



HAL
open science

Flux of tritium from the sea to the atmosphere around a nuclear reprocessing plant: Experimental measurements and modelling for the Western English channel

O. Connan, P. Bailly Du Bois, L. Solier, Didier Hebert, C. Voiseux

► To cite this version:

O. Connan, P. Bailly Du Bois, L. Solier, Didier Hebert, C. Voiseux. Flux of tritium from the sea to the atmosphere around a nuclear reprocessing plant: Experimental measurements and modelling for the Western English channel. *Journal of Environmental Radioactivity*, 2023, 257, pp.107068. 10.1016/j.jenvrad.2022.107068 . hal-03859893

HAL Id: hal-03859893

<https://hal.science/hal-03859893>

Submitted on 21 Nov 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License

Flux of tritium from the sea to the atmosphere around a nuclear reprocessing plant: Experimental measurements and modelling for the Western English Channel.

⁽¹⁾Connan O., ^(1,2)Bailly du Bois P., ⁽¹⁾Solier L., ⁽¹⁾Hebert D., ⁽¹⁾Voiseux C

⁽¹⁾Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-ENV/SRTE/LRC, BP 10, rue Max Pol Fouchet, 50130 Cherbourg-En-Cotentin, France.

⁽²⁾LUSAC-Intechmer, Conservatoire National des Arts et Métiers, Boulevard de Collignon, Tourlaville, 50110, Cherbourg-en-Cotentin, France.

* Corresponding author:

[E-mail adress: olivier.connan@irsn.fr](mailto:olivier.connan@irsn.fr)

IRSN

Cherbourg-En-Cotentin Radioecology Laboratory

Rue Max Pol Fouchet

BP10

50130 Cherbourg En Cotentin

Tel (33) 2 33 01 41 08

Abstract

Tritium is a radionuclide released to the atmosphere and in rivers or sea by nuclear industries in various forms, mainly HTO. In general, in impact studies leading to estimated doses for the on-land population, only atmospheric discharges are taken into consideration for tritium. However, the tritium released in liquid environments can, under some conditions, be transferred to the atmosphere by evaporation and then be transported to terrestrial ecosystems by wind. This study was carried out in north west France, near to a fuel reprocessing plant (RP) at La Hague, which discharges tritium into the western English Channel. Firstly, we highlighted the influence of a mass of water enriched with tritium on the measurable HTO activity levels in atmospheric water vapour downwind of this mass of water by using 18 field campaigns. Secondly, a hydrodynamic model able to simulate tritium activity in the water of the western English Channel was coupled with an evaporation model and a Gaussian type atmospheric transport model. It allows to reconstitute variations in atmospheric tritium activity on the coast, depending on actual liquid discharges of tritium from the fuel reprocessing plant in La Hague (RP) and weather conditions. On this basis, when a mass of water containing approximately 20 to 100 Bq.L⁻¹ of tritium flows between 0 and 10 km off the coast, variations in atmospheric activity measured onshore can increase of 2 to 15 Bq.L⁻¹ downwind. Study data was then used to estimate in the western English Channel (~21,000 km²) including the La Hague region, the Normand-Breton Gulf (NBG) and the Bay of Seine (BS). Mean tritium quantities released by the sea into the atmosphere reached 130 TBq.y⁻¹ over the 2017-2020 period, with emissions varying in function of the geographical area. These fluxes depends principally on the distance from the liquid discharge point, and therefore mean tritium activity in seawater and local current hydrodynamics emissions, were estimated at 0.9 - 11.3 GBq km⁻².y⁻¹. If we compare the "marine" source term, in HTO form, with the direct source term for gaseous discharges from the RP into the atmosphere, the marine source term is one order of magnitude greater for the entire marine region affected by liquid discharges. Finally, thanks to these works, we were able to estimate that approximately 1.1% of tritium stock discharged at sea (liquid discharge regulated and controlled) return to the atmosphere each year, at the scale of the English Channel. Although tritium is negligible in terms of dose impact, it may be necessary to take into consideration the quantities of tritium released by the sea and then transported to land in order to improve the precision of impact studies. Such assessment is subject to the availability of an atmospheric transport model able to integrate

the surface source and differentiate between tritium transported to land and tritium which remains diluted in the atmosphere labelled above the sea.

Keywords: Tritium, Ocean-atmosphere interface, Flux, Marine source term, marine dispersion, atmospheric dispersion.

1. Introduction

Tritium (^3H) is a low energy β -emitting radionuclide with a half-life of 12.32 years. Tritium is produced by naturally-occurring processes through the interaction of atmospheric gases and cosmic radiation. Natural abundance is estimated at about 4.5 kg of tritium. It has mostly been incorporated in the oceans since the late 1950's and the early 1960's by fallout from the atmospheric nuclear tests with an estimated atmospheric input of about 520–550 kg of tritium (Michel, 1976; UNSCEAR, 2000, Oms et al., 2019). About 99% of atmospheric tritium is in the form of tritiated water molecules (HTO) and thus follows the water cycle. In addition, tritium is actually mainly produced by the nuclear industry, and tritium is one of the major radionuclides discharged into the the atmosphere, rivers and seas in terms of quantity. Many nuclear facilities worldwide are located near to oceans and discharge various radionuclides with government authorisation. Such radionuclides include tritium, either directly at sea via a discharge pipe, generally a few km from the coast, or via rivers that lead to the oceans and can result in tritium labelling (Oms, 2018). Industrial discharges of gaseous and liquid tritium worldwide are thus estimated at 11.7 PBq.y^{-1} and 16 PBq.y^{-1} respectively (UNSCEAR 2016). In France, liquid discharges from nuclear powerplants (mainly into rivers) represent approximately 1 PBq.y^{-1} and liquid discharges from the La Hague reprocessing plant represent roughly 11 PBq.y^{-1} . In addition to existing nuclear facilities worldwide (436 units in 2013, Zeng et al, 2016), several EPR type reactors are currently under construction on coasts in Finland, France, the United Kingdom and China. Other traditional nuclear facilities are under construction, particularly in China where 26 units are currently taking shape over the entire length of the east coast of China (Zeng et al, 2016). Tritium as HTO cannot be eliminated in the decontamination process, therefore tritium discharges are not likely to decrease in the future.

Excluding normal operations leading to declared and controlled discharges, nuclear accidents such as at Fukushima Daiichi Nuclear Power Plant (FDNPP) can occur to accidentally inputs of radionuclides into the oceans. At Fukushima, the quantities of ^3H discharged into the Pacific Ocean were estimated at between 0.1 and 0.5 PBq (Povinec et al., 2013). When considering other potential sources of tritium contamination in the event of an accident, it is worth remembering that a growing number of nuclear submarines and vessels are currently sweeping across the oceans (between 150 and 200 according to current estimates <http://www.world-nuclear.org>) implying a potential, although limited, risk. We could also report the launch of the first floating nuclear reactor in Russia in May 2020.

Oceans and seas, as well as estuaries and rivers, represent a reservoir of anthropic substances including radionuclides, which could, given specific transfer conditions at the interface, be transported to land via spray and aerosols, or via simple exchange at the ocean-atmosphere interface and atmospheric transport by wind (Maro et al, 2005; Sheppard et al., 2006; Upstill-Goddard, 2006; Marang *et al et al* 2011).

Ocean-atmosphere exchanges of tritium were recently considered in studies (Nie et al, 2020, 2021, Zhao et al., 2021), particularly aiming to estimate the tritium evaporation from a liquid surface. The objective is to determine if this source term, operating via emissions from a liquid reservoir such as sea, should be taken into consideration to better estimate the dose absorbed by people on land. This is true even if tritium is clearly not a particularly high-impact radionuclide in terms of dose, thanks to a low dose factor (1.8×10^{-11} Sv/Bq, ICRP, 2012). These questions on how much tritium in the sea evaporates to the atmosphere are also partially relevant for understanding the Fukushima configuration, where 1000 tanks containing ≈ 1 million tons of water contaminated principally with tritium are currently stored after the accident. The quantities of tritium at Fukushima are estimated at 1 PBq (TEPCO, 2020), and the plan is to discharge this tritium into the Pacific Ocean in the coming years. Consequently, the ability to predict the behaviour and fate of this tritium will become a critical feature for both understanding and controversies (Buessler, 2020; Zhao *et al et al* 2021). Detailed knowledge of the behaviour of tritium dissolved at sea and the percentage released to the atmosphere is therefore required. This information will help to knowledge this transfer to populations in coastal areas, and to avoid exclusively considering the ingestion of aquatic products when calculating doses (Nie et al, 2021).

The aim of our work is to check the impact of a mass of tritiated water on nearby atmospheric activity, and the percentage of liquid discharge evacuated to the atmosphere. We focused on a suitable study area near to the Orano La Hague fuel reprocessing plant, in northwest France (hereafter RP). We launched experimental campaigns in the immediate proximity of the offshore discharge point for this reason. Orano RP discharges large quantities of tritium in a controlled manner into the English Channel, in the north part of the Normand-Breton Gulf (hereafter NBG) 1 km from the coast. To begin with, we checked that the mass of seawater flowing near to the coast, and influenced by the tritium discharged from RP, can lead to higher terrestrial HTO activity levels in atmospheric water vapour after tritium exchanges at the water-air interface and transport in a landward direction. G. Bacon (Bacon *et al.*, 2011) shows the first indications of this influence. This aspect was checked by taking experimental

measurements on the coast, downwind from the sea as the tritiated water plume passes through. In a second step, after having checked that this impact is real, we developed a model coupling a hydrodynamic model (Mars-2D; Bailly du Bois et al, 2012, 2020b; Fievet et al, 2020), which can be used to simulate tritium activity in seawater throughout the region, with an evaporation model which can be used to calculate a marine source term. We applied a Gaussian atmospheric transport model to the calculated source term in order to attempt to reproduce the activity measured of HTO induced on land by the mass of water, depending on weather conditions. Finally, we estimated the quantities of tritium over 2017-2020 period, which could be re-released considering a marine surface including the Normand-Breton Gulf (NBG) and the Bay of Seine (BS), which covers several thousand km² of the Western English Channel clearly affected by tritium. The importance of this "secondary" marine source term for the atmosphere will be discussed in comparison with "direct" gaseous emissions from the Orano RP.

2. Materials and methods

2.1 Study Area

The Orano reprocessing plant (RP) at La Hague is located in north west France, at the north west tip of Cotentin Peninsula, facing the western English Channel (Fig 1). The La Hague region is located between the Normand-Breton Gulf (NBG) to the west and the Bay of Seine (BS) to the east, in the center of the English Channel. The offshore discharge point for liquid effluents is located 1 km from the coast in the north part of the NBG (Fig 1). The Flamanville nuclear power plant operated by EDF is also located in this region on the west coast of the Cotentin peninsula, but its liquid radioactive discharges are much smaller than those of the RP (two orders of magnitude smaller). Thanks to the many studies completed on the dispersal of radionuclides in this region, the hydrodynamics of the area are well known and a model (Mars 2D, Model for Applications at Regional Scale, described below) was especially adapted for this region, with multiple experimental campaigns used for its validation (Bailly Du Bois et al., 1999; 2005; 2012; 2020a). The long-term dispersal of dissolved material in seawater in these areas is very complex, principally in the NBG due to the presence of the Channel Islands. However many studies have been completed by the French Institute for Radiological Protection and Nuclear Safety (IRSN) and have contributed precise details of the dispersion of tritium and other radionuclides in this region (Bailly du Bois et al., 1995; 2010; 2020b; Fievet et al, 2020). The hydrodynamics and transport of water masses due to residual currents are

described in the works of Salomon et al., (1991), Bailly du bois et al, (2012), Fievet *et al et al* (2020) (supplementary material A1). Globally, if we consider a period of time of approximately one year, the Normand-Breton Gulf area most affected by discharges is the central and eastern part of the NBG, running from Cap de Carteret up to Cap Fréhel, including the island of Jersey (zone located between -1.6 and -2.4°W , and to the south of 49.3°N , Figure 1). Our supplementary material A2 includes a description of mean tritium activity in seawater in the NBG for the years 2014 and 2015 for real weather conditions and discharges (Fievet et al.,2020). However, over the long term, radionuclides released by the RP mainly disperse to the northwest, by crossing the BS from west to east (Salomon et al., 1991), with a gradient which decreases in proportion to distance from the French coast towards the British coasts. On this basis, a water mass would take 110 to 152 days to transit from Cap de la Hague to the Strait of Dover (Bailly du Bois et al, 1995). For more details on the general circulation of water masses in the English Channel, refer to Bailly du Bois and Dumas (2005).

2.2 General method, objective and means

Details of the general method used for this study, mentioned in the introduction and adapted from Bacon et al., 2011, are confirmed below and sum up in the Figure 2.

a) Obtain coastal measurements of tritium in atmospheric water vapour, each hour, for approx. 6 to 9 hours, when a sea breeze is blowing, and ideally when a liquid discharge was released by the RP.. Tritium is also measured in coastal seawater simultaneously to meteorological measurements.

b) Hydrodynamic model simulation of tritium activity in seawater around the discharge point using the Mars-2D model, for different geographic zones downwind (Fig. 3), for these experimental campaign days (over an area covering roughly a hundred km).

c) Considering meteorological parameters, seawater temperatures and salinity, and tritium activities simulated in seawater, calculation of water evaporation (and therefore tritium evaporation) from seawater, and the quantity of tritium released to the atmosphere.

d) Estimate the "source" geographic zone based on wind direction when taking measurements on the coast, and when the source term is known (calculated according to the protocol under point c), apply a surface Atmospheric Transfer Coefficient (ATC) in order to ultimately

estimate the atmospheric HTO activity induced by the seawater (considering atmospheric water content).

e) Compare this induced HTO activity as calculated with the experimental HTO measurement taken on the coast.

f) Validate the previous model, then extrapolate the tritium marine source term reaching the atmosphere at the scale of the Bay of Seine and the Normand-Breton Gulf (zones influenced by the tritiated liquid discharges from the Orano RP). This is done by considering actual meteorological conditions to calculate evaporation, and tritium activity simulated in seawater. Run this process over several years (2017-2020), to take into account meteorological variations and varying discharge levels from year to year.

g) Compare the secondary source term with direct atmospheric emissions of gaseous HTO via the Orano RP stacks and estimate volumes.

2.3 Tritium discharges from the Orano reprocessing plant of La Hague

The quantities of tritium discharged in liquid and gaseous form by Orano RP La Hague are published annually (<https://www.orano.group>). Liquid effluents are intermittently discharged at sea via a pipe located 1 km from the coast (Fig. 1), and can last between several minutes and several hours, while both volumes and radionuclide activity vary. The number of monthly discharges has varied from 36 to 50 over the last 10 years. Discharges are mainly scheduled over a set period depending on tide times to promote optimal hydrodynamic dispersal towards the northern and eastern English Channel (Bailly du bois et al, 2012). Over the 2017-2020 period, the mean annual liquid discharge level for tritium was equal to $11,975 \text{ TBq.y}^{-1}$ with a minimum of $11,400 \text{ TBq.y}^{-1}$ (in 2018 and 2020) and a maximum of $13,200 \text{ TBq.y}^{-1}$ (in 2019). Each individual discharge of tritium to seawater from the La Hague reprocessing plant was transmitted by Orano for the period from 2016 to 2020. The quantity discharged and the date/time of the start and end of each discharge are given. Details of these releases are given in Bailly du Bois et al, 2020.

2.4 Sampling campaigns

We ran 18 air sampling campaigns, each over one day, in order to measure tritium in atmospheric water vapour at regular intervals for 6 to 8 hours, downwind from the sea breeze. These campaigns were run at the Goury Semaphore (Fig. 1) when the sea breeze was active from sea to land, and during periods when liquid discharges containing tritium were likely to occur from the Orano RP. 6 campaigns had been run in 2011 and 12 were run between 2018 and 2021.

Atmospheric water vapour was sampled using SDEC H3R7000 systems. The H3R7000 system condenses the atmospheric water vapour over a 30-min. period and measures the humidity as $\text{g}\cdot\text{m}^{-3}$ of H_2O in air (less than 10% accuracy). The condensed water is recovered for liquid scintillation analysis at a later stage. After sampling, the water was stored in darkness at 4°C prior to analysis. For analysis, a volume of 10 cm^3 of collected water (8 cm^3 for seawater) was placed in a vial with 10 cm^3 of liquid scintillation cocktail Ultima gold LLT (12 cm^3 for seawater). The samples activity was measured by liquid scintillation counting (TRI-CARB 3180 TRSL, PerkinElmer) for 16 h with a detection limit of $\sim 1\text{ Bq}\cdot\text{L}^{-1}$.

The experimental results of these campaigns, HTO measurements in atmospheric water vapour in $\text{Bq}\cdot\text{L}^{-1}$ on the coast, were then compared with the results of the full modelling process. Details of experiment days can be found in table 1.

3. Modelling description

Fig. 2 gives an overview of the modelling process used in this work to obtain the tritium activity induced by the evaporation of tritium dispersed at sea after liquid discharges into atmospheric water vapour.

The process used for this work integrates various modules used to ultimately obtain HTO activity in atmospheric water vapour at a given terrestrial location:

- A tritium hydrodynamic dispersal model for seawater (Mars-2D);
- A water evaporation model (Rohwer's model);
- Consideration of the surface area in m^2 ;
- An Atmospheric Transfer Coefficient (ATC) to "transport" the source term obtained towards land;
- Calculate atmospheric water content.

These different modules are described below. The general equation corresponds to equation 1

$$[\text{HTO}]_{\text{air}} = \frac{\text{HTO}_{\text{sea}} \times \text{Evaporation} \times \text{Surface} \times \text{ATC}}{\text{H}_2\text{O}_{\text{air}}} \quad (\text{eq. 1})$$

With

[HTO]_{air} : activity of tritium in water vapour of atmosphere in Bq.L-1

[HTO]_{sea} : activity of tritium in seawater in Bq.L-1

Evaporation : in L.m⁻².s⁻¹

Surface : in m²

ATC : Atmospheric Transfert Coefficient in s.m⁻³

H₂O_{air} : quantité of water in air in L.m⁻³

3.1 Hydrodynamics modelling – Mars 2D

Radionuclide activity concentrations in seawater were calculated using the Mars-2D model described in Bailly du Bois and Dumas (2005), Bailly du Bois et al. (2012, 2020b), Fievet *et al et al* (2020). The Mars-2D model uses two-dimensional horizontal approximation (i.e. shallow-water equations) to predict dissolved-substance transport in the English Channel. Full details concerning the algorithm are given by Lazure and Dumas (2008). In this study, the model used involved a nesting strategy. The first model (0 rank) starting from a broad region covering the entire NorthWest European continental shelf (with a 5.6 km grid resolution, 40-65°N, 15°E -20°W) down to a detailed domain covering the whole English Channel with a mesh size of 1500 m (rank 1, 46.84-52.34°N, 4.75°E -7.58°W) and down to two final domain with a mesh size of 400 m for the Hague area (rank 2, model named Gnbb, 48.46-50.01°N - 0.99 -2.74°W) or with a mesh size of 500 m for the Normand Breton Gulf and the Bay of Seine (rank 2, model named Ma05, 48.47-50.95°N, 1.79°E -4.34°W).

Mars-2D incorporated actual 3H discharges, tidal and meteorological forcing to simulate instantaneous currents and tritium dispersion. Real atmospheric pressure and wind data were

provided by Météo France (ARPEGE data). Tidal conditions were provided according to fes2004 (Lyard et al., 2006). Bathymetric data were estimated from various sources using the method described in Bailly du Bois (2011). The English Channel is shallow, therefore the concentration is already homogeneous in the water column as little as 1500 m from the outlet (Bailly du Bois et al., 2020b), which supports the use of a 2D model. The model was extensively validated on a short to long-term basis by comparing models/measurements as described in Bailly du Bois and Dumas (2005), Bailly du Bois et al. (2012, 2020b), Bailly du Bois (2014). Individual tritium discharges to seawater from the La Hague reprocessing plant were transmitted by Orano for the period from 2016 to 2020.

The rank 2-Gnbb model was used to compare data directly with our experimental campaigns performed at Goury Semaphore, as this model was specifically configured for this zone; calculations were run over a timescale of approximately one day which is sufficient to account for the more recent and concentrated plumes. The Ma05 model with the largest scope was used to calculate the source term in the NBG and BS, and calculations were run over a 5-year period (2016-2020). In terms of the precision and robustness of the model, supported by IRSN works for many years, data are known to be good quality, particularly when working on quantities of radionuclides (stock global of ^3H in a given geographic area). On this basis, Bailly du Bois *et al et al* (2020b) report a mean deviation between the modelled values and those deduced from measurements taken during oceanographical campaigns as systematically less than 10% for stocks of radionuclides in the English Channel and in the south of the North Sea. Bailly Du Bois and Dumas (2005) report a 1% difference between overall quantities of radionuclides calculated and measured for six campaigns completed between 1988 and 1994 at the scale of the English Channel and the North Sea. In terms of concentrations at each specific point, with the model covering the western English Channel, the mean deviation between individual concentrations was estimated as 6% (with standard deviation of 44%) (Bailly du Bois, 2014).

For the purposes of comparing models/measurements for experimental campaign days, tritium mean activities in seawater were calculated every 30 minutes in each upwind zone with a potential impact (50 possible zones for each calculation). 120 zones were predefined with 30° cones, at distances from the coast of 1, 3, 5 and 10 km respectively (Figure 3). Zones between 1-5 km, 3-5 km or 1-10 km, to give examples, were differentiated in order to obtain mean tritium activities for each zone, and identify a footprint zone matching our terrestrial measurements as best possible (see results section).

Then, as one of the objectives of this work was to estimate the quantities of tritium released from the marine area to the atmosphere, we focused on the region encompassing the Normand-Breton Gulf (NBG) and Bay of Seine (BS), which are the more affected by tritium discharges from the Orano La Hague. For this purpose, we calculated variation in tritium activity over time in seawater in various zones identified from 1 to 21 (Figure 4), from 01/01/2016 to 31/12/2020, every 3 hours. Data were used from 2017 (an equilibrium state was achieved in 2016 for tritium activity throughout the zone, Bailly du Bois et al., 2018).

3.2 Tritium evaporation model

Many equations can be used to calculate an evaporation rate (Singh and Xu, 1997; Xu and Singh, 2001; 2002). The methods used to determine evaporation can be separated into several categories, primarily empirical, water budget, energy budget and mass transfer, but the mass-transfer methods are generally easiest to use and the only practical method available (Singh and Xu, 1997). The mass transfer method is based on Dalton's law. The formula can be written as per equation 2.

$$E(\text{H}_2\text{O}) = f(u) (e_s - e_a) \quad (\text{Eq. 2})$$

Where E is free water surface evaporation, e_s is the saturation vapour pressure at the temperature of the water surface, e_a is the saturation vapour pressure at the dew point temperature and $f(u)$ is the wind dependant function. Many formulas with different $f(u)$ have been derived based on this approach.

We used a formula taken from the works of Rohwer (Rohwer, 1931). The evaporation rate obtained corresponds to water flux evaporated from the sea surface to the atmosphere, in $\text{L}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, and for our study, this water flux will carry ^3H activity ($\text{Bq}\cdot\text{L}^{-1}$) as calculated according to the description in section 3.1.

The following formula is used

$$E(\text{H}_2\text{O}) = 0.372 (1 - 0.000374 \text{ Pa}) (1 + 0.6 u) (e_s - e_a) \quad (\text{Eq. 3})$$

where:

$E(H_2O)$: Water vapour flux ($L.m^{-2}.d^{-1}$),

P_a : Atmospheric pressure (mbar),

u : Wind speed ($m.s^{-1}$),

e_s : Saturated vapour pressure at the sea surface (mbar),

e_a : Atmospheric vapour pressure above the water (mbar),

At the water surface, the air is saturated, and vapour pressure (e_s) can be calculated using the formula given by Schmidt (1989) :

$$\ln(e_s/p_c) = (T_c / T) \times (a_1 \cdot \tau + a_2 \cdot \tau^{1.5} + a_3 \cdot \tau^3 + a_4 \cdot \tau^{3.5} + a_5 \cdot \tau^4 + a_6 \cdot \tau^{7.5}) \quad (\text{Eq. 4})$$

where e_s is the partial pressure of atmospheric water vapour at saturation (bar), $p_c = 220.64$ bars,

T is air temperature in K, $T_c = 647.14$ K, $\theta = T/T_c$, $\tau = 1 - \theta$.

Constants: $a_1 = -7.85823$, $a_2 = +1.83991$, $a_3 = -11.7811$, $a_4 = +22.6705$, $a_5 = -15.9393$, $a_6 = +1.77516$.

P_a from equation 1 (partial pressure of atmospheric water vapour at humidity U) is deduced from the equation provided by Triplet et Roche (1986):

$$P_a = e_s U / 100 \quad (\text{Eq. 5})$$

where U is relative humidity as a %, and e_s is the saturated vapour pressure of water as calculated previously.

According to Singh and Xu (1997), the different equations which can be used to calculate evaporation give similar results, which are satisfactory on the scale of monthly evaporation data.

3.3 Atmospheric transport of the source term

Once the marine source term (quantity of tritium released in Bq. s^{-1} per unit of surface area in km^2), is calculated as described previously from ^3H activity in water (section 3.1) and evaporation (section 3.2), we applied a surface atmospheric transfer coefficient (ATC) to this source term to reflect its transport landwards. This ATC will mainly depend on weather conditions (stability) and the distance between the source term and land. The ATC is given in s.m^{-3} and is the ratio between an activity measurement at a given point (a tritium activity in Bq.m^{-3} of air on land in our case) and the initial source term (Bq.s^{-1}). In other words, the ATC is a dilution factor relating to atmospheric transport. As the marine emission source term for tritium and this ATC are known, we can deduce the induced atmospheric HTO activity in Bq.m^{-3} at a distant point as per equation 6

$$[\text{HTO}]_{\text{land}} (\text{Bq.m}^{-3}) = [\text{HTO}]_{\text{sea evaporate}} (\text{Bq.s}^{-1}) \times \text{ATC} (\text{s.m}^{-3}) \quad (\text{Eq. 6})$$

As is the case for evaporation, many options exist for calculating the ATC. The ATC is based on Gaussian models, which represent a simplified solution to of ? the diffusion transport equation, which describes spatial variation in the concentration of a pollutant over distance, in the event of constant release under uniform meteorological conditions. The use of a Gaussian model requires the standard deviation of the horizontal and/or vertical dispersion of the plume to be determined. For first generation Gaussian models, such as the models of Pasquill (1961), Doury (1981) and Briggs (1985), the standard deviations for dispersion were determined from experimental campaigns.

We opted for the Pasquill equation which is still widely used in the atmospheric domain. In our work, we defined "mean" ATC based on distance between the source zone (the sea) and land. We applied a different mean ATC depending on distance of source (1 to 10 km) with the land. We estimated surface atmospheric transfer coefficients according to an approximation whereby virtual distance from the source is increased to integrate the spreading width of the plume due to the surface (IAEA, 1981).. In other words, and to give an example, to integrate the surface effect of a source located at a distance of 5 km with neutral atmospheric stability, the source was moved 0.82 km (according to the Pasquill formula) farther away to integrate

plume spreading, and will thus be virtually considered as a source located at a distance of 5.82 km rather than 5 km. Using this approach, we calculated mean surface ATC for distances of 1, 3, 5, 10 & 25 km, assuming neutral atmospheric stability (D as per Pasquill's classification), which corresponded to our weather conditions for each experimental campaign day involving the Gourey semaphore. The ATC calculated in this way can be found in table 2. This approach is clearly simplified as we apply the same ATC for a distance of 6 or 10 km for example. However, as the origin of the source term is uncertain (e.g. does the 3H come from a zone located at a distance of 4 to 5 km ?, or 8 to 10 km ?), this solution has the advantage of being quick and easy to implement. This point must be improved in the future. These ATC were necessary in order to obtain the results to be discussed in section 4.2.

4. Results

4.1 Estimating evaporation rates

Figure 5 shows variation in evaporation rate as calculated every 3 hours over the 2017-2020 period, with a mean value over 72h. According to our calculations, variation is minor, and depends on the temperate oceanic climate in the region, which leads to minor variations in mean air and water temperatures. Mean monthly seawater temperatures in this region (NBS and BS) are estimated at 9°C in winter and 18°C in August (https://marc.ifremer.fr/resultats/temperature_et_salinite/modele_mars3d_manche_gascogne). Maximum evaporation rate is recorded from August to December (mean value of 4.4 ± 0.7 L.m⁻².d⁻¹) and minimum evaporation from January to July (mean value of 2.8 ± 0.4 L.m⁻².d⁻¹). Mean evaporation calculated over the 01/01/2017-30/06/2021 period for the La zone Hague zone is 3.4 ± 2.6 L.m⁻².d⁻¹, with a median value of 2.8 L.m⁻².d⁻¹, and minimum and maximum values of 0 and 23 L.m⁻².d⁻¹ respectively. We could compare these values with that proposed by Yu (2008) equal to 3.08 L.m⁻².d⁻¹ (112 cm.y⁻¹) estimated for the global ocean in 2005 with an upward trend of approx. 10% annually. Nie *et al et al* (2020) estimate mean evaporation (in the China sea) of 1 m y⁻¹, i.e. 2.74 L.m⁻².d⁻¹.

4.2 HTO in atmospheric water vapour: comparing measurements with models

The modelling process described above is used to calculate ³H activity in seawater by Mars 2D in downwind zones for period around the measuring date (Figure 2), calculate evaporation

and apply the ATC. We simulated variation in induced HTO activity on the coast at our measuring point at Gouy Semaphore for each experiment day (Table 1). 2 days are detailed for illustration below: 24/05/2019 and 12/09/2019. Measured and simulated values are compared for other experiment days and shown in supplementary material A3.

4.2.1 Experiment A6: 24/09/2019

During this experiment day, the wind was blowing west northwest (304° on average, 2.7 m s^{-1}). The results of ^3H measurements in atmospheric water vapour are shown in Figure 6, with the ^3H activity simulated in seawater for the corresponding downwind zone (zone k, $300\text{-}330^\circ$, from 1 to 5 km, Figure 3). The activity measured in atmospheric water vapour varies from 2.4 to 6.5 Bq.L^{-1} at 8:40 and 13:10 UTC respectively. This activity increases from 07h to 13h before decreasing. Mean tritium activity in seawater, for zone k from 1 to 5 km, varies in the same way, with minimum and maximum values of 12 and 35 Bq.L^{-1} respectively. For interpretation purposes, we also reproduced the ^3H activity maps as simulated in seawater in Figure 7 at 09:00, 11:30 and 13:00 UTC. The passing of a plume of tritiated water is clearly visible downwind with wind from 304° at approx. 11:30 (the white arrow indicates wind direction), with a maximum in activity levels of 60 Bq.L^{-1} , which corresponds, prior to the subsequent arrival of the maximum value measured onshore at approx. 13:00 (HTOair, Figure 6). At 13:00, the concentrated plume has already passed through to the north and no longer appears to be downwind of the measuring point on the coast, which is confirmed onshore with a lower measurement at 14:00 UTC, indicating the start of a decreased impact of the water mass on the atmosphere.

Finally, we modelled HTO activity onshore induced by the water mass according to the process described in section 3, and Figure 8 compares measured values for activity with modelled values. The model is based on ^3H data in zone k ($300\text{-}330^\circ$) downwind and at a distance of 0-10 km from the coast, with evaporation data, ATC at 10 km (Table 2), the surface area of zone 0-10 km, weather conditions on 24/05/2019, coupled with measuring background noise of 2 Bq.L^{-1} in atmospheric water vapour. Measuring dynamics are correctly reproduced by the model.

4.2.2 Experiment A8: 12/09/2019

Atmospheric HTO measurements on 12/09/2019 can be found in Figure 9. Mean wind on the sampling day was blowing west-south-west from 252°, at 5.1 m.s⁻¹. ³H in seawater was simulated in zones h (210-240°), i (240-270°) and j (270-300°). ³H activity in seawater, as modelled for zone i, is indicated in Figure 9 for the 1-5 km zone. Atmospheric HTO activity varied from 3.6 to 13.9 Bq.L⁻¹ over the day, which is a significant variation for ambient conditions. In parallel, mean activity modelled in seawater between 1 and 5 km matches the same dynamics for zone i (240-270°) downwind, with activity varying from 20 to 50 Bq.L⁻¹.

The first 3 atmospheric measurements taken at 6:25, 7:12 and 8:25 increase from 8.9 to 13.9 Bq.L⁻¹, with the sampling point located downwind from the plume circulating in seawater offshore at that point in time, characterised by high concentrations with activity in excess of 100 Bq/L (Figure 10 shows the development of the marine plume of ³H every hour between 6 and 13 h). From 9:00, due to the tide changing direction, the plume moving northwards started to move southwards (Figure 10) and the tritium concentration of the downwind water mass decreased progressively. Consequently, atmospheric activity decreased after 08:30 (Figure 9).

HTO activity onshore induced by the water mass was modelled according to the process described in section 3, and values are compared with measurements on Figure 11. The model is based on ³H data in zone i (240-270°) downwind and at a distance of 0-10 km from the coast, with evaporation data, ATC at 10 km (Table 2), the surface area of zone 0-10 km, weather conditions on 12/09/2019, coupled with measuring background noise of 2 Bq.L⁻¹ in atmospheric water vapour. Measuring dynamics are correctly reproduced by the model.

4.2.3 Discussion

Several zones likely to influence the onshore measuring point were tested on each experiment day, and the zone with values most similar to experimental measurements was selected. The logic was clearly managed based on wind direction. Modelled data correspond to a zone and influence distance selected to match the measurements as far as possible. Background HTO in atmospheric water vapour of 2 Bq.L⁻¹ was added to the results of the model. The results are presented in supplementary materiel A3 for the 18 days. After this first modelling work, it appears that the dynamics of HTO variation in the atmosphere at the coast can globally be reproduced considering the presence of a marine plume offshore. In most of our experiments,

simulation by model for variations of HTO in water vapour at the coast are correctly reproduced (days A1 A2 A3 A4 A5 A6 A8 A9 GB1 GB3 GB5 and GB6). On days when the model and measurements are not really comparable either in dynamic or in activity levels (days A7, A10, A11, A12), it generally appears that the wind conditions are not ideal. For A7 experiment, wind are variable all the days. For A10, A11, A12 experiments, winds are from south to south east, with origin not only from the sea, and in addition, activities in the sea appears not important with no liquid release during these three days..

In most cases, the model corresponds best to the measurements by considering the overall impact zone 0-10 km. In some rare cases (experiment GB6 for example, supplementary material A3) it will be rather the zone 0-5 km which will be impactful and will best match the model to the measurements (in the case of GB6, a very concentrated plume runs along the coast less than 3 km, which did not happen on the other days of experiments). It is therefore important to understand that each moment is different; one day the plume of concentrated tritiated water can be along the coast 1-5 km for example, and the next, the concentrated plume may be closer to the 10 km. This depends primarily on the tide and when the discharge occurred. In addition, due to the alternation of tides one can also be influenced by an older rejection. In the end, the spatio-temporal gradient is always different.

Considering these results, it would appear that, even if simulations could be improved to better correspond to experimental measurements, we are able to almost-systematically reproduce the dynamics of HTO activity in atmospheric water vapour, if a “concentrated” plume of ^3H travelled offshore in seawater, in the 0-10 km zone, upwind. At this stage of the work, a steady wind is also necessary to be able to be sure of relying on the calculated tritium activities for a well-defined area of marine origin.

On the basis of this comparison, we estimate that this method can be used to simulate atmospheric values on the coast, and therefore explain increases in atmospheric HTO activity. If we can reproduce onshore HTO activity, we can also consider that the method can be used to estimate source term at sea in a satisfactory manner.

4.3: Estimating a global marine source term, Hague area, Normand-Breton Gulf and Bay of Seine

Considering that the liquid discharges from the Orano RP affect the entire NBG and BS on a long-term basis, we estimated the potential quantities of ^3H released to the atmosphere over one year for this entire marine region. For this purpose, we simulated ^3H activity in seawater for each zone defined in Figure 4, from 2017 to 2020, every 3 hours, based on the individual discharges released by Orano RP and actual weather conditions. Figure 12 shows variation in, for example, ^3H activity in seawater for area 4 in BS, area 13 near to the La Hague zone, area 18 in the NBG and area 19, which is the most north-western zone in the central part of the western English Channel where Atlantic waters first enter (Figure 4 and Table 3). Between these 4 zones, area 13 has the highest activity and the most significant variations, due to the nearby discharge point. On the other hand, zone 19, shows the lowest activity levels as it is less affected by discharges. Between these two extreme zones, zone 18 in the NBG varies little, however mean activity levels (5.6 Bq.L^{-1}) are relatively high (Table 3). This could be explained by smoothing effect of the NBG with long term recirculation of water masses as showed in Fievet et al, 2020. Finally, zone 4 in the Bay of Seine records clear variations, due to influencing by discharges, while showing strong sensitivity to meteorological effects such as periods with an east wind, but with a lower mean activity (4.6 Bq.L^{-1}) reflecting the relatively fast water flow and the mixing with central Channel Atlantic waters.

We used the model described previously (3.2 and 3.3) by coupling the hydrodynamic dispersal model with Rohwer's evaporation model to estimate the source term for this marine zone, i.e. the quantity of tritium released by the sea to the atmosphere. The quantities of tritium released are then calculated per unit surface area in each zone defined in Figure 4. The aim is to obtain mean values over a 4-year period and to see possible spatial variations. Table 4 shows tritium activity released in TBq calculated for each year from 2017 to 2020 per zone. If we total these emissions per year, the global value for marine emissions range between 104 TBq for 2020 and 150 TBq for 2017, with a mean value of 130 TBq.y^{-1} . It appears that zones 13 and 14 are always the most active zones with 12 - 21 TBq.y^{-1} released (Figure 13).. Zones 16, 17, 19 and 20 in the north west have the lowest emissions (Figure 13). Zones 19 and 16 are the nearest to the central part of the English Channel, which has low contamination levels due to mixing with Atlantic water. The water in this region is more extensively diluted with Atlantic water, and can even sometimes be totally replaced during strong meteorological events (storms or continuous southwest winds). We observe that emissions are very homogeneous in Bay of Seine, ranging from 6.4 to $7.7 \text{ GBq.km}^{-2}.\text{y}^{-1}$?. In the south of the NBG, emissions vary from 4.4 in the west to over 8.2 in the east of the Gulf. In the northwest

part of the NBG emissions are lower, from 0.9 to 3.5 GBq.km⁻².y⁻¹?, and finally the highest emissions are concentrated in the La Hague zone, near to La Hague Cape and near to the discharge point, ranging from 8.2 to 11.3 GBq.km⁻².y⁻¹?

Table 5 indicates mean and total emissions, for combined values corresponding to the 3 large zones defined herein: La Hague, NBG and BS. We obtain mean emission values of 51, 44 and 34 TBq.y⁻¹ for the Normand-Breton Gulf (10,813 km²), Bay of Seine (6,332 km²) and La Hague (3,521 km²) respectively.

4.4: Comparing the marine source terms with direct atmospheric discharges from the Orano La Hague RP

The total values in Table 5 were compared with direct gaseous atmospheric discharges released via the chimneys of the Orano RP (table 6). If we consider the direct discharges (for HT+HTO) from stacks representing 58 to 72 TBq.y⁻¹ between 2017 and 2020 (<https://www.orano.group>), it appears that marine emissions (130 TBq.y⁻¹ on average) is more important by a factor 1.8 to 2.2. If we exclusively consider atmospheric HTO discharges (estimated at approx. 15% of total direct discharges from chimneys with the remaining 85% comprising HT, Connan et al, 2017), then marine emissions contribute 12 to 15 times more than atmospheric direct atmospheric emissions of HTO.

We estimated that marine HTO emissions were over 10 times greater than atmospheric emissions from stacks. However, we must also point out that this source area covers large distances of several hundred km from west to east (representing roughly 20,000 km²) sometimes far from land. If we consider the prevailing wind directions, i.e. northeast or west in this region, these "marine" emissions will generally be dispersed seawards rather than landwards. However, more specific studies could be carried out to estimate the percentage of these emissions which actually return to land, considering weather conditions and the corresponding atmospheric transfers. A coupled marine and atmospheric dispersion model could be developed for this purpose. Another question would be to consider as impacted terrestrial regions located far from the direct gas source, but close to the sea. For example, in our case, the north of Brittany, located far from the gaseous point source, is close to the southern part of the NBG. Finally, reasoning to the extreme, it could be argued that the north-west zone of France would be under the potential influence of a source which would be the English Channel.

As these works are unique in their kind, little information is available in the literature on tritium emissions from marine zones for comparison purposes. The only available data are provided by Maro et al. (2005), who had proposed a tritium emission rate of 39 TBq.y⁻¹ for a surface area of 4,400 km² in Bay of Seine. This value had been calculated on the basis of tritium gradient measurements between seawater and the atmosphere, over a dozen ad hoc measuring points during a marine campaign. When given per 1 km² for Bay of Seine, as in our works, the emission rate given by Maro *et al* (2005) is equivalent to 8.9 GBq km⁻². If we consider our mean value calculated for Bay of Seine (44.4 TBq.y⁻¹ for 6,332 km², table 5) converted to 1 km², we obtain 8.5 GBq.km⁻², which is extremely similar.

Finally, we recall that the quantities of liquid discharged from the Orano RP, from 2017 to 2020, varied from 11,400 to 13,200 TBq.y⁻¹ with a mean value of 11,975 TBq.y⁻¹. With a mean marine emission rate of 130 TBq.y⁻¹ as calculated in our work, 1.1% of tritium is re-released to the atmosphere (It should be noted, however, that from a regulatory point of view, this re-emitted quantity is already reported as a liquid activity). Little data are available in the literature, however, we could mention Nie *et al* (2020), who estimated the fraction of tritium which could evaporate in Hangzhou Bay in the China sea at 10-12.5%, however the hydrodynamic and climatic conditions are completely different to our study zone (semi-closed estuary in the eastern China Sea). In another parametric study on reservoirs, the same authors (Nie *et al.*, 2021) estimated a mean value of 2.5% evaporation (approx. 1 to 4.5% depending on the vertical diffusion coefficient and the selected water evaporation rate), but which could be lower depending on the depth of the discharge. Nie *et al* (2021) also estimated, in their work, that for a HWR (Heavy Water moderated and cooled reactor) Nuclear Power Plant, the annual tritium evaporation rate from liquid release would be comparable to direct gaseous tritium discharges.

5. Conclusions

Thanks to 18 field campaigns, we were able to highlight the influence of a mass of water enriched with tritium on the potentially measurable HTO activity levels in atmospheric water vapour downwind of this mass of water. We demonstrated that evaporation at the ocean-atmosphere interface can transfer a measurable fraction of tritium quantities to the atmosphere, which can then be carried to land by wind. The Mars-2D hydrodynamic model can be used to obtain tritium levels in seawater in the western English Channel. It is coupled

with an evaporation model depending on weather conditions, and a Gaussian type atmospheric transport model. These coupled models allow to reconstitute variations in atmospheric tritium activity on the coast, depending on sea conditions based on liquid discharges of tritium from the Orano La Hague fuel reprocessing plant. On this basis, when a mass of water containing approximately 20 to 100 Bq.L⁻¹ of tritium flows between 0 and 10 km off the coast, variations in atmospheric activity measured onshore can reach 2 to 15 Bq.L⁻¹ downwind. If we consider the Normand-Breton Gulf and the Bay of Seine in the western English Channel, a marine region influenced by liquid discharges from the Orano RP, mean tritium emissions by the sea to the atmosphere were estimated for the entire region (~21,000 km²). Over the 2017-2020 period a flux of 130 TBq.y⁻¹ was calculated, with emissions which vary depending on distance from the discharge point. Emissions near to the discharge point were estimated at 8.2 - 11.3 GBq km⁻².y⁻¹. In the Bay of Seine, emissions are homogeneous and range from 6.4 to 7.7 GBq km⁻² y⁻¹ while in the NBG, due to the complex hydrodynamics at play, emissions vary from 0.9 GBq.km⁻².y⁻¹ in the northwest part of the NBG, to 8.2 GBq.km⁻².y⁻¹ in the central part of the NBG. If we compare the "marine" source term for tritium (only HTO form) with the direct source term for gaseous discharges (HTO and HT) from the RP, the marine source term is two orders of magnitude greater for the entire marine region. If we consider only gaseous discharges of HTO from chimneys, difference are one order of magnitude greater. In terms of impact, because of the very low dosimetric impact of tritium, there is no particular topic on this aspect. Finally, if we consider the mean liquid discharge from the Orano RP over the 2017-2020 period, our work led us to estimate that approximately 1.1% of this discharge was re-released to the atmosphere each year.

Even if tritium has very little impact in terms of dose, the atmospheric impact is higher than the marine one and it may be necessary to take into consideration the quantities of tritium released by the sea and transported to land in order to improve the precision of impact studies. It requests the availability of an atmospheric transport model coupled with a marine dispersion model able to integrate the surface source. It will be necessary to differentiate between tritium transported to land and tritium which remains diluted in the atmosphere above the sea. Finally, when tritiated water is expected to be released at Fukushima, this study provides relevant answers on the aspects of quantities that could possibly pass into the atmosphere.

Acknowledgements

The authors thank Orano RP of La Hague for their partnership during this research and for providing release gaseous and liquid data, and Mr Fievet B. for helpful discussions on the dispersal of radionuclides. Finally we thank G. Bacon for the older samples, method and measurements used in this work.

6. References

- AIEA 1981. Dispersion atmosphérique et choix des sites de centrales nucléaires, guide de sûreté, N°50-SG-S3., Agence Internationale de l'énergie atomique, Vienne, 1981.
- Bacon G., Bailly du Bois P., Maro D., Solier L., 2011. Etude du transfert entre l'eau et l'atmosphère d'un rejet marin de tritium (HTO) en zone côtière. Etude préliminaire. CESI, Rapport 3ème année école d'ingénieur, 38 p.
- Bailly du Bois, P., Salomon, J.C., Gandon, R., Guegueniat, P., 1995. A quantitative estimate of English Channel water fluxes into the North Sea from 1987 to 1992 based on radiotracer distribution. *J. Mar. Syst.*, 6, 457–481.
- Bailly du Bois, P., Guegueniat, 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, 1977–2002.
- 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*, 169-210.
- Bailly du Bois P., Dumas F., 2010. Short term model in a macro tidal sea : precision and sensitivity of modelling compared with tracers and physicals measurements. IRSN/DEI-SECRE/2010-51, 29pp.
- Bailly du Bois P., 2011. Automatic calculation of bathymetry for coastal hydrodynamic models. *Computers & Geosciences* 37 (2011), pp. 1303-1310. 10.1016/j.cageo.2010.11.018
- Bailly du Bois P., Dumas F., Solier L., Voiseux C., 2012. In-situ database toolbox for short-term dispersion model validation in macro-tidal seas, application for 2D-model. *Continental Shelf research*, 36, 30, 63-82.
- Bailly du Bois P., 2014. Dispersion des radionucléides dans les mers du nord-ouest de l'Europe : observations et modélisation. Mémoire d'habilitation à diriger des recherches HDR, IRSN, 329 pp, ISBN 978-2-954237-2-9
- Bailly Du Bois P., Morillon M., Solier L., Laguionie P., Fievet B., Voiseux C., Le Cavalier S., 2018. Investigation sur la dispersion et le transfert de radionucléides dans le golfe normand breton. IRSN, rapport PSE-ENV/2018-00050, 26 pp.
- Bailly du Bois, P., Dumas, F., Morillon, M., Furgerot, L., Voiseux, C., Poizot, E., Mear, Y., Bennis, A.C., 2020a. Alderney Race, general hydrodynamic and particular features. *Phil. Trans. Roy. Soc. A* 378: 20190492. <http://dx.doi.org/10.1098/rsta.2019.0492>
- Bailly du Bois, P.B., Dumas, F., Voiseux, C., Morillon, M., Oms, P.E., Solier, L., 2020b. Dissolved Radiotracers and Numerical Modeling in North European Continental Shelf Dispersion Studies (1982-2016): Databases, Methods and Applications. *Water*, 12, 1667, doi :10.3990/w12061667.
- Briggs, G.A., 1985. Analytical parametrizations of diffusion e the convective boundary layer. *J. Clim. Appl. Met.* 14, 1167-1186.
- Buesseler K.O., 2020. Opening the floodgates at Kukushima, *Science*, 369, 6504, 621-622.
- Connan O, D. Hebert, L. Solier, D. Maro, G. Pellerin, C. Voiseux, M. Lamotte, P. Laguionie. 2017. Atmospheric tritium concentrations under influence of AREVA NC La Hague reprocessing plant (France) and background levels. *Journal of Environmental Radioactivity*, 177, 184-193.
- Doury, A., 1981. Le vadémécum des transferts atmosphériques. rapport n_CEA-DSN- 440. CEA.
- ERA-Interim reanalysis, 2011. Configuration and performance of the data assimilation system. D. P. Dee S. M. Uppala A. J. Simmons P. Berrisford P. Poli S. Kobayashi U. Andrae M. A. Balmaseda G. Balsamo P. Bauer P. Bechtold A. C. M. Beljaars L. van de Berg J. Bidlot N. Bormann C. Delsol R. Dragani M. Fuentes A. J. Geer L. Haimberger S. B. Healy H. Hersbach E. V. Hólm L. Isaksen P. Kållberg M. Köhler M. Matricardi A. P. McNally B. M. Monge-Sanz J.-J. Morcrette B.-K. Park C. Peubey P. de Rosnay C. Tavolato J.-N. Thepaut F. Vitart. First published: 28 April 2011 <https://www.ecmwf.int/node/8946>.

- Fievet B., Bailly Du Bois P., Voiseux C., Godinot C., Cazimajou O., Solier L., De Vismes Ott A., Cossonet C., Habibi A., Fleury S., 2020. A comprehensive assessment of two decade radioactivity monitoring around the Channel Islands, *Journal of Environmental Radioactivity*, 223-224, 106381.
- ICRP, 2012. Compendium of dose coefficient based upon ICRP publication 60. ICRP Publication 109, 132 pp.
- Lazure P., Dumas F., 2008. An external internal mode coupling for a 3D hydrodynamical model for applications at regional scale (MARS). *Advances in Water resources*, 31, 32, 233-250.
- Lyard F., Lefevre F., Letellier T., Francis O., 2006. Modelling the global ocean tides: modern insights from FES2004. *Ocean Dyn. Earth Environ. Sci.* 56, 394–415. (doi:10.1007/s10236-006-0086-x)
- Marang L., Siclet F., Luck M., Maro D., Tenailleau L., Jean Baptiste P., Fourré E., Fontugne M., 2011. Modelling tritium flux from water to atmosphere: application to the Loire River. *Journal of Environmental Radioactivity*, 102, 244-251.
- Maro D., Tenailleau L., Fontugne M., Germain P., Hebert D., Rozet M., 2005. Tritium transfer between sea and atmosphere in the English Channel (North Cotentin and Bay of Seine). *Radioprotection*, 40, 1, 589-594.
- Michel R., 1976. Tritium inventories of the world oceans and their implications. *Nature*, 263, 103-106. <https://doi.org/10.1038/263103a0>.
- Nie B., Yang J., Wang W., Gu Z., Yuan Y., Li F., 2020. Numerical study on tritium dispersion in coastal waters : The case of Hangzhou Bay, China. *Journal of hydrology*, 590, 125532.
- Nie B., Yang J., Yuan Y., Li F., 2021. Additional radiation dose due to atmospheric dispersion of tritium evaporated from a hypothetical reservoir, *Applied Radiation and Isotopes*, 167, 109475.
- Oms P.E., 2018. Tritium as an original continental waters tracer in the Bay of Biscay: measurements and modelling. XVIth International Symposium on Oceanography of the Bay of Biscay (ISOBAY 16) Anglet, France 5-7 June 2018.
- Oms P-E, Bailly du Bois P., Dumas F., Lazure P., Morillon M., Voiseux C., Le Corre C., Cossonet C., Solier L., Morin P., 2019. Inventory and distribution of tritium in the oceans in 2016. *Science of The Total Environment*, 656, 1289-303.
- Pasquill, F., 1961. Estimation of the dispersion of windborne material. *Meteorol. Mag.* 90, 33-49.
- Povinec, P. P.; Aoyama, M.; Biddulph, D.; Breier, R.; Buesseler, K.; Chang, C. C.; Golser, R.; Hou, X. L.; Jeřkovský, M.; Jull, A. J. T.; Kaizer, J.; Nakano, M.; Nies, H.; Palcsu, L.; Papp, L.; Pham, M. K.; Steier, P.; Zhang, L. Y., 2013. Cesium, iodine and tritium in NW Pacific waters; a comparison of the Fukushima impact with global fallout. *Biogeosciences*;10(8):5481-96
- Rohwer C., 1931. Evaporation from free surface water surface, *USDA Tech Null*, 217, 1-96.
- Singh V.P., Xu C.Y., 1997. Evaluation and generalization of 13 mass transfer equations for determining free water evaporation. *Hydrological Process*, 11, 311-323.
- Salomon J.C., Guéguéniat P., Breton M. 1991 Mathematical model of 125Sb transport and dispersion in the Channel. In *Radionuclides in the study of marine processes*, Norwich, UK, 10– 13 September 1991 (eds JP Kershaw, DS Woodhead), pp. 74–83. London, UK: Elsevier Applied Science.
- Scheppard S.C., Ciffroy P., Siclet F., Damois C., Sheppard M.I., Stephenson M., 2006. Conceptual approaches for the development of dynamic specific activity models of 14C transfer from surface water to humans, *Journal of environmental Radioactivity*, 87, 32-51.
- Schmidt E., 1989. *Properties of Water and Steam in SI-Units*. Grigull U. (ed.), Springer-Verlag Berlin, 206p.
- TEPCO, 2020. Draft study responding to the subcommittee report on handling ALPS treated water.
- Triplet J.P., Roche G., 1986. *Météorologie Générale*. Météo-France, Trappes, 317pp.
- UNSCEAR, 2000. Sources and effects of ionizing radiation, Vol 1 Sources: United Nations Scientific Committee on the effects of Atomic Radiation. UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation; 2000 p. 659.

UNSCEAR, 2016. Sources, effects and risks of Ionizing Radiation. In Annex C, biological effects of selected internal emitters, tritium. UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation; 2016 p. 122.

Upstill-Goddard, R.C., 2006. Air sea gas exchange in the coastal zone. *Estuarine Coastal and Shelf Science*, 70, 388-404.

Xu C.Y., Singh, V.P., 2001. Evaluation and generalization of temperature based methods for calculating evaporation. *Hydrological Processes*, 15, 305-319.

Xu C.Y., Singh, V.P., 2002. Cross comparison of empirical equations for calculating potential evapotranspiration with data from switzerland. *Water Ressources Management*, 16, 197-219.

Yu L., 2008. Global Variations in Oceanic Evaporation (1958–2005): The Role of the Changing Wind Speed. *Journal of Climate*, 20, 5376-5390.

Zeng M. Liu Y., Ouyang S., Shi H., Li C., 2016. Nuclear energy in the post-Fukushima Era : research on the developments of the Chines and worldwide nuclear power industries. *Renew. Sustain. Energy Rev*, 58, 147-156.

Zhao C., Wang G., Zhang M., Wang G., de With G., Bezhenar R., Maderich V., Xia C., Zhao B., Jung K.T., Periañez R., Akhir M.F., Sangmanee C., Qiao F., 2021. Transport and dispersion of tritium from the radioactive water of the Fukushima Daiichi nuclear plant. *Mar Pollut Bull*, 169,: 112515. doi: 10.1016/j.marpolbul.2021.112515.

Figure 1 : Map of study area : Goury Sempahore is the location of experimental measurements in air (black dot). Position of release in sea from Orano La Hague Reprocessing Plant (RP) is represented by a cross.

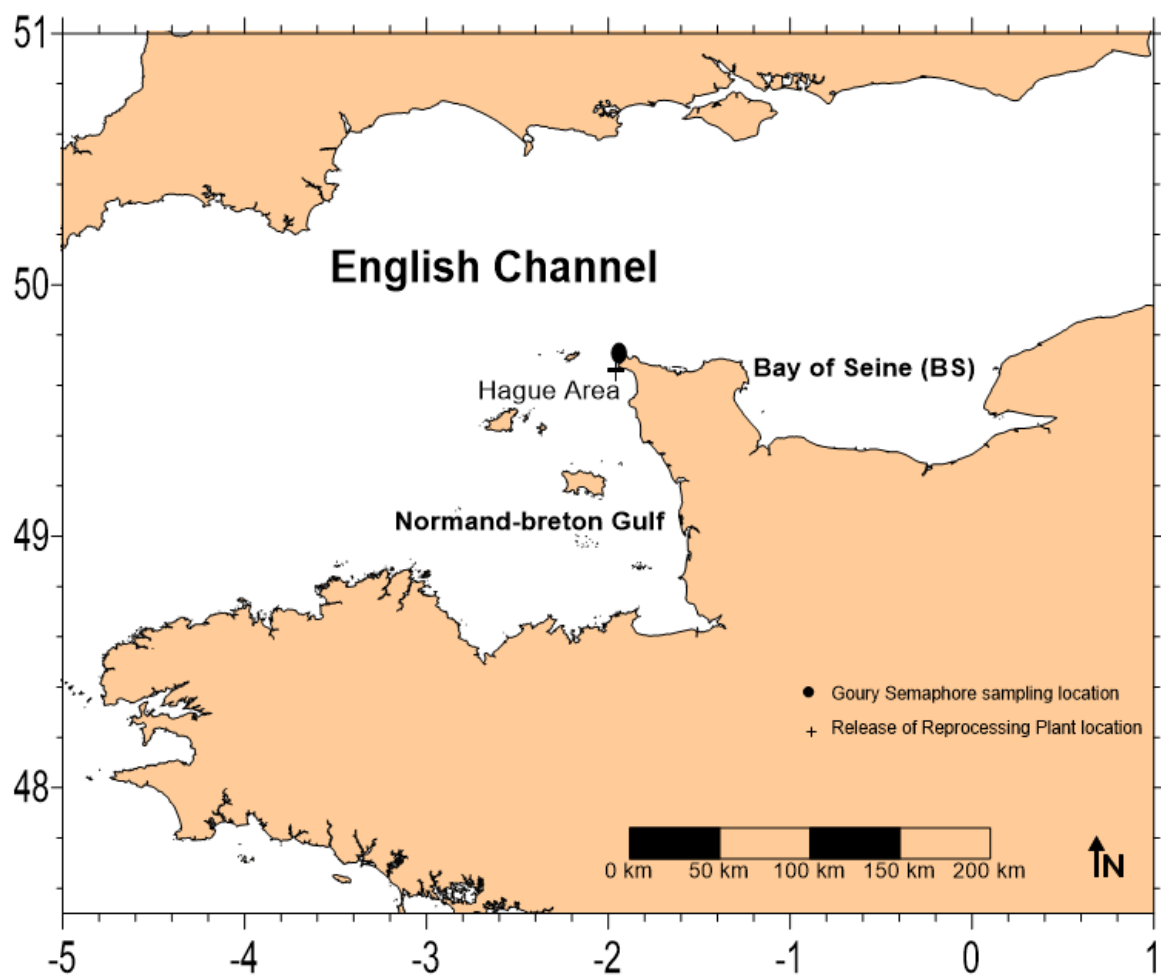


Figure 2 : Diagram of the model used for the calculation of HTO activity in air due to the water mass.

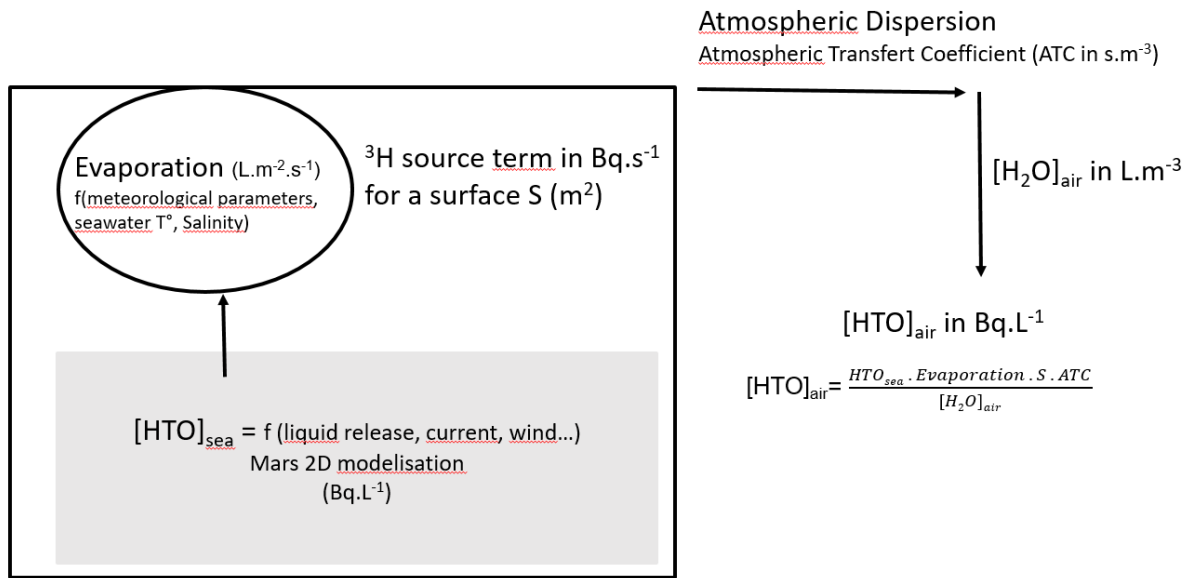


Figure 3 : Map of potential influence zones for the location of the Semaphore of Gourey according to wind directions, for simulations by Mars-2D. Each zone has an opening of 30° , and is subdivided into 4 boxes at distances of 1, 3, 5, 10 km from the coast.

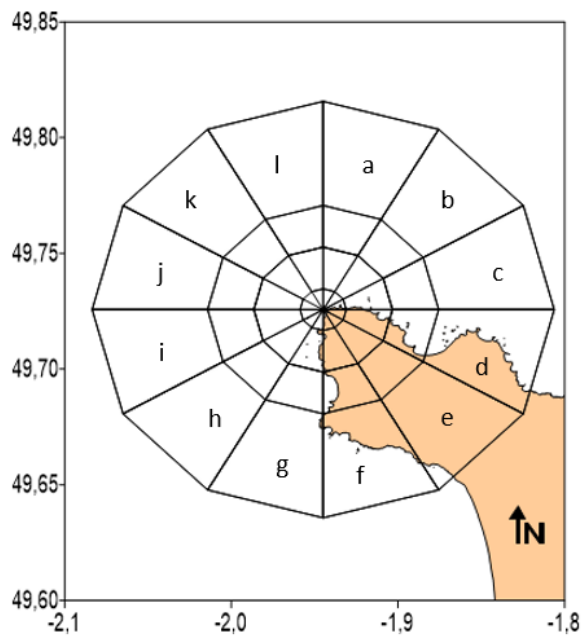


Figure 4 : Map and definition of box numbering used for marine source term calculation (numbering is arbitrary and has no particular meaning).

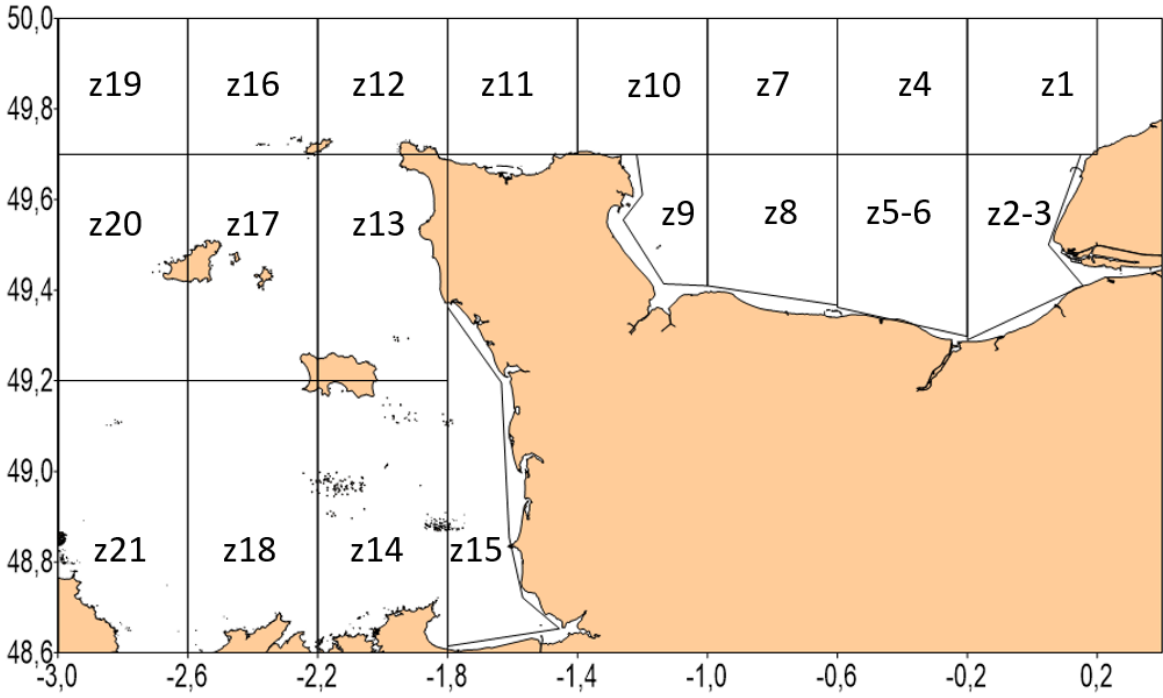


Figure 5 : Evaporation (in $L \cdot m^{-2} \cdot d^{-1}$) calculated every 3h for the Hague area (z12, figure 4) by the Rohwer method for the 2017-2020 period (black line represent the 72h average).

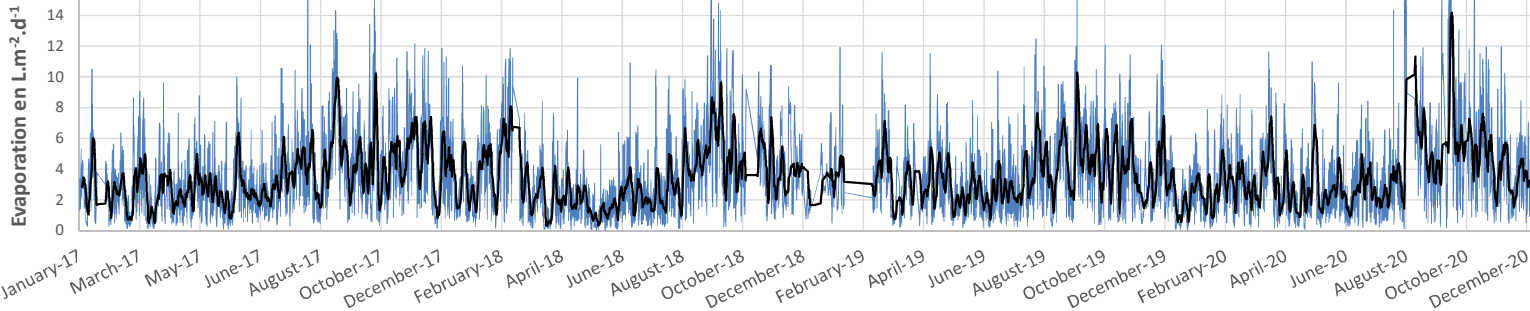


Figure 6 : 24/05/2019 ; measurements of HTO in air water vapour (Bq.L⁻¹) at Goury Semaphore location and simulated HTO in seawater (Bq.L⁻¹) for area k from figure 3 (300-330°, distance 1-5 km) ; the dotted lines represent the hours of simulation of the figure 7.

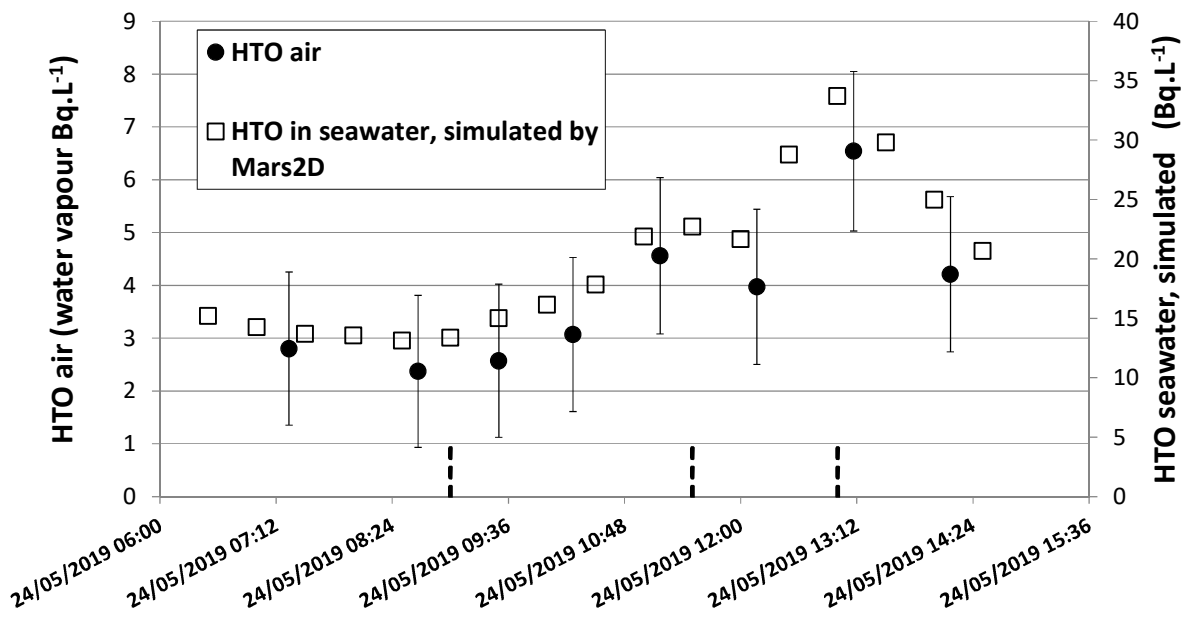


Figure 7 : 24/05/2019 ; [HTO] simulated in seawater (Bq.L⁻¹) at 09h, 11h30, 13h TU. The white arrow indicates the wind direction.

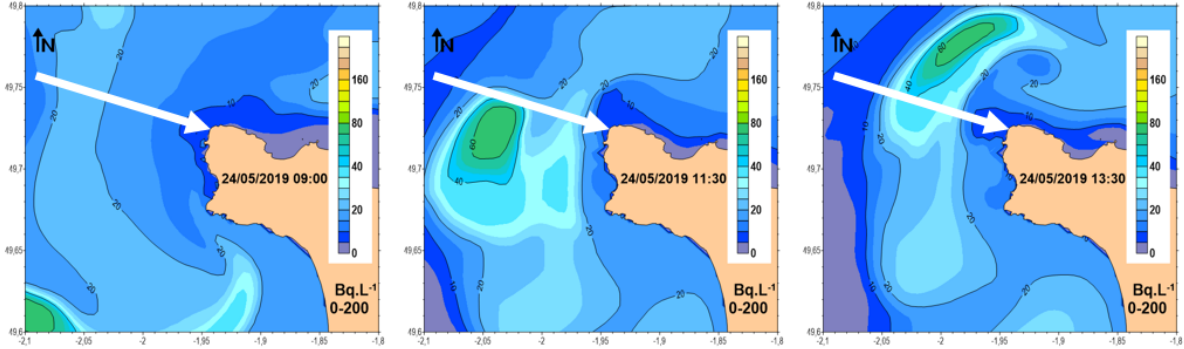


Figure 8: 24/05/2019; [HTO] in water air vapour (Bq.L⁻¹) at Goury Semaphore location, measurements in black circles, simulated (marine dispersion + evaporation + atmospheric dispersion) in white squares.

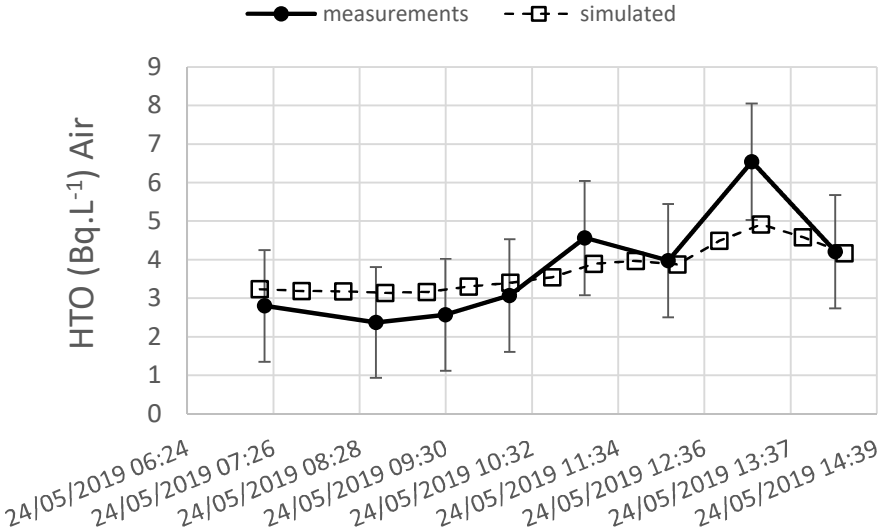


Figure 9 : 12/09/2019 ; measurements of HTO in air water vapour(Bq.L⁻¹) at Goury Semaphore location and simulated HTO in seawater (Bq.L⁻¹) for area i from figure 3 (240-270°, distance 1-5 km) ; the dotted lines represent the hours of simulation of the figure 10.

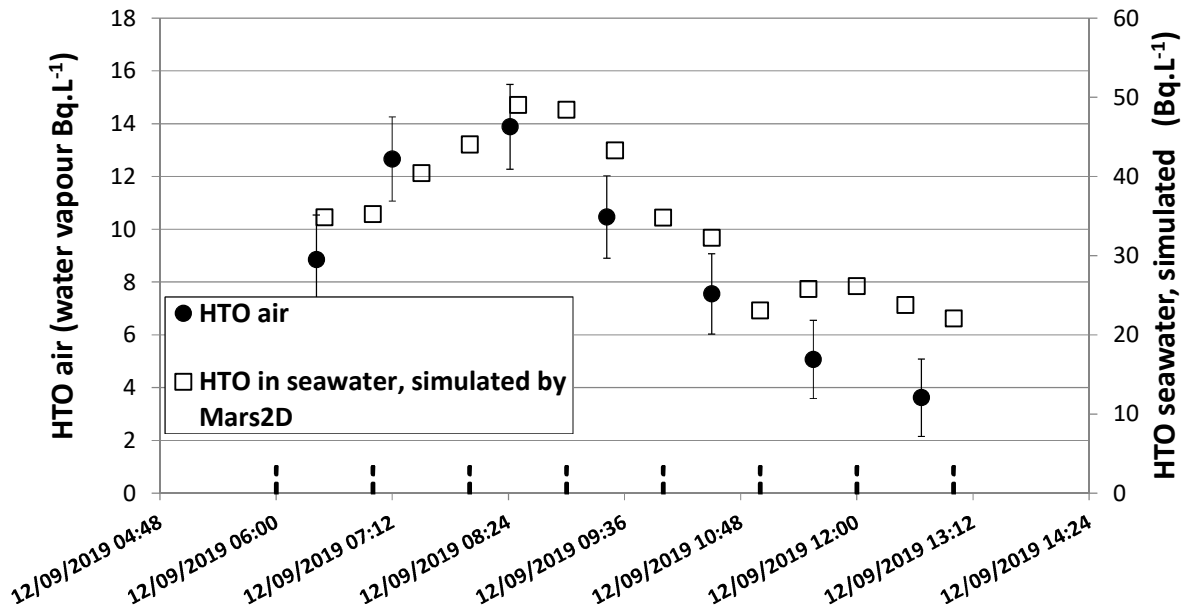


Figure 10 : 12/09/2019 ; [HTO] simulated in seawater (Bq.L^{-1}), 06h, 07h, 08h, 09h, 10h, 11h, 12h, 13h. The white arrow indicates direction of wind.

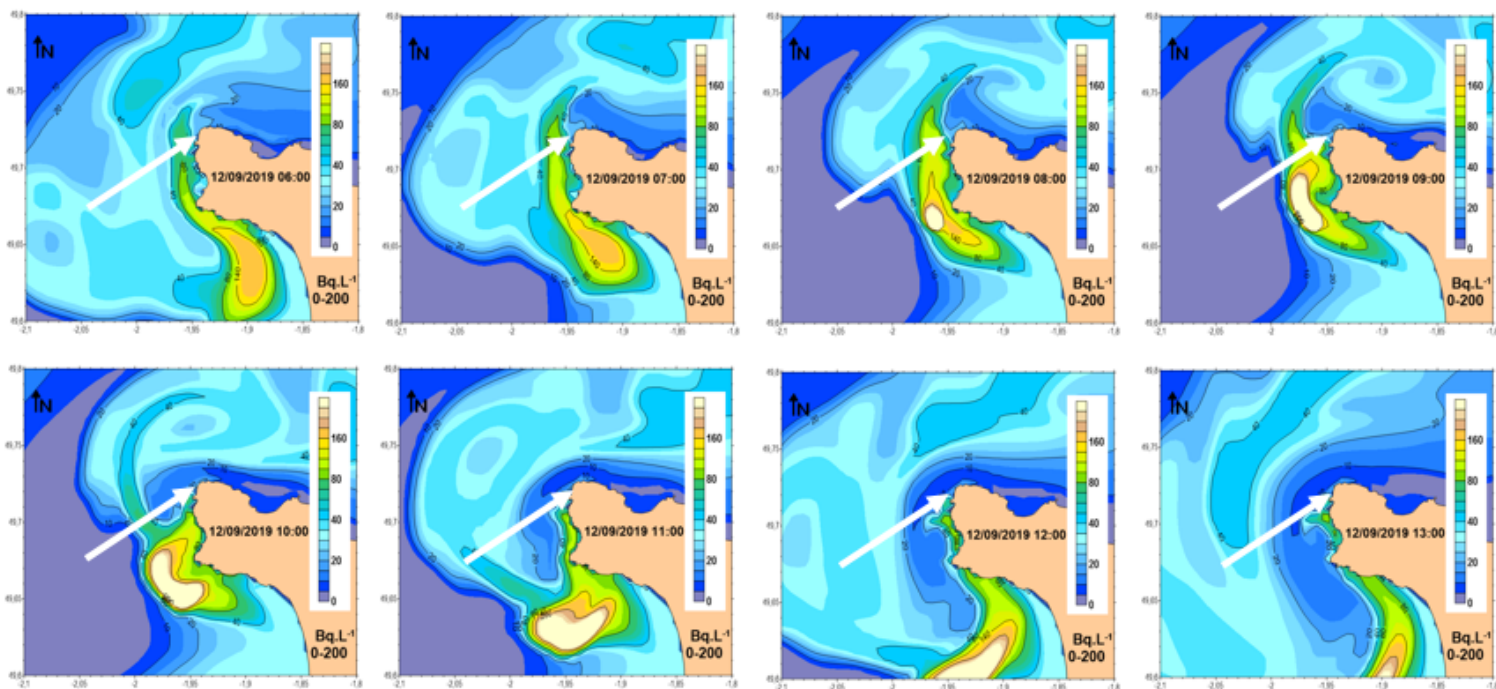


Figure 11: 12/09/2019 ; [HTO] in air water vapour (Bq.L⁻¹) at Goury Semaphore location, measurements in black circles, simulated in white squares (marine dispersion + evaporation + atmospheric dispersion).

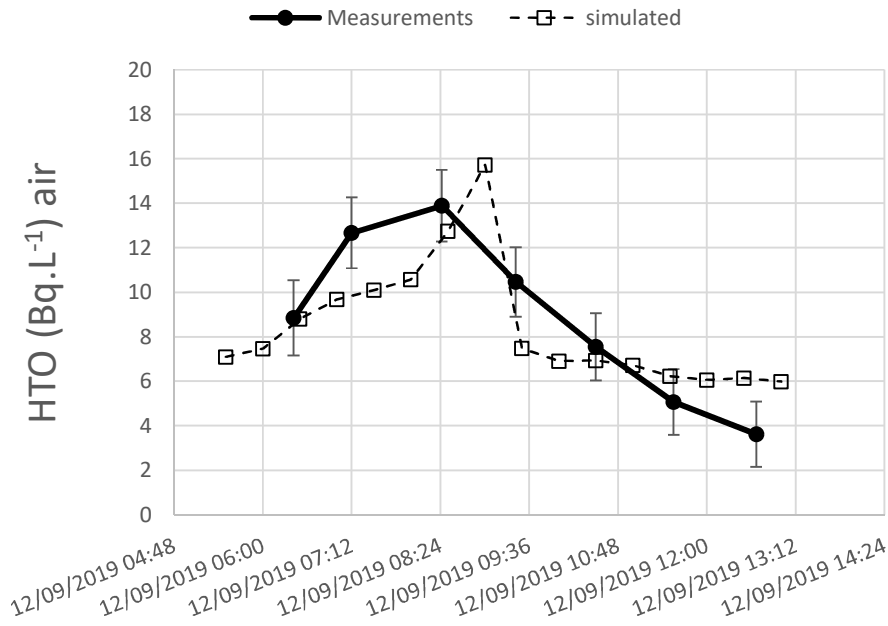


Figure 12 : Mean additional ³H (Bq.L⁻¹) in seawater simulated by Mars-2D for 2017-2020 period for area z4, z13, z18, z19 (see Figure 4 for position of zone).

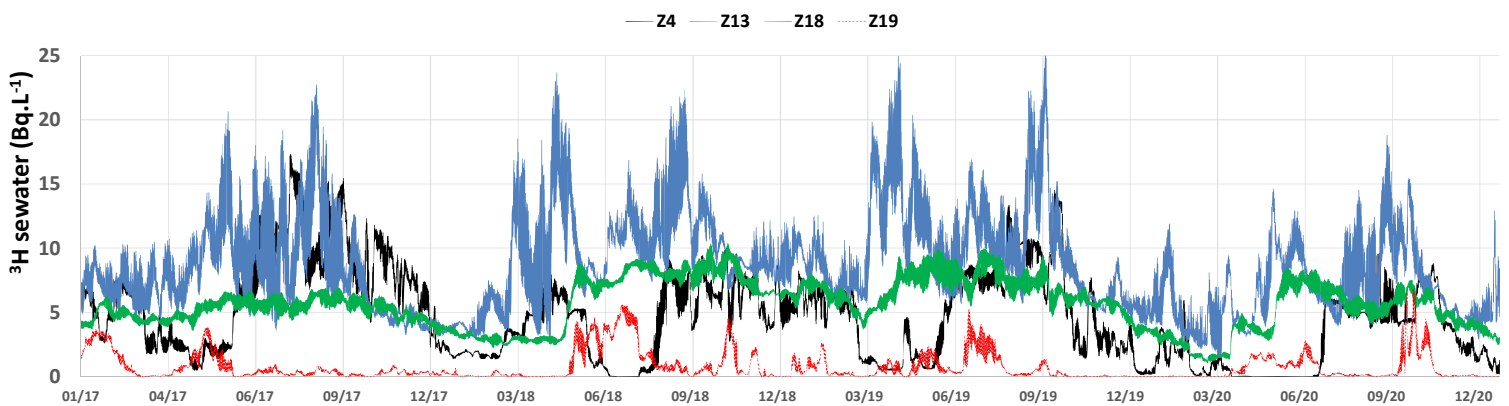


Figure 13 : Mean ³H emission from seawater for different area: in GBq.km⁻².y⁻¹ (in bold), in TBq.y⁻¹ (italic) for different area (name of area from figure 4 and indicate in bracket).

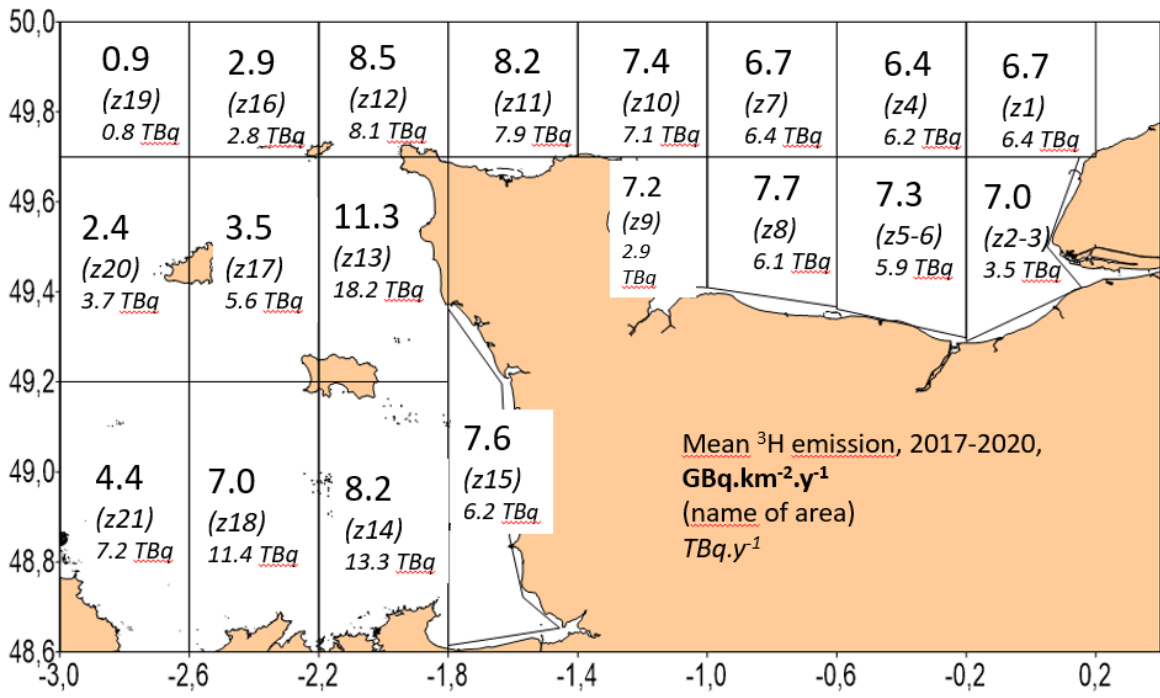


Table 1 : Characteristics of experimental days for the measurement of HTO in the water vapour of the air at the Goury Semaphore .

| N° campaigns | time start (TU) | time end (TU) | Wind direction (°) | Wind speed | Activity max | Activity min |
|--------------|------------------|------------------|--------------------|----------------------|-------------------------------|-------------------------------|
| | | | | (m.s ⁻¹) | HTO air (Bq.L ⁻¹) | HTO air (Bq.L ⁻¹) |
| A1 | 21/02/2018 08:45 | 21/02/2018 14:37 | 50 | 6.6 | 2.7 | 1.7 |
| A2 | 10/09/2018 05:45 | 10/09/2018 13:10 | 246 | 5.3 | 6.9 | 3.2 |
| A3 | 03/10/2018 10:30 | 03/10/2018 15:00 | 264 | 4 | 4.5 | 2.8 |
| A4 | 09/05/2019 07:10 | 09/05/2019 12:55 | 262 | 7.7 | 13.5 | 5.8 |
| A5 | 21/05/2019 05:30 | 21/05/2019 13:15 | VRB (300°-40°) | 2.6 | 6.3 | 5.4 |
| A6 | 24/05/2019 06:45 | 24/05/2019 14:00 | 304 | 2.7 | 6.5 | 2.4 |
| A7 | 19/06/2019 05:35 | 19/06/2019 11:45 | 245 | 1.7 | 9.6 | 5.9 |
| A8 | 12/09/2019 06:32 | 12/09/2019 12:40 | 252 | 5.1 | 13.9 | 3.6 |
| A9 | 04/02/2020 08:10 | 04/02/2020 14:25 | 301 | 8.1 | 6.2 | 4.7 |
| A10 | 24/03/2021 07:55 | 24/03/2021 16:10 | 196 | 4 | 3.7 | 4.9 |
| A11 | 25/03/2021 07:15 | 25/03/2021 15:00 | 202 | 8.8 | 2.8 | 3.6 |
| A12 | 26/03/2021 06:15 | 26/03/2021 11:30 | 187 | 14.5 | 2.8 | 4.3 |
| GB-R1 | 18/01/2011 18:15 | 18/01/2011 22:00 | 345 | 5.3 | 7.3 | 4.3 |
| GB-R2 | 21/01/2011 16:00 | 22/01/2011 03:30 | 40 | 6.7 | 5.8 | 2.6 |
| GB-R3 | 23/01/2011 06:22 | 23/01/2011 17:32 | 33 | 9.1 | 9.6 | 1.3 |
| GB-R4 | 26/01/2011 07:50 | 26/01/2011 20:35 | 41 | 9.3 | 5.9 | 2.3 |
| GB-R5 | 08/03/2011 19:27 | 08/03/2011 22:31 | 180 | 4.8 | 9.3 | 4.7 |
| GB-R6 | 17/03/2011 14:23 | 17/03/2011 22:20 | 258 | 9.2 | 12.1 | 2.4 |

Table 2 : Surface Atmospheric Transfert Coefficient (ATC) in s.m⁻³ used for atmospheric transport from 1, 3, 5, 10 km from the coast (from Pasquill gaussian model).

| distance | 1 km | 3 km | 5 km | 10 km |
|--------------------------|----------|----------|----------|----------|
| ATC (s.m ⁻³) | 2.00E-05 | 3.70E-06 | 1.80E-06 | 7.50E-07 |

Table 3 : Min, max, mean and median ^3H activities in Bq.L^{-1} , for the 2017-2020 period, for the 4 area Z4, Z13, Z18, Z19 (see Figure 4 for position of zone).

| ^3H (Bq.L^{-1}) | Z4 | Z13 | Z18 | Z19 |
|-------------------------------------|------|------|------|-----|
| min | 0.0 | 1.4 | 1.0 | 0.0 |
| max | 17.3 | 25.2 | 10.4 | 6.8 |
| mean | 4.6 | 8.7 | 5.6 | 0.7 |
| median | 4.4 | 8.2 | 5.7 | 0.2 |

Table 4 : ^3H emission in TBq.y^{-1} from the different area of figure 4, for 2017-2020 period.

| | 2017 | 2018 | 2019 | 2020 | Average (TBq) |
|--------------|------------|------------|------------|------------|---------------|
| Z1 | 10.8 | 3.6 | 7.0 | 4.1 | 6.4 |
| Z2-3 | 5.9 | 1.5 | 4.2 | 2.5 | 3.5 |
| Z4 | 9.4 | 4.9 | 6.2 | 4.1 | 6.2 |
| Z5-6 | 9.7 | 2.7 | 6.9 | 4.1 | 5.9 |
| Z7 | 8.9 | 5.6 | 6.5 | 4.6 | 6.4 |
| Z8 | 9.4 | 3.2 | 7.3 | 4.6 | 6.1 |
| Z9 | 4.1 | 1.5 | 3.5 | 2.4 | 2.9 |
| Z10 | 9.1 | 6.0 | 7.9 | 5.3 | 7.1 |
| Z11 | 9.6 | 7.5 | 8.6 | 5.8 | 7.9 |
| Z12 | 8.5 | 8.5 | 8.8 | 6.8 | 8.1 |
| Z13 | 16.8 | 18.4 | 21.7 | 15.7 | 18.2 |
| Z14 | 12.3 | 13.0 | 16.0 | 11.8 | 13.3 |
| Z15 | 5.7 | 6.1 | 7.4 | 5.6 | 6.2 |
| Z16 | 2.4 | 3.2 | 2.9 | 2.6 | 2.8 |
| Z17 | 4.8 | 6.8 | 5.8 | 4.8 | 5.6 |
| Z18 | 10.6 | 11.5 | 13.4 | 10.0 | 11.4 |
| Z19 | 0.8 | 0.8 | 0.6 | 1.0 | 0.8 |
| Z20 | 3.7 | 5.0 | 3.4 | 3.1 | 3.8 |
| Z21 | 7.0 | 8.3 | 8.0 | 5.6 | 7.2 |
| Total | 150 | 118 | 146 | 104 | 130 |

Table 5 : Mean emission of ^3H (in $\text{GBq.km}^{-2}.\text{y}^{-1}$ and TBq.y^{-1}) over the period 2017-2020, for the Normand-Breton Gulf (NBG), Bay of Seine (BS) and Hague area.

| Area | $\text{GBq.km}^{-2}.\text{y}^{-1}$ | TBq.y^{-1} |
|-------------------|------------------------------------|---------------------|
| NBG (Z14 à Z21) | 4.7 | 51.0 |
| BS (Z1 à Z10) | 7.0 | 44.4 |
| Hague (Z11 à Z13) | 9.7 | 34.2 |

Table 6 : Comparison of tritium emissions from the marine environment with direct gaseous emissions from the Orano RP stack .

| | 2017 | 2018 | 2019 | 2020 |
|---|-------------|-------------|-------------|-------------|
| Marine Emission (TBq/year) | 150 | 118 | 146 | 104 |
| Gaseous Release (TBq/year) | 71.6 | 60.2 | 65.9 | 58.1 |
| HTO (15% of gaseous emission) | 10.7 | 9.0 | 9.9 | 8.7 |
| Ratio Marine emission/Total gaseous release | 2.1 | 2.0 | 2.2 | 1.8 |
| Ratio Marine emission/HTO gaseous release | 14 | 13 | 15 | 12 |