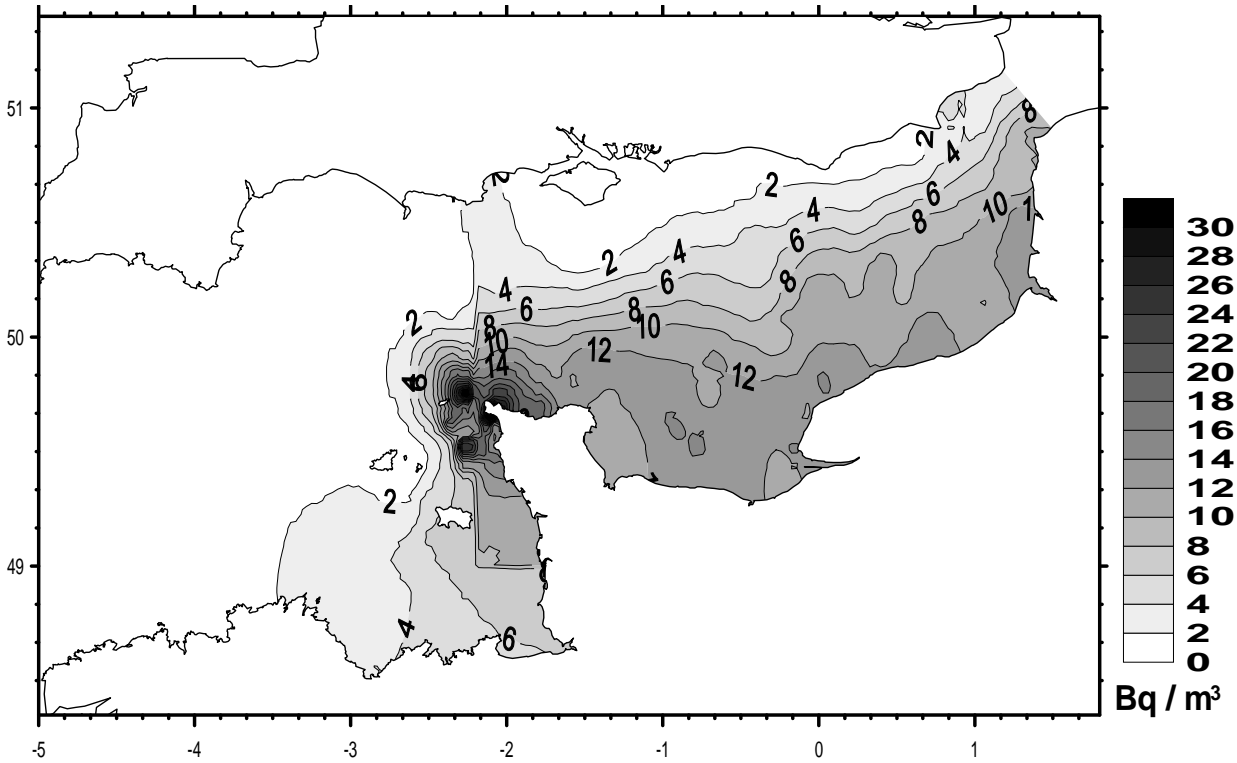


Figure 8 a : Average impact of la Hague reprocessing plant in the English Channel for antimony 125, with a constant release of 1MBq/s (deduced from campaigns of 1983, 1986, 1988 and 1994)

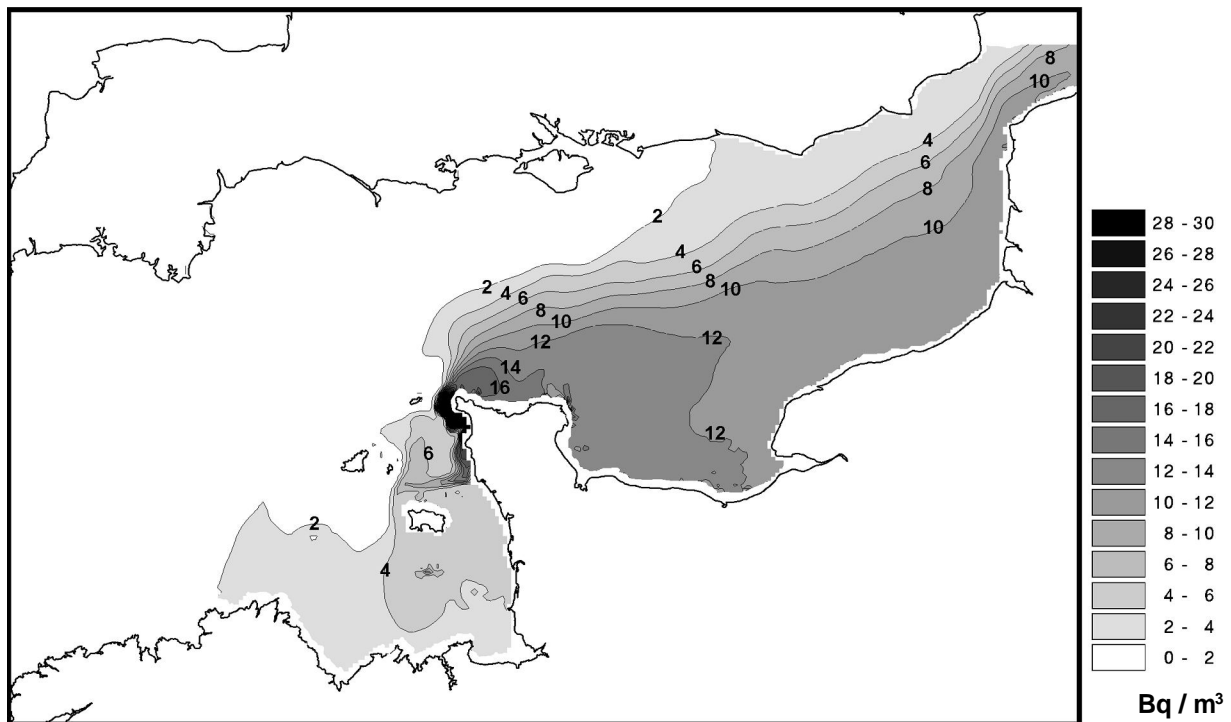
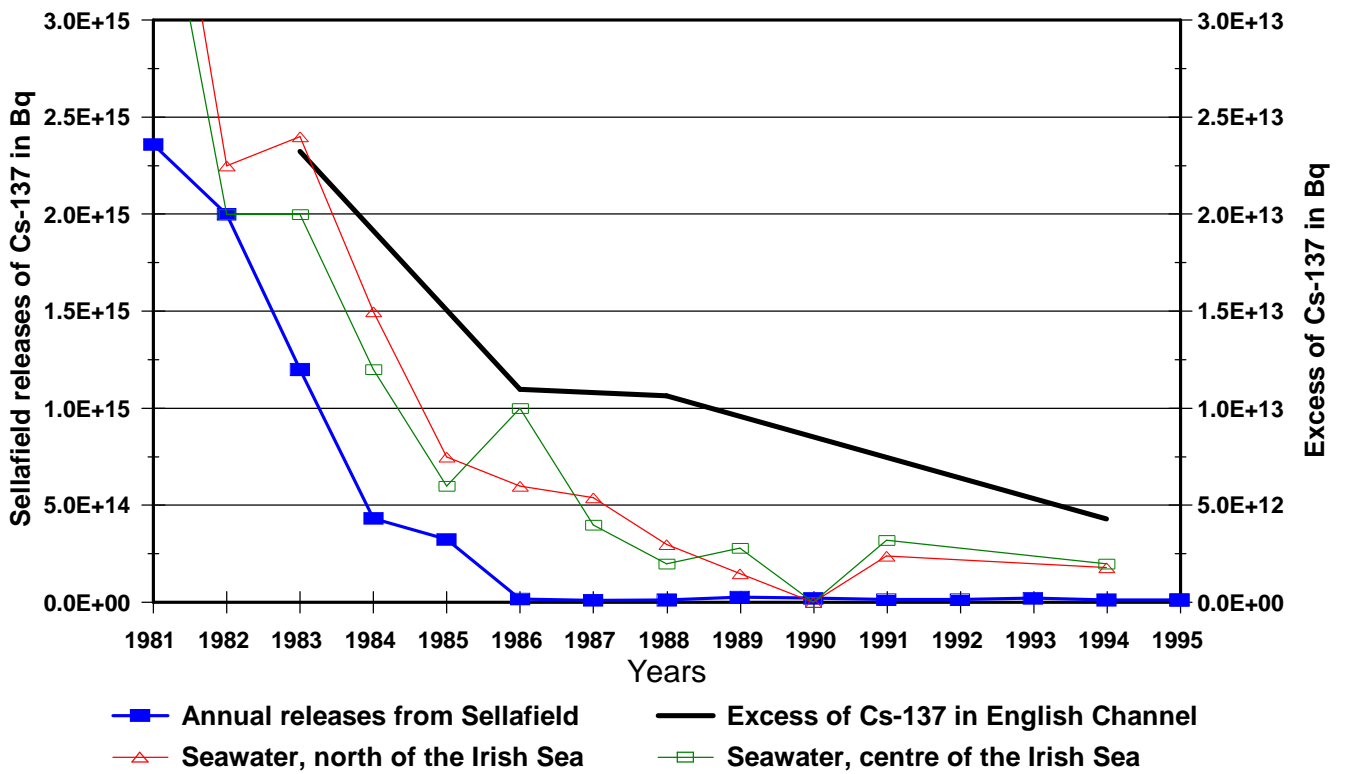


Figure 8 b : Model simulation of a constant release of 1MBq/s with a constant SW wind of 7m/s

3.4.2. Average distribution of ^{134}Cs (Fig. 7b)



The average distribution of ^{134}Cs is based on only two campaigns (the years 1986 and 1988 are excluded because of the Chernobyl input), but is nevertheless very similar to the distribution of ^{125}Sb , thus verifying the hypothesis that ^{134}Cs is globally conservative in the English Channel and that there is no other major source for this element in this sea. However, a decrease in ^{134}Cs radiolabelling along the French coast is seen that does not appear to affect ^{125}Sb levels. It is possible that a reduction of dissolved ^{134}Cs may be associated with a higher suspended load near the coast. table 6 give estimation of these losses, which would represent about 14% of ^{134}Cs at the scale of the English channel.

3.4.3. Average distribution of ^{137}Cs (Fig. 7c)

The average distribution of ^{137}Cs is relevant to three campaigns (1986 is excluded because of the Chernobyl contribution). If the plume of La Hague's releases is perfectly identifiable in the English Channel, and comparable in the East to the ^{134}Cs distribution, the level of the average impact of ^{137}Cs is distinctly superior to that of ^{125}Sb . Particularly, an impact west of Cotentin, in zones which are unaffected by the labelling of La Hague, is clearly perceptible. Since the background of Atlantic surface sea water was deducted (2.5 Bq/m^3), and the Chernobyl impact was excluded (1986), it would seem that another source of ^{137}Cs exists and labels waters entering in the English Channel in a nearly homogenous manner. This source is equivalent to the la Hague release during this period (cf. Table 6).

It is possible to quantify these excesses at the time of the campaigns whilst taking into account the known sources (La Hague releases, Atlantic background water, Chernobyl fallout deduced from the content of ^{134}Cs), supposing that ^{137}Cs is totally conservative. The calculated quantities will undoubtedly be a little under-estimated, because we observe, as for ^{134}Cs , losses in ^{137}Cs along the French coast that are not taken into account.

Figure 9 : Excess of ^{137}Cs found in the Channel compared with Sellafield releases

The temporal variation in excess ^{137}Cs activity within waters of the English Channel (Fig. 9), when compared with the pattern of release from Sellafield and the activities measured in the Irish Sea, reveals that :

1) the evolution is quite similar, notably with regard to levels measured in the centre and north of the Irish Sea. It has in fact been shown (Kershaw *et al.*, 1987 ; Condren *et al.*, 1996) that the

sediment appears as a secondary source for ^{137}Cs in the Irish Sea, attenuating the strong decrease of industrial releases ;

2) the excess of ^{137}Cs measured in the English Channel represents approximately 1% of the Sellafield releases. This gives a plausible magnitude for the flow of radioactivity leaving the Irish Sea towards the south and entering the English Channel. If the influence from Irish Sea is confirmed, the source-term would be the instantaneous releases from Sellafield, associated with re-dissolution from sediments of the Irish Sea.

3.4.4. Average distribution of ^3H (Fig. 7d)

Only 1994 data are available to establish the impact of ^3H coming from the La Hague plant, so the number of measurements are clearly fewer than in the case of the gamma emitters. Nevertheless, the observed distribution is almost identical to that for ^{125}Sb , both in terms of spatial distribution of the labelling, as well as the measured activities in waters away from the coast. Closer to the French coast, the labelling is significantly enhanced in the Baie de Seine and along the coast of the Pays de Caux (6000 Bq/m^3). These relatively higher levels could be explained by several ways :

- measurements carried out in the Seine river shows concentrations of ^3H in the same order of magnitude ($3000 - 12000\text{ Bq/m}^3$) in fresh waters ;

- releases of ^3H by coastal nuclear power plants (Palluel and Penly), represents only 1% of releases from La Hague. In view of the shallow water depths in this sector, the possibility of local labelling cannot be excluded ;

- variations of La Hague releases / or wind conditions.

It is not currently possible to decide the respective influence of each contribution, these first results required validation by further measurements.

3.4.5. Average distribution of ^{106}Ru (Fig. 7e)

The distribution of ^{106}Ru is indicative of non-conservative behaviour in seawater, where the element is adsorbed onto sediments and living organisms. Considering its relatively short half-life (one year), it is normal to observe a deficit, but the evaluated average transit time of about six months in the English Channel is insufficient to explain the losses of this radionuclide. In view of what we know about the long-term circulation of waters masses, the loss of ^{106}Ru appears proportional to the residence time in seawater. In particular, the more highly labelled areas in the middle of the English Channel are the result of quicker transit times than in coastal zones.

3.4.6. Average distribution of ^{60}Co (Fig. 7f)

The distribution of ^{60}Co is clearly different and shows two sources of comparable intensity in the English Channel : the La Hague reprocessing plant, and probably the Winfrith nuclear power plant. The attenuation of the ^{60}Co signal towards the east - as with ^{106}Ru - reveals the non-conservative behaviour of this element in seawater. The dispersion towards the east, more perturbed than in the case of ^{106}Ru , seems nevertheless proportional to the residence time in seawater. The low levels of measured activities, the associated analytical errors and the uncertainties concerning the extraction yield make it impossible to go any further in the interpretation of results for this element.

4. CONCLUSION

The new tools developed in this study (correction of sample locations with tide and interpolation methods) makes it possible to establish quantitative inventories of measured radioactivity and re-utilise previously obtained data (years 1983, 1986 and 1988) in the English Channel. A comparison of these results with known releases of artificial radionuclides into the sea provides constraints on the transit-times of water masses in the Channel, as well as an evaluation of losses of non-conservative radiotracers from seawater (table 6). The contribution from the Chernobyl fallout material is also estimated (Fig. 6, 13 TBq at the end of 1986).

The comparison of different yearly situations make it possible to represent the average impact of a source on the scale of the English Channel (activity in Bq/m³ obtained in seawater corresponding to a release of 1 MBq/s from La Hague reprocessing plant). This is carried for one element (¹²⁵Sb, Fig. 7a, 8a) showing conservative behaviour in seawater, while comparisons with other less conservative radiotracers (¹³⁷Cs, ¹³⁴Cs, ¹⁰⁶Ru and ⁶⁰Co, Fig. 7 b-f) show the extent of loss for the respective radionuclides, as well as the area concerned. Such results are of great interest for understanding the fixation of these tracers onto sediments. In other respects, ¹³⁷Cs exhibits an excess in its inventory for all the studied years, equivalent to the La Hague plant releases for this radionuclide. A comparison between this excess and Sellafield releases into the Irish Sea suggest a possible influence of this source in the English Channel, the excess of ¹³⁷Cs representing about 1% of the Sellafield releases. These data will serve as a basis for understanding long - term transport and the origins of water masses coming into the English Channel.

All the results obtained could be refined taking account of all campaigns carried out in the English Channel by IPSN and the different European laboratories over a period of twenty years. Nevertheless, the results are already directly comparable with numerical models (fig 8a, 8b), so confirming the efficiency of the tools used in this study. They provide more precise limits for refining the calibration of models, and serve as a basis for studying the fixation of tracers onto particulate matter, sediments and living organisms.

The information obtained using artificial radionuclides can generally be transposed to other substances present in seawater. It may also be applied to understanding the impact of industrial installations, as well as transport mechanisms in the marine environment under normal or accidental scenarios. Future studies will link model results more strongly with observations in the real environment, in order to specify and supplement the descriptions of geochemical and biological processes that govern the behaviour of radionuclides in marine ecosystems.

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