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**Natural and Artificial Radionuclides in the Study of Water Movements
in Coastal and Open Ocean Systems**

**IMPROVING KNOWLEDGE OF WATER-MASS CIRCULATION IN THE ENGLISH
CHANNEL USING RADIOACTIVE TRACERS.**

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Improving Knowledge of Water-Mass Circulation in the English Channel Using Radioactive Tracers

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Abstract : 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 , ^{125}Sb and ^3H in the English Channel and Irish Sea.

The re-utilization 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 would represent about 1% of Sellafield 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

New tools have been 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 characterized sources (Chernobyl accident fallout).

2. TECHNIQUES

2.1. Sampling and analysis

The data studied here originates from surface (3 m) sampling campaigns carried out in 1983, 1986, 1988 and 1994 in the English Channel and Irish Sea on board ships of the IFREMER, CNRS and MAFF (G.B.). Campaigns from 1994 onwards were carried out in the framework of the CCE MAST II FluxManche programme.

2.2.1. ^{125}Sb , ^{137}Cs , ^{134}Cs

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 [1].

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; measurement of ^3H by liquid scintillation was made after electrolytic enrichment by the CEA- SMSRB laboratory.

2.2. Data exploitation

The data are re-worked, by correcting the original positions according to the hour and tide at the moment of sampling, using instantaneous currents generate by modelization. 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. After repositionning, measurements were interpolated over the whole set of studied areas. These techniques allow a quantitative utilization of the point measurements in a sea with strong tide as the English Channel, and easier comparisons between the different campaigns.

3. RESULTS AND DISCUSSION

3.1. General description of radiotracer distribution (figs. 1 a-d)

Figures 1 give a good illustration of water mass circulation in 1994 on the scale of the Irish Sea and English Channel. The different radionuclides (^{137}Cs , ^3H , ^{134}Cs and ^{125}Sb) yield a mutually consistent set of measurements which follow the general distribution pattern recorded previously. The results for ^3H are of interest in this context, confirming that releases from La Hague and Sellafield can be readily distinguished from other source terms (background, rivers or rain) using this radionuclide.

The distribution of ^{137}Cs or ^3H (fig. 1a-b) provides a good overview of the circulation of water masses in the English Channel and the Irish Sea. Levels representative of Atlantic background increase slightly in the western approaches of the Channel ($2.9 - 4.1 \text{ Bq/m}^3$). The influence of La Hague releases extends from the centre of the Channel and along the French coast towards the east, with little influence in the north.

In the Irish Sea, the Sellafield impact is much more pronounced east the Isle of Man, with a strong decrease southward. South of lat. 51°N , activities become close to the background, but are generally higher than levels expected for normal Atlantic background (2.5 Bq/m^3 for ^{137}Cs , 300 Bq/m^3 for ^3H). The levels of ^{137}Cs are 10 to 100 times higher than those measured

in the English Channel (2000 - 50 Bq/m³ in 1983 ; 150 - 20 Bq/m³ in 1994). The labelling observed in this area is also more marked for other radionuclides (¹²⁵Sb, ³H, and ¹³⁴Cs fig. 1).

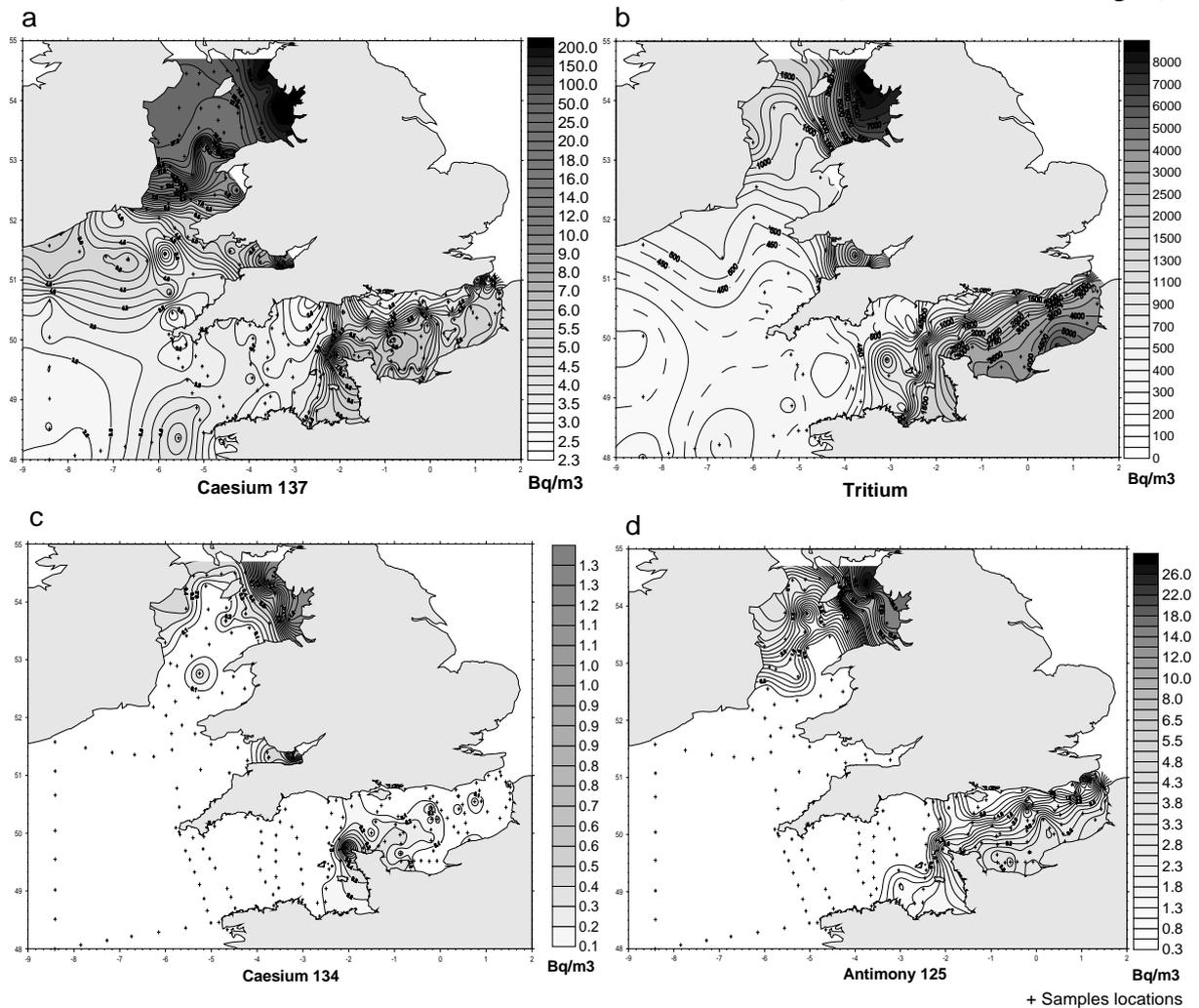


Figure 1 : GedyMac campaign, 28/08 - 13/09/1994, radioactivity in seawater

3.2. Fallout from the Chernobyl accident

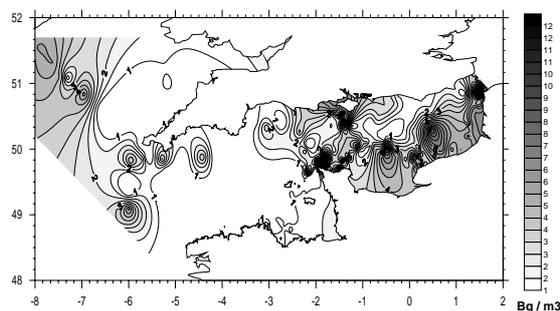


Figure 2 : ¹³⁴Caesium coming from Chernobyl accident in the English Channel at the end of 1986

Fallout material from the Chernobyl accident extended over all of Europe, and can be recognized by a diffuse labelling of surface 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 ratio of ¹³⁷Cs/¹³⁴Cs at the time of the accident and characteristics of other known sources (Atlantic waters background and La Hague's releases). Radiolabelling attributable to the presence of Chernobyl fallout in the English Channel is represented in figure 2. The levels observed in

Atlantic waters are probably due to direct atmospheric fallout, while the labelling in the eastern part of the Channel 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. In the English Channel, 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.

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 . The total inventory of radioactivity present has been calculated for all the measured radionuclides and all the campaigns. The quantities obtained 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 ^{125}Sb 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 remains 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 5 to 6 months of releases ;

Assuming that the flow is non-laminar and that all zones do not participate in the same manner to the flow, the average transit time of 4 - 5 months obtained by other methods ([2], [3], [4]) is compatible with these new data.

On the basis of these estimates, it is possible to evaluate the percentage of losses or excesses of radioactivity measured for the other radionuclides. It is nevertheless necessary to subtract all known sources in order to calculate the inventories precisely (Atlantic waters background, industrial releases and Chernobyl fallout).

Table 1 : Proportions of radioactivity in the English Channel coming from La Hague during 1983 to 1994 in comparison to ^{125}Sb .

equivalent release duration :	Whole English Channel 4702 Km ³ 32 weeks				Eastern English Channel 1576 Km ³ 25 weeks			
	^{137}Cs	^{134}Cs	^{125}Sb	^3H	^{137}Cs	^{134}Cs	^{125}Sb	^3H
Number of campaigns	3	2	3	1	5	3	5	1
Fraction of the La Hague release	231 %	78 %	98 %	101 %	139 %	78 %	96 %	119 %

3.4. Average circulation and fluctuations (figure 3)

In order to eliminate random factors influencing dispersion at a given moment (variability of releases and meteorological conditions), the activities measured during a campaign were normalized with respect to the average release of this radionuclide from La Hague during the six preceding months. These preliminary results give a good approximation of the impact of releases from La Hague in an average situation, while indicating the measured dissolved activity in Channel waters corresponding to a constant flow of 1 MBq/s.

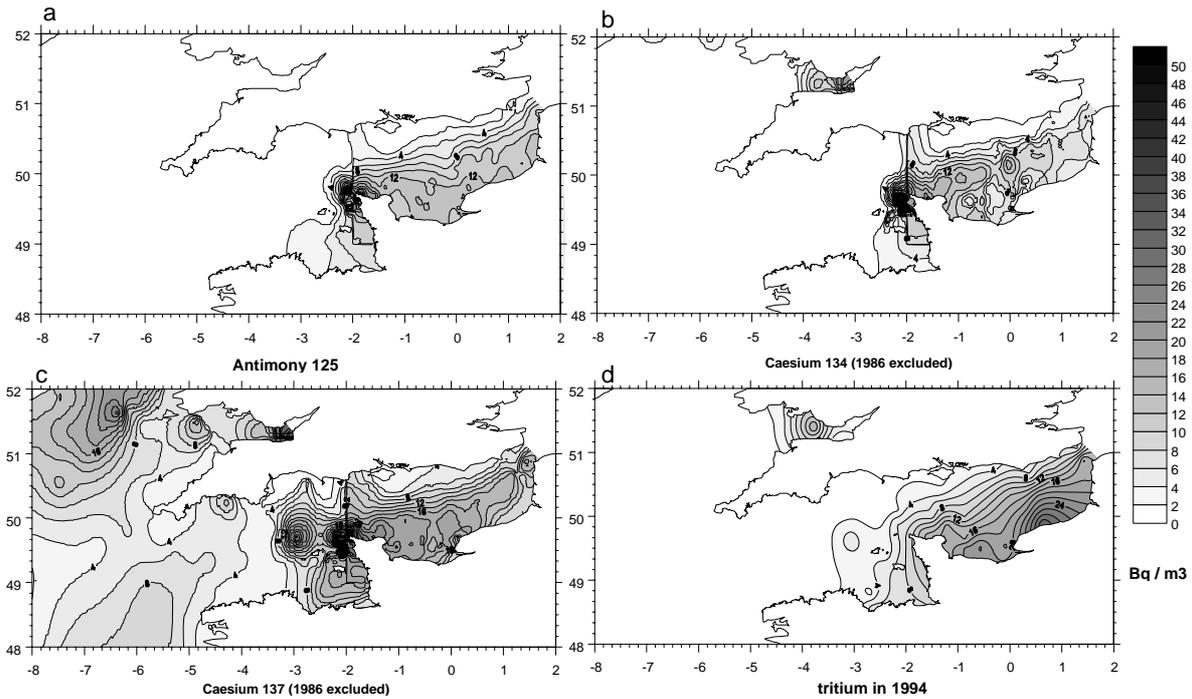


Figure 3 : Average impact of la Hague reprocessing plant in seawater for a constant release of 1 MBq/s

- Average distribution of ^{125}Sb (fig. 3a).

^{125}Sb is used as a reference conservative tracer originating exclusively from the La Hague plant. The mapped distribution of radiolabelled waters is closely similar to the pattern obtained by hydrodynamic modelling [5] and exhibits the average impact of the la Hague plant in English Channel waters.

- Average distribution of ^{134}Cs (fig. 3b).

The average distribution of ^{134}Cs is based on only two campaigns, but is nevertheless very similar to the distribution of ^{125}Sb , thus verifying the hypothesis that ^{134}Cs is overall 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 1 : about 20% of ^{134}Cs).

- Average distribution of ^{137}Cs (fig. 3c).

The average distribution of ^{137}Cs comprises data from four campaigns. Even though the plume of releases from La Hague is clearly identifiable in the English Channel, and comparable in the east to the ^{134}Cs -labelled plume, the average impact of ^{137}Cs is clearly higher than that of ^{125}Sb . This is especially the case for the clearly identifiable impact observed to the west of the Cotentin in Atlantic waters, an area unaffected by labelling from La Hague. Since the seawater background is taken into account (2.5 Bq/m^3), and the Chernobyl impact is excluded (year 1986), it may be inferred that another source of ^{137}Cs exists that labels waters entering the English Channel in a nearly homogenous manner. It is possible to quantify these excesses by making allowance for the known sources (releases from La Hague, Atlantic seawater background, Chernobyl fallout), while assuming that ^{137}Cs is totally conservative. Since it may be envisaged that English Channel waters are labelled by releases from Sellafield, we compared the evolution of this excess with the Sellafield releases, as well as the ^{137}Cs activities measured in the Irish Sea. The evolution obtained is quite similar, notably as regards the levels measured in the centre and the north of the Irish Sea. The excess of ^{137}Cs measured in the English Channel would represent about 1% of the Sellafield release. This would give a

plausible magnitude for the flux of radioactivity leaving the Irish Sea towards the south and penetrating into the English Channel.

- Average distribution of ^3H (fig. 3d).

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. Measurements carried out in the Seine river show only a weak contribution of ^3H in fresh waters, despite the releases of ^3H by coastal nuclear power plants (Palluel and Penly), representing 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. In any case, these first results required validation by further measurements.

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-utilize 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 1). The contribution from the Chernobyl fallout material is also estimated (fig. 2).

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/m^3 obtained in a seawater sample for a release of 1 MBq/s from La Hague reprocessing plant). This is carried for one element (^{125}Sb , fig. 3a) showing conservative behaviour in seawater, while comparisons with other less conservative radiotracers (^{137}Cs and ^{134}Cs , fig. 3b-d) show the extent of loss for the respective radionuclides, as well as the area concerned. Such results could be of great interest for understanding the fixation of these tracers onto sediments. In other respects, ^{137}Cs exhibits an excess in its inventory for all the studied years. 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 ^{137}Cs 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 other radionuclides and 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, they provide more precise limits for refining their calibration, and serve as a basis for studying the fixation of tracers onto particulate matter, sediments and living organisms.

Références

- [1] Gandon R., Guéguéniat P., "Preconcentration of ^{125}Sb onto MnO_2 from Seawater Samples for Gamma-ray Spectrometric Analysis", *Radiochimica Acta* (1992) 57, pp. 159-164.
- [2] Guéguéniat P., Salomon J.C., Wartel M., Cabioch L., Fraizier A., "Transfer Pathways and Transit Time of Dissolved Matter in the Eastern English Channel Indicated by Space-Time Radiotracers Measurement and Hydrodynamic Modelling", *Estuarine, Coastal and Shelf Science* (1993) 36, pp.477-494.

- [3] Bailly du Bois P., Salomon J.C., Gandon R., Guéguéniat P., "A quantitative estimate of English Channel water fluxes into the North Sea from 1987 to 1992 based on radiotracer distribution", *Journal of Marine Systems* (1995) Vol. 6 N° 5-6 pp 457-481.
- [4] Salomon J.C., Breton M., Guéguéniat P., "Computed residual flow through the Dover Strait", *Oceanologica Acta* (1993) 16, 5-6, pp. 449-455.
- [5] Salomon J.C., Guéguéniat P., Breton M., "Mathematical model of ¹²⁵Sb transport and dispersion in the Channel", In : *Radionuclides in the study of marine processes*. Norwich, UK, 10-13 september 1991, Ed. Kershaw J.P., Woodhead D.S. Elsevier Applied Science (1991) pp.74-83.