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Olivier Connan, Donovan Maire, Didier Hebert, Luc Solier, Philippe Laguionie, Marianne Rozet, Marianne Lamotte, Denis Maro

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1 **Tritium in precipitation on 5 sites in North-West France during the 2017-**  
2 **2019 period.**

3 <sup>(1)</sup>Connan O., <sup>(2)</sup>Maire D., <sup>(1)</sup>Hébert D., <sup>(1)</sup>Solier L., <sup>(1)</sup>Laguionie P., <sup>(1)</sup>Rozet M., <sup>(1)</sup>Lamotte  
4 M., <sup>(1)</sup>Maro D

5 <sup>(1)</sup>Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-ENV/SERIS/LRC, Radioecology Laboratory, ,  
6 BP10, rue Max Pol Fouchet, 50130 Cherbourg-Octeville, France.

7 <sup>(2)</sup> Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSE-ENV/SAME/LMN, Nuclear Metrology  
8 Laboratory, BP35, rue de l'écluse, 78119 Le Vésinet, France

9 \* Corresponding author:

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

11 *IRSN*

12 *Cherbourg-Octeville Radioecology Laboratory*

13 *Rue Max Pol Fouchet*

14 *BP10*

15 *50130 Cherbourg Octeville*

16 *Tel (33) 2 33 01 41 08*

17 *Fax (33) 2 33 01 41 30*

18

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## 2 **1. Introduction**

3 Tritium ( $^3\text{H}$ ) is a low energy  $\beta$ -emitting radionuclide with a half-life of 12.32 years. Tritium is  
4 produced by naturally-occurring processes through the interaction of gases and cosmic radiation.  
5 Natural abundance is estimated at about 4.5 kg of tritium, produced at an average rate of 0.32  
6  $\text{atom}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$  (Masarik and Beer, 2009; UNSCEAR, 2016), corresponding to  $\approx 258$  g.y (Oms et al.,  
7 2019). Atmospheric tests of nuclear weapons between 1945 and 1980 led to an estimated release of  
8 668 kg, corresponding to about  $242 \cdot 10^6$  TBq, during that period (Okada and Momoshima, 1993;  
9 UNSCEAR, 2000); in 2016, the tritium remaining from the fallout from atmospheric nuclear weapons  
10 testing is estimated to be 22-23.5 kg (Oms et al., 2019). Most of the  $^3\text{H}$  released by these nuclear  
11 explosions was injected into the stratosphere in its hydrogen gas (HT) and methyl tritium gas ( $\text{CH}_3\text{T}$ )  
12 forms. The tritium then crosses the tropopause from stratosphere to troposphere. The flux varies over  
13 time, with higher fluxes in spring and early summer (Ehhalt et al., 2002). Today,  $^3\text{H}$  is mainly released  
14 into the environment by nuclear power plants, military nuclear facilities and nuclear reprocessing  
15 plants (CNSC, 2009; ASN, 2010). Globally, gaseous releases for all nuclear facilities are currently  
16 estimated at  $11,700 \text{ TBq}\cdot\text{y}^{-1}$  ( $16,000 \text{ TBq}\cdot\text{y}^{-1}$  for liquid releases), with reprocessing plants representing  
17 approximately  $300 \text{ TBq}\cdot\text{y}^{-1}$ . Artificial gaseous emissions of tritium globally are estimated at around 33  
18  $\text{g}\cdot\text{y}^{-1}$ . In France, gaseous  $^3\text{H}$  releases from nuclear power plants are estimated at between 23 and 50  
19  $\text{TBq}\cdot\text{y}^{-1}$ , with the Orano La Hague fuel reprocessing plant releasing around 70-80 TBq into the  
20 atmosphere ( $\approx 0.2 \text{ g}\cdot\text{y}^{-1}$ ) depending on the year. Other French facilities (different industries and  
21 research centres) release approximately  $400 \text{ TBq}\cdot\text{y}^{-1}$  ( $\approx 1.1 \text{ g}\cdot\text{y}$ ), the majority of which is attributed to  
22 the Valduc and Marcoule centres (UNSCEAR, 2000: UNSCEAR, 2016; IRSN, 2019). In the  
23 troposphere, HT forms are oxidized into HTO before being extracted from the atmosphere through  
24 precipitation. Since the early 1960s, tritium activities in precipitation have steadily decreased (Tadros  
25 et al., 2014, Eyrolle-Boyer et al., 2014).

26 Tritium activities in precipitation can be very variable locally due to the direct influence of nuclear  
27 facilities, but also to the mixing of air masses between the stratosphere and the troposphere, and to the  
28 origin of the air mass (marine or continental) at the time of the precipitation. In the northern  
29 hemisphere, when the continental surface heats up in the spring, the height of the tropopause increases,  
30 causing a larger than normal tritium influx from the stratosphere to the troposphere. This increases its  
31 overall tritium concentration, and eventually leads to an increase in the tritium activity measured in  
32 rainwater in spring and summer, a phenomenon known as 'spring leak' (Newell, 1963; Gat et al.,  
33 2001). The activities in rainwater (but obviously also in water vapour in the air) also depend on  
34 latitude, altitude and distance from the sea. When an air mass is marine in origin, water vapour  
35 exchanges above the oceans at the sea/atmosphere interface tend to reduce the activity in the air, so

36 precipitations have lower tritium activities when the origin of the air mass is oceanic (Gat et al., 2001;  
37 Ehhalt et al., 2002; Crawford et al., 2013; Cauquoin et al., 2015; Tadros et al., 2014; Dului et al.,  
38 2018).

39 Tritium activities in precipitation are monitored by the Global Network of Isotopes in Precipitation  
40 (GNIP) in partnership with the World Meteorological Organization (WMO). The database is available  
41 through the IAEA (IAEA/WMO, 2019). As part of the GNIP's activities, monthly precipitation  
42 samples were collected at 1000 stations in more than 125 countries in the past (International Atomic  
43 Energy Agency, 2019). For France, data for precipitations exist for 47 sites but historical tritium data  
44 only exist for a few sites (principally Avignon, Brest, Le Mans, Marseille, Orléans, Strasbourg and  
45 Thonon Les Bains). The oldest data date from 1963 and the most recent from 2013. For some sites,  
46 historical records are intermittent or cover only a few years. In France, the continental site of Thonon  
47 Les Bains (46.3722°N, 6.4708°E) has the longest and most regular monitoring record with 571  
48 measurements spread evenly over the period 1963 to 2012. In the context of our study, the Brest site  
49 (48.3600°N, -4.5700°W), will also be of interest, even though only 78 measurements between 1996  
50 and 2002 are available, because it is right on the coast, like the sites we worked with in this study.  
51 Other sites across Europe monitored by the GNIP will also be of use to us in this study for comparison  
52 (Madrid, Valentia, Punta Delgada).

53 This study was launched on the basis of the finding that we had few low-level tritium measurements  
54 (below 1 Bq.L<sup>-1</sup>) for precipitations in France. There are plenty of measurements taken as part of the  
55 regulatory monitoring of nuclear facilities by health institutions and the nuclear operators themselves  
56 for environmental monitoring purposes, but they are almost always at detection limits of several Bq.L<sup>-1</sup>  
57 (in particular because of water drinking standards) and are usually taken close to facilities so there is  
58 a potential direct influence (for example, there are several thousand rainwater measurements for the  
59 RNM site (French National Measurement Network (RNM), 2019) but the detection limit for these is  
60 usually between 3 and 5 Bq.L<sup>-1</sup>). In previous studies (Crawford et al., 2013), measurements of HTO in  
61 water vapour in the air revealed extremely low activities on French background sites (where there was  
62 no influence from nuclear activities), which prompted us to ask what the real levels in rainwater in  
63 France might be. Furthermore, tritium is particularly heavily monitored in France by environmental  
64 protection and anti-nuclear associations, which communicate extensively on the subject, so it is  
65 important for institutional bodies to have expertise in this area. We therefore decided to measure  
66 tritium at low levels in rainwater, mainly on the site of the Cherbourg En Cotentin-LRC  
67 (Radioecology Laboratory), based on samples taken twice a month, accompanied by periodic  
68 measurements on several other sites. Particular attention was given in this study to quantifying very  
69 low values. The aim was ultimately to provide recent data on tritium levels in rainwater in north-west  
70 France, the area of particular interest to us (Connan et al., 2017; Maro et al., 2017); a more extensive  
71 study covering the whole of France would obviously have been desirable, but this was not possible to

72 do rapidly for reasons of low level measurement capacity, so the decision was made initially to restrict  
73 it to our normal area of study. In this study we present the results of measurements taken on these  
74 sites, most of which are less than 100 km from nuclear facilities and are therefore potentially impacted  
75 by tritium releases into the atmosphere. The results provide up-to-date data, which can be used for  
76 comparison purposes. The study does not aim to continue in the long term or to replace monitoring,  
77 such as that done by the GNIP.

78

## 79 **2. Materials and methods**

### 80 **2.1 Sampling location and collection**

81 Rainwater was sampled in north-west France (Figure 1b), in the Normandy region on sites at  
82 Cherbourg-LRC (49.634°N, -1.645°W), Cherbourg-Town Center (49.639 °N -1.628°W), Omonville  
83 La Petite-PTILH (49.694°N , -1.8733°W), Coutances (49.044°N, -1.455°W), Saint-Lô (49.119°N, -  
84 1.107°W) and in the Brittany region on the Pontrieux site (48.702°N , -3.1644°W). The Cherbourg-  
85 LRC site is at the Radioecology Laboratory of IRSN (the French Institute for Radiological Protection  
86 and Nuclear Safety). The Normandy region has several nuclear facilities (Flamanville NPP, Orano NC  
87 La Hague reprocessing plant, Cherbourg submarine naval base, Andra's La Hague disposal facility)  
88 which could potentially have an impact as regards tritium on atmospheric water vapour and rainwater.  
89 The Pontrieux site in the Brittany region is some distance from the Normandy region and is subject to  
90 prevailing west or south-west winds from the Atlantic Ocean, so overall it is outside the influence of  
91 nuclear facilities. This will be our reference site.

92 The samples from the Cherbourg-LRC site (30 samples) were taken using an Eigenbrodt pluviometer  
93 (Figure 2a), situated 2.5 m above the ground. This pluviometer has a rain sensor that causes it to open  
94 during precipitation and close in dry periods, preventing exchanges between the rainwater and the  
95 atmosphere. The tank is a 5-litre amber glass bottle and the pluviometer's surface area measures 452  
96 cm<sup>2</sup>. The 30 samples were taken between 29/09/2017 and 28/05/2019 over periods of 12 to 43 days  
97 (mean = 20 days). The samples from the Coutances and Saint-Lô sites (18 samples) and the  
98 Cherbourg-Town Centre site (11 samples) were taken between 25/09/2018 and 25/03/2019 using a  
99 pluviometer specially constructed for periodic short samples, with a surface collection area of 53 cm<sup>2</sup>,  
100 and a 500 ml glass tank containing a floating ball covering the whole surface to limit exchanges  
101 between the water and the atmosphere (Figure 2b). Sampling periods varied from 8 to 43 days, with a  
102 mean of 16 days (it should be noted that these samples were taken as part of an education programme  
103 promoting scientific discovery and openness to society). The samples from the Pontrieux site (15  
104 samples) were taken periodically over short periods (2 to 24 hours) using a pluviometer (funnel and  
105 polyethylene bottle) with a surface area of 660 cm<sup>2</sup> between 19/10/2017 and 18/03/2019. The samples

106 from the Omonville la Petite-PTILH site (Figure 1b) were taken as part of other research work and are  
107 presented here for comparison with our other sites. The Omonville la Petite-PTILH site is 2 km north  
108 of the Orano NC La Hague plant and is often affected by tritium releases from the plant. Rainwater  
109 samples are taken there at a high frequency every 48 hours (if it rains) using a pluviometer with a large  
110 surface area (0.25 m<sup>2</sup>) and an automatic sampling system (Bülher BU2000 system) where each  
111 rainwater sample is stored and refrigerated in a separate glass receptacle in which the surface is  
112 covered by a floating ball to prevent exchanges with the atmosphere (if it does not rain for 48 hours,  
113 the corresponding bottle is not filled and remains empty). For this site, we present the results obtained  
114 over the period 09/2017 to 04/2019 (196 samples). Finally, we also took 3 opportunistic samples on a  
115 continental site in central France, at Lusignan (46.416°N, 0.1188°E) in July 2018 (Figure 1a).

## 116 **2.2 Tritium measurements**

117 The samples taken at the different sites are kept in darkness in cold storage before being prepared and  
118 measured using the low level liquid scintillation counting method. Each sample was filtered to 0.45  
119 µm then mixed with a liquid scintillation cocktail in a low diffusion polyethylene vial (with a teflon  
120 coating inside the vial). The liquid scintillation cocktail used is Ultima Gold LLT (Perkin Elmer). The  
121 water/liquid scintillation cocktail ratio is 70/70 ml or 10/10 ml depending on the liquid scintillation  
122 counter (LSC) used.

123 The device with the best performance is an ALOKA LSC-LB07 (Hitachi), in which each sample is  
124 counted for 24h. The background count rate and counting efficiency were  $2.77 \pm 0.11$  cpm and  $20.7 \pm 2.4$   
125 % respectively. The whole batch of samples is counted over 6 cycles (6 times the whole batch) of 4  
126 hours for each sample. One batch contains 7 samples maximum in order to ensure the samples remain  
127 in the counter for less than one week. Each batch of samples contains one reference sample with a  
128 known <sup>3</sup>H activity in order to check measurement quality. The LB07 counter is calibrated using  
129 groundwater (low level of tritium) spiked with a known quantity of a <sup>3</sup>H standard solution. Several  
130 standards were made and quenched with nitromethane in order to cover the whole quenching index  
131 range of the samples. The decision threshold (SD) was 0.15 Bq.L<sup>-1</sup>. Measurement uncertainty  
132 (average) was 23%, 18% for samples > 0.5 Bq.L<sup>-1</sup> (depending on the activity) and 50% for samples <  
133 0.5 Bq.L<sup>-1</sup>. Below the SD, the uncertainty was 100%. For the samples counted with the LB07, the  
134 results below SD=0.15 Bq.L<sup>-1</sup> were indicated in the tables. These 'uncensored' values were used in the  
135 mean calculations (Vivier et al., 2012; Puydarrieux et al., 2015). For other samples, we used a TRI-  
136 CARB 3180 TRSL (PerkinElmer) with a counting time of 16 hours per sample. With this 3180 TRSL  
137 LSC, the decision threshold (SD) is 1.0 Bq.L<sup>-1</sup> in the sample. The background count rate and counting  
138 efficiency were 2.09 cpm and 14.9%. The measurement uncertainty depends on the level of activity,  
139 but based on 151 significant measures (>1 Bq.L<sup>-1</sup>) on the PTILH site, average uncertainties is 31%.

140 Our measurement practices are regularly validated by means of comparison exercises between French  
141 and international laboratories. Our measurement laboratories are also currently involved in writing a  
142 standard on tritium measurement (XP NF M60-824, test method for analysis of tritium in free water  
143 and organically bound tritium in environmental matrices). During this study, 67 samples were  
144 measured with the better-performing ALOKA LSC-LB07 (Hitachi) and 10 samples were measured  
145 with the TRI-CARB 3180 TRSL (PerkinElmer) for the Pontrieux, Cherbourg-LRC, Cherbourg-Town  
146 Centre, Coutances, Saint-Lô and Lusignan sites. For the Omonville-PTILH site, the 196 samples were  
147 measured by the TRI-CARB 3180 TRSL (PerkinElmer).

148

## 149 **3. Results**

### 150 **3.1: Cherbourg-LRC site**

151 The results for the Cherbourg-en-Cotentin-LRC site are presented in Figure 3 and Table A1. The  
152 values range from 0.2 to 2.42 Bq.L<sup>-1</sup>. The mean value is 1.00 Bq.L<sup>-1</sup>. In Figure 3, it can be seen that  
153 the minimum values are measured in the October to March periods, corresponding to winter. The  
154 maximum values are measured between May and July 2018, then in May 2019 (an upwards trend in  
155 activities is observed from April 2019), which accords with the 'spring leak' phenomenon (the increase  
156 is around 0.7 Bq.L<sup>-1</sup>). Two values seem to stand out in the graph: the highest value ( $2.42 \pm 0.22$  Bq.L<sup>-1</sup>)  
157 was observed between 15/10/2018 and 05/11/2018 and a value of  $1.35 \pm 0.20$  Bq.L<sup>-1</sup> was observed  
158 between 03/02 and 01/03/2019. The value 2.42 Bq.L<sup>-1</sup> is confirmed by the measurement over a similar  
159 period (23/10 to 06/11/2018) of an activity of  $3.02 \pm 0.23$  Bq.L<sup>-1</sup> on the Cherbourg-Town Centre site  
160 1.3 km away (Figure 3 and Table 2). Examination of the pluviometer data and wind directions  
161 indicates that, during 2 rain events (26/10 and 01/11/2018) during this period, the wind was coming  
162 from the direction of the Orano NC plant 18 km to the north-west, with a pluviometry volume over  
163 these two episodes representing 10% to 15% of the total over the whole period. In the case of the 1.35  
164 Bq.L<sup>-1</sup> value measured between 03/02 and 01/03/2019, the higher value can be explained by a rainfall  
165 episode on 01/03/2019, between 0:00 hrs and 08:00 hrs, with 10mm of rain (i.e. 15% of the total  
166 collected during the period), which also originated in an air mass from the north-west, the direction of  
167 the Orano plant (this is confirmed by significant <sup>85</sup>Kr activity, a marker of atmospheric releases from  
168 the Orano plant, Connan et al., 2017). In these conditions, where rainfall is concomitant with an air  
169 mass affected by anthropogenic atmospheric releases (of HTO, HT and <sup>85</sup>Kr), it is entirely possible for  
170 the value to double or triple over a sampling period of 15 to 30 days, just with a single rainfall episode  
171 (specific experiments conducted by our team in 2016-2017 to collect rain from the plume emitted by  
172 the Orano plant at a distance of only 1 to 3 km from the plant, showed values ranging from 09 to 54  
173 Bq.L<sup>-1</sup>; so if for example 10% of the volume collected has an activity of 9 Bq.L<sup>-1</sup>, for 100mm of

174 rainwater, the activity can be multiplied by 3 compared to the mean activity). Overall, for the  
175 Cherbourg-LRC site, rainwater values below 1 Bq.L<sup>-1</sup> are the norm, but it only take one episode of  
176 rain has to be associated with winds from the Orano NC plant at La Hague for activities to increase  
177 significantly.

### 178 **3.2: Pontrieux site (reference)**

179 The results for the Pontrieux site are presented in Figure 3 and Table 1. This site is some distance  
180 away from any nuclear facilities, in an area where the prevailing winds are from the south-west; it is a  
181 site influenced by the sea, with the Channel approximately 10 km to the north and the Atlantic Ocean  
182 100 km to the west (Figure 1). The values range from 0.01 to 1.19 Bq.L<sup>-1</sup>. Only one value (1.19 Bq.L<sup>-1</sup>)  
183 exceeds 1 Bq.L<sup>-1</sup> and in nearly half of cases the values are below the SD (7 values < 0.15 Bq.L<sup>-1</sup> out  
184 of 15 samples). The activities measured on this site are clearly the lowest in our study. For each  
185 sample, we traced the back trajectories (NOAA/HYSPLIT model;  
186 [https://ready.arl.noaa.gov/HYSPLIT\\_agreement.php](https://ready.arl.noaa.gov/HYSPLIT_agreement.php)) arriving at this site to have an idea of the origin  
187 of the air masses when each sample was taken (Figure A1), and we have indicated the direction of  
188 origin deduced from this in Table 1; we show that, when the wind comes from the south or east with a  
189 continental component (land), the values for HTO in rainwater are at their highest. When the wind for  
190 the last hours before sampling comes from the north/north-west with an influence of the Channel,  
191 which is more affected by tritium than the Atlantic (Oms et al., 2019), values are also significant. On  
192 the other hand, when the air masses come from the south-west or the west, from the Atlantic Ocean,  
193 the values are always the lowest, below the decision threshold of 0.15 Bq.L<sup>-1</sup>. The values are shown in  
194 Figure 3 to compare with the values measured on the two sites of Cherbourg. The highest activities are  
195 measured during the same period (May to July 2018) as on the Cherbourg-LRC site (Figure 3) which,  
196 combined with the continental component of the air masses at these times, contributes to this  
197 significant increase (a general increase can be seen from values of 0.15 to values of 0.4 to 1 Bq.L<sup>-1</sup>). It  
198 is worth noting that on this site the activities in rainwater are very low, and that values of around 0.15  
199 Bq.L<sup>-1</sup> and below are probably the level background for French sites mainly under the influence of air  
200 masses from the sea and not affected by nuclear industries. The mean calculated on the basis of all the  
201 activity values (including uncensored values < SD) is 0.36 Bq.L<sup>-1</sup>.

### 202 **3.3: Coutances, Saint-Lô and Cherbourg-Town Centre sites**

203 The results for the 3 sites at Coutances, Saint-Lô and Cherbourg-Town Center are presented in  
204 Table 2. In view of the prevailing winds, the Cherbourg-Town Centre site is the most susceptible to  
205 the impact of the region's nuclear facilities. The Coutances and Saint-Lô sites are respectively 76 and  
206 83 km south of these facilities (Figure 1b), and the prevailing wind direction on these sampling sites is  
207 not from these facilities. In the case of Coutances and Saint-Lô, the maximum values are obtained  
208 during the same period:  $1.56 \pm 0.19$  Bq.L<sup>-1</sup> from 18/10/2018 to 08/11/2018 and  $2.15 \pm 0.20$  Bq.L<sup>-1</sup>



209 from 19/10/2018 to 06/11/2018 respectively. The other values indicate low activity levels  $< 0.4 \text{ Bq.L}^{-1}$ .  
210 The mean values for Coutances and Saint-Lô are  $0.55$  and  $0.64 \text{ Bq.L}^{-1}$  respectively (Figure 5). In this  
211 geographical area to the south of the region, the activities tend to be low overall, though higher than  
212 those at the reference site at Pontrieux. These slightly higher activities are probably explained by the  
213 tritium levels in the seawater of the Channel, a few tens of kilometres to the west. Finally, the  
214 activities at the Cherbourg-Town Centre site range from  $0.38$  to  $3.02 \text{ Bq.L}^{-1}$  with a mean activity of  
215  $1.19 \text{ Bq.L}^{-1}$ . This mean is higher than at the other two sites and agrees well with those for Cherbourg-  
216 LRC, which is 2 km away (Figure 1).

### 217 **3.4: Omonville La Petite-PTILH site**

218 The measurements in rainwater at the Omonville La Petite (PTILH) site every 48 hours between  
219 25/09/2017 and 30/04/2019 (196 measurements) are presented in Figure 4. The PTILH site is used  
220 extensively by IRSN for monitoring and research, which explains why data are available for very short  
221 time intervals of 48 hours. This site is 2 km north of the Orano NC La Hague plant (Figure 1) and is  
222 regularly affected by gaseous tritium releases from the plant (87% in the form of HT, but with median  
223 HTO activities in water vapour in the air of  $66 \text{ Bq.L}^{-1}$  in the plume, Connan et al., 2017), which can  
224 have an impact on the activity in rainwater in the precipitation event takes place on an air mass coming  
225 from the plant. The HTO activities in rainwater on this site range from  $< 1 \text{ Bq.L}^{-1}$  to  $25.5 \text{ Bq.L}^{-1}$   
226 maximum (Figure 4). The mean value is  $3.8 \text{ Bq.L}^{-1}$  (mean calculated by using  $1 \text{ Bq.L}^{-1}$  when the value  
227 is  $< 1 \text{ Bq.L}^{-1}$ ) but would be lower if we have better precision  $< 1 \text{ Bq.L}^{-1}$ . This value reflects the non-  
228 continuous but recurrent influence of the Orano NC plant compared with data measured at the other  
229 sites presented above. It is worth noting that, despite the plant's proximity, many measurements are  
230 nevertheless below  $1 \text{ Bq.L}^{-1}$ , especially between October 2017 and February 2018. During this period  
231 (from 08/10/2017 to 21/01/2018 to be precise), the two main stacks emitting tritium at the Orano NC  
232 plant were shut down for maintenance. The measurements at this site are therefore representative of  
233 activities to be found on a site heavily impacted by atmospheric tritium releases, with high significant  
234 variability due to its short distance from the industrial site. Finally, to provide further information  
235 about the activities in rainwater under the direct influence of the Orano plant, specific experimental  
236 work was conducted by our team in 2016-2017 to collect rainwater under the plume emitted by the  
237 Orano plant. The rainwater samples were taken, checking by means of real-time measurements of  $^{85}\text{Kr}$   
238 that the rainwater episode was in an area under the influence of the gaseous release (rainwater was  
239 sampled in a tank with a large surface area installed on a trailer instrumented to measure  $^{85}\text{Kr}$ , that  
240 could be moved to the location of the release while in the rain): the values obtained from 6 samples at  
241 separate times range from 9 to  $54 \text{ Bq.L}^{-1}$ .

242

## 243 4. Discussion

244 Figure 5 summarises the results obtained at our different measurement sites. Unsurprisingly, the graph  
245 shows levels that increase from our reference site at Pontrieux, where measurements were very similar  
246 to those at Saint-Lô and Coutances, to the Cherbourg-LRC and Cherbourg-Town Centre sites, where  
247 the tritium levels were higher. Finally, the Omonville-PTILH site is distinctive in character due to its  
248 situation 2 km from the Orano reprocessing plant at La Hague, with higher activity levels and wider  
249 variations. We sought to verify the precipitation amount effect (Tadros et al., 2014), which is reflected  
250 in an inverse correlation between the tritium activity in rainwater and the amount of rain. Tritium in  
251 the form of HTO in water vapour in the air condenses in rain, concentrating the tritium in light rain  
252 and diluting it in more important rain. This effect is more significant on continental sites and also  
253 depends on the timing between stratosphere-troposphere exchanges and the annual rain cycle on a site  
254 (Tadros et al., 2014). If the spring-summer tritium peak related to stratosphere-atmosphere exchanges  
255 coincides with light rainfall, the effect will be maximised. To determine whether this amount effect is  
256 observed on the Cherbourg-LRC site, we normalised each tritium activity to the total average and  
257 correlated this with the precipitation (Figure 6a). Although this is a coastal site, the precipitation  
258 amount effect is observed, with a very clear negative correlation. The same plot was done for the  
259 Omonville-PTILH site (Figure 6b); in this case the effect is not visible because the tritium activities  
260 are affected too much by the atmospheric tritium emissions from the Orano reprocessing plant.

261 Our measurements have the particular characteristic of having been done (except in the case of the  
262 Pontrieux site) in a region significantly influenced by the sea (the Cherbourg peninsula is surrounded  
263 by water, Figure 1b) with prevailing winds coming from the sea (south-west or north-east for 75% of  
264 the time), but also potentially impacted by atmospheric tritium releases from nearby nuclear facilities.  
265 It should also be mentioned that the Orano plant also releases tritium into the sea in liquid waste  
266 discharges (12 to 14 PPq.y<sup>-1</sup>) leading to activities in the seawater that range from 2 to 20 Bq.L<sup>-1</sup>. Based  
267 on our seawater monitoring over the last 3 years, the mean activity in seawater at the north-west tip,  
268 near the release stack, is 12 Bq.L<sup>-1</sup>. It is 6 Bq.L<sup>-1</sup> in the eastern part of the region, and the activities are  
269 lower, at between 1 and 3 Bq.L<sup>-1</sup> in the southern part of the Channel (Gulf Normand-Breton), i.e. to  
270 the west of the Coutances and Saint-Lô sites. Figure 5 could reflect that, i.e. that the PTILH site is  
271 directly influenced by atmospheric releases from the Orano plant, and the Cherbourg-LRC and Town  
272 Centre sites are also influenced but to a lesser degree, whereas the Coutances and Saint-Lô sites,  
273 which are not under the prevailing winds of the plant, are influenced by marine air masses from the  
274 west, which pass over waters with slightly higher tritium levels than normal. Finally, the Pontrieux  
275 site, under the influence of air masses from the Atlantic most of the time, with a few exceptions  
276 presents very low activities.

277 It is therefore worth comparing our results with existing GNIP data for France and Europe. In Figures  
278 7 and 8 we have reproduced the GNIP data from several stations chosen by us (Figure 1a): the Thonon  
279 Les Bains site (46.37°N, -6.47°E) because this is the best documented site in France (continental site)  
280 and the one most often used as a reference, the Brest site at the western tip (48.36°N, -4.57°W, data  
281 from 1996 to 2002 only), the Valentia site (51.93°N -10.25°W) at the south-western tip of Ireland, and  
282 the Ponta Delgada site (37.77°N, -25.65°W) in the middle of the Atlantic (Azores), because these sites  
283 are a long way from any nuclear facilities but are strongly influenced by the Channel and the Atlantic,  
284 and in this respect they are similar to our measurement sites. Finally, the Madrid site (40.41°N, -  
285 3.67°W) in the centre of Spain, which is a continental site but in a region with few nuclear facilities  
286 and winds that, on a larger scale, come mainly from the Atlantic, seemed to be worth looking at.

287 First, we compared the data for 1996-2016 from the Thonon Les Bains, Madrid, Brest, Valentia and  
288 Ponta Delgada sites (Figure 7). Over the whole period, much lower values are observed (difference of  
289 approximately 0.5 to 0.7 Bq.L<sup>-1</sup>) at the marine sites of Brest, Valentia and Ponta Delgada than at the  
290 Madrid and Thonon Les Bains sites, both of which always have higher values. This reflects the  
291 respective marine and continental influences of these two groups of stations (with comparable  
292 latitudes for Valentia, Brest and Thonon Les Bains in particular). A difference between the values for  
293 Thonon Les Bains and Madrid is also always observed, with Madrid having lower activities than  
294 Thonon Les Bains (by around 0.2 to 0.5 Bq.L<sup>-1</sup>). Figure 8 shows the data for Thonon Les Bains, Brest  
295 and Valentia, focusing on the period 1995-2005 when data was simultaneously acquired for all 3 sites.  
296 There is a high level of agreement between the Brest and Valentia data (despite the fact that they are  
297 600 km apart, separated by the Atlantic Ocean, the Channel and the Irish Sea), and there is always a  
298 difference of around 0.5 Bq.L<sup>-1</sup> compared to the Thonon Les Bains data. Because the GNIP data stop  
299 in 2002 (Brest), 2012 (Thonon Les Bains, Ponta Delgada, Valentia), and 2015 (Madrid), we used a  
300 model inspired by Jean-Baptiste et al. (2019), based on a sine function fitted to the most recent data  
301 from Thonon Les Bains and Valentia to simulate the data for each site up to 2019 so that we could  
302 compare it with our measurements. This model is the sum of two components, the first corresponding  
303 to the exponentially decreasing mean value of tritium in rainwater over time, and the second  
304 describing seasonal variability, with an amplitude that diminishes over time (see Jean-Baptiste et al.  
305 (2019), for more details).

306 Figure 9 presents the data simulated on the basis of the models for the Thonon Les Bains and Valentia  
307 sites, together with our measurements taken in 2018-2019 on the Cherbourg-LRC and Pontrieux sites.  
308 The Valentia measurements are shown on the graph up to 2012, to observe the agreement between the  
309 model and the measurements. It can be seen that the measurements from our reference site at  
310 Pontrieux appear to be similar (or even lower than) to the measurements at the Valentia site in the case  
311 of the lowest activities. However, the few highest activities on the Pontrieux site ( $\approx 1$  Bq.L<sup>-1</sup> on 3  
312 occasions) are higher than those given by the model as a maximum. This reflects the continental

313 influence in certain periods, as explained in section 3.2; this influence should not exist in Valentia.  
314 The measurements taken at Cherbourg-LRC are rather similar to the simulated measurements for the  
315 Thonon Les Bains site than that of Valentia, but the amplitude is greater, and higher values ( $> 1 \text{ Bq.L}^{-1}$ )  
316 on several occasions bear witness to the sporadic impact of the nearby nuclear facilities (though in  
317 80% of cases the Cherbourg-LRC measurements are consistent with the model's predictions for  
318 Thonon Les Bains). Furthermore, over the period from 02 to 04/07/2018, we took 3 individual samples  
319 of rainwater from a site in central France (Lusignan,  $46.416^{\circ}\text{N}$ ,  $0.1188^{\circ}\text{E}$ , Figure 1a) with a mean  
320 result of  $1.11 \pm 0.16 \text{ Bq.L}^{-1}$  (Figure 9, green dots); these 3 measurements coincide well with the model  
321 for Thonon les bains.

322 The different comparisons show that the Thonon Les Bains site may be affected by diffuse tritium  
323 releases from nuclear facilities because, when we see the measurements (Figures 7 and 8) the site  
324 always has slightly higher activities than coastal sites and especially larger variations. The prevailing  
325 winds come mainly from the south-west and north-east at this site, and there are a number of nuclear  
326 sites less than 200 km away to the south-west (several NPPs, the CEA site at Marcoule); moreover,  
327 even when the prevailing winds are not blowing, the CEA's nuclear site at Valduc, which is the largest  
328 emitter of gaseous tritium in France, is only 140 km to the north-west and can probably sporadically  
329 affects the tritium content of the air and rainwater in this region. In view of our measurements at  
330 Pontrieux and the simulations for the Valentia site, a site on the Atlantic coast in western France  
331 would certainly show lower tritium levels in rainwater than at Thonon Les Bains and would be of  
332 interest. The Cherbourg site, although under the potential influence of many nuclear sites, nevertheless  
333 does not, with rare exceptions, give very high measurements for tritium content in rainwater, and the  
334 measurements appear comparable with the simulations for Thonon Les Bains, which would not have  
335 seemed obvious before this work.

336 Several recent studies in different countries throughout the world have recorded tritium activities in  
337 rainwater in the last few years. For European sites in Germany, Romania and Ukraine (Berlin,  
338 Stuttgart, Regensburg, Vienna, Krakow, Rm.Valcea), Dului et al. (2018) give mean annual values of  
339  $1.1$  to  $1.4 \text{ Bq.L}^{-1}$ , with a maximum between May and August corresponding to the presence of the  
340 spring leak mechanism. They also indicate a decrease of around 1.4% per year in background levels in  
341 rainwater since 2000. For a site in Hanoi (at a latitude of  $21^{\circ}\text{N}$ ) in the period 2011-2016, based on  
342 monthly measurements, Anh et al. (2018) give values ranging from  $0.06$  to  $0.83 \text{ Bq.L}^{-1}$ , with a mean of  
343  $0.30 \pm 0.15 \text{ Bq.L}^{-1}$ , showing that the site is influenced by the tropical climate and particularly by the  
344 effect of moisture from the sea. Tadros et al. (2014) did a comprehensive study of tritium in rainwater  
345 in Australia during the period 1963-2011. They give lower tritium values in rainwater on coastal sites,  
346 with less marked seasonal variations than on inland sites. For the period 2005-2011, the authors give a  
347 range of values from  $0.10$  (northern Australia) to  $0.38 \text{ Bq.L}^{-1}$  (southern Australia, Melbourne). Harms  
348 et al. (2016) give measurements in California (USA) for 18 sites, including coastal sites near the

349 Pacific Ocean and more continental sites. The mean value for the coastal sites is  $0.23 \pm 0.04 \text{ Bq.L}^{-1}$ ,  
350 while for the other sites the values range from 0.5 to  $0.66 \text{ Bq.L}^{-1}$ . Our results are consistent with these  
351 various studies. Finally, it could be note in the literature the very particular data of Chae and Kim  
352 (2018), who took measurements in rainwater around the Wolsong nuclear power plant site in Korea,  
353 which has four pressurized heavy water reactors and two pressurized water reactors. The activities in  
354 rainwater vary from 0.3 to  $1090 \text{ Bq.L}^{-1}$  and the least affected site presents activities of 0.8 to  $10 \text{ Bq.L}^{-1}$   
355 with a mean of  $2.3 \text{ Bq.L}^{-1}$ . It is worth noting the difference in magnitude of the values linked to the  
356 presence of pressurized heavy water reactors. Finally, mention should be made of recent work done at  
357 Corte ( $42.300570^\circ\text{N}$ ,  $9.148592^\circ\text{E}$ ) on the island of Corsica (France) in the Mediterranean Sea,  
358 between 18/04/2017 and 04/04/2018 on 46 rainfall episodes. Juhlke et al. (2019) give a range of  
359 activities from 0.19 to  $1.19 \text{ Bq.L}^{-1}$ , with a mean of  $0.53 \text{ Bq.L}^{-1}$ , which are very similar values to those  
360 measured on our sites at Coutances and Saint-Lô in particular.

361

## 362 **5. Conclusion**

363 We measured tritium activity in rainwater at 5 sites in north-west France between October 2017 and  
364 May 2019. The aim was to provide recent data for a reference site (Pontrieux) most of the time  
365 influenced by marine air masses from the Atlantic Ocean, and for several sites in the Manche  
366 department in north-west France, where several major nuclear facilities are located. The activities  
367 measured were compared with IAEA-GNIP data, using measured data and simulations from a model  
368 to complete the data sets up to 2019. On our reference site (Pontrieux), the activities were often below  
369 the decision threshold ( $<0.15 \text{ Bq.L}^{-1}$ ) and are comparable with the activities expected at GNIP stations  
370 with a marine influence (Valentia, Ireland; Ponta Delgada, Azores). The measurements taken in the  
371 south of the Manche department (Coutances, Saint-Lô) also show that, for most of the time, the  
372 nuclear facilities (approximately 80 km further north) have no impact. For the Cherbourg-LRC and  
373 Cherbourg-Town Center sites, which are closer to the Orano NC La Hague reprocessing plant  
374 (approximately 20 km East South east away), the activities are low overall (mean values of 1.00 and  
375  $1.19 \text{ Bq.L}^{-1}$  respectively) but these values probably bear witness both to the direct influence of air  
376 masses with a slightly higher tritium content (explained by rain pushed by winds from the Orano  
377 plant), and probably a continuous emission/exchange phenomenon at the interfaces with the  
378 surrounding sea, in which the tritium activity is higher than elsewhere. Despite this, the data from the  
379 Cherbourg-LRC site are with few exceptions comparable with data who can be predicted for the GNIP  
380 reference site in France at Thonon Les Bains. We also showed that a single episode of rain on a  
381 marked tritium air mass, even if it represents only 10% of the total volume of water from the sample  
382 for example, could significantly increase tritium activity. The precipitation amount effect could be  
383 shown on the Cherbourg LRC site, which is not the case on the Omonville-PTILH site, which is too

384 influenced by the Orano plant, with important variation of activities between  $<1$  and  $25 \text{ Bq.L}^{-1}$ , and a  
385 average activity of  $3.8 \text{ Bq.L}^{-1}$  for 48-hour period rain event.

386 This work also show that IAEA-GNIP data from the European stations nearest to France with a marine  
387 influence (those we worked with were Valentia and Punta Delgada) would be more relevant than the  
388 French IAEA site of Thonon Les Bains (with a sine function model for example) to correctly estimate  
389 tritium activities in rainwater for regions of France on the Atlantic coast. These activities are mostly  
390 less than  $0.2 \text{ Bq.L}^{-1}$ . This work, which compares periodic measurements on a few sites with sets of  
391 measurements taken over a long period of time, shows the interest of the IAEA-GNIP long-term data  
392 sets but also the need for more recent data than the existing data, the latest of which dates from 2012.  
393 Finally, our work shows that a reference station monitored regularly over the long term on France's  
394 west coast, bordering the Atlantic Ocean, would be very useful because a one-off study like ours is not  
395 enough to provide data for use in climate models.

396

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401

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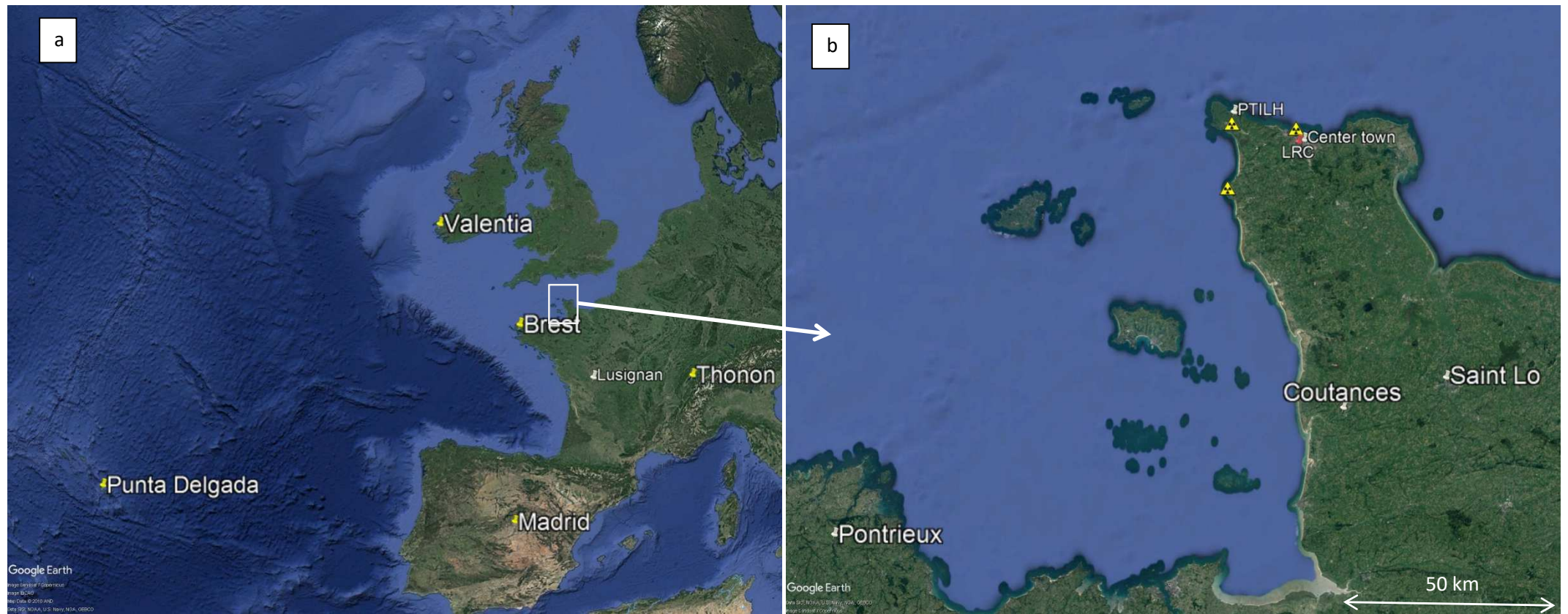


Figure 1 : (a) Location of the global network of isotopes precipitation (GNIP) stations (IAEA/WMO 2019) used in this work (yellow dots) and (b) location of local sampling site of our study (white and red dots). Positions of nuclear facilities were reported on (b).

Figure 2 : (a) Pluviometer Eigenbrodt used at Cherbourg–LRC site, (b) Pluviometer used on others sites.



Figure 3 : Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) at Cherbourg-LRC site (bi-Monthly measurements), at Pontrioux and Cherbourg Center Town sites (intermittent measurements). The dotted line represent the decision thresholds for our measurements (below its ‘uncensored’ values).

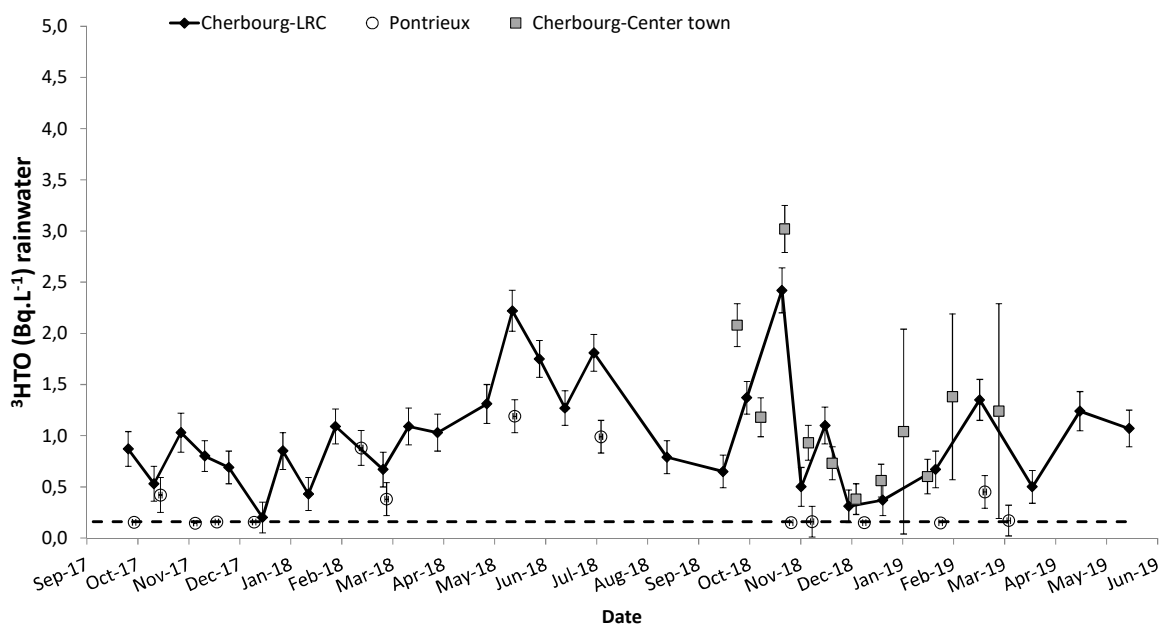


Figure 4 : Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) at Omonville La Petite PTILH site (the black bar represent activities  $< 1 \text{ Bq.L}^{-1}$ ).

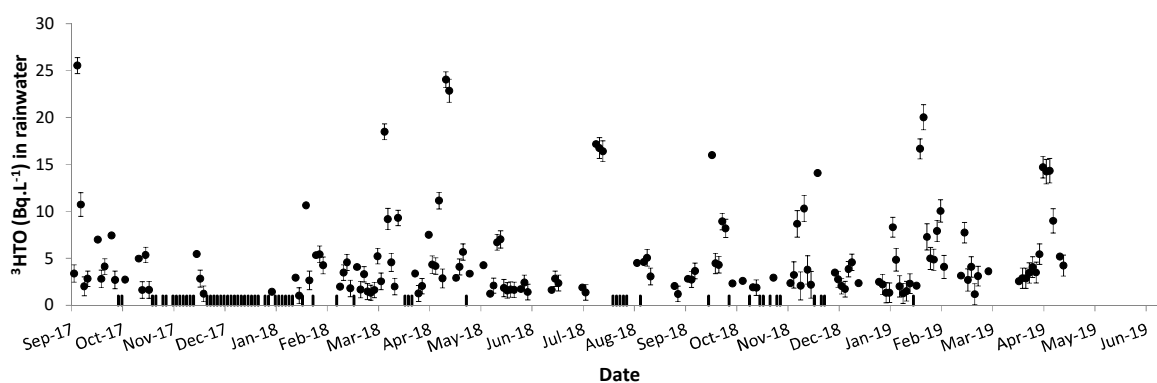


Figure 5: BoxPlots of  $^3\text{H}$  ( $\text{Bq.L}^{-1}$ ) in rainwater in our site of measurements. Cross represent median, The two ends of the rectangle represents 25-75% percentile, horizontal bar are average, circle marks min and max measurements (values of average are in italics, values of median are in bold).

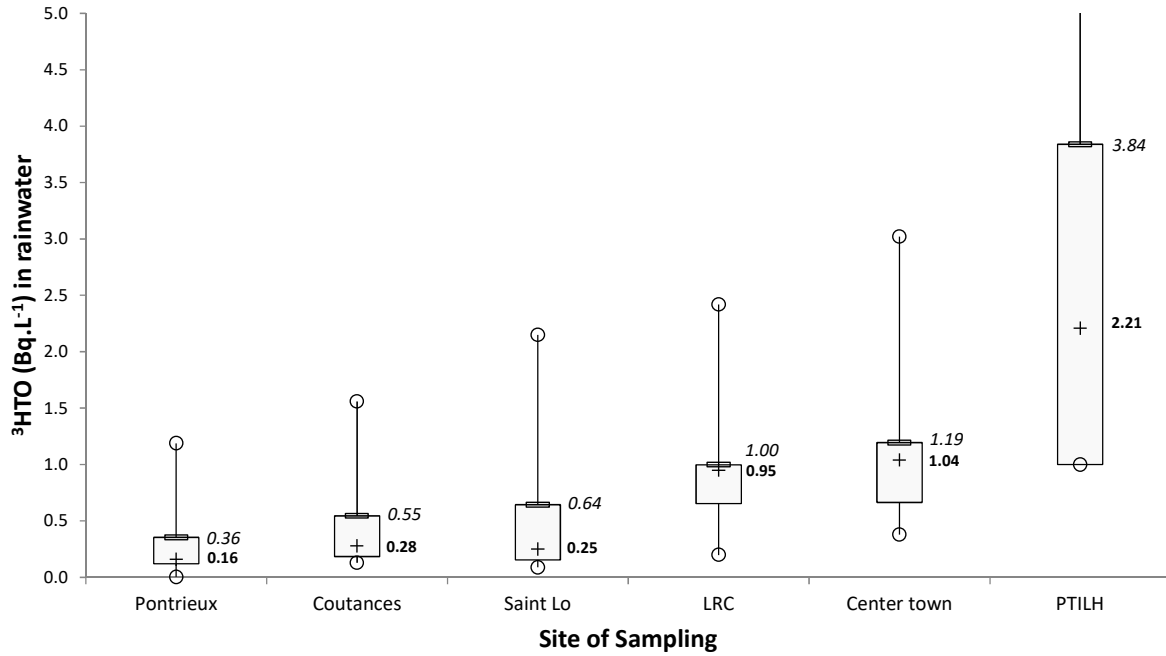


Figure 6 : Normalised  $^3\text{H}$  in function of rainfall (mm) at Cherbourg-LRC site (a) and PTILH site (b).

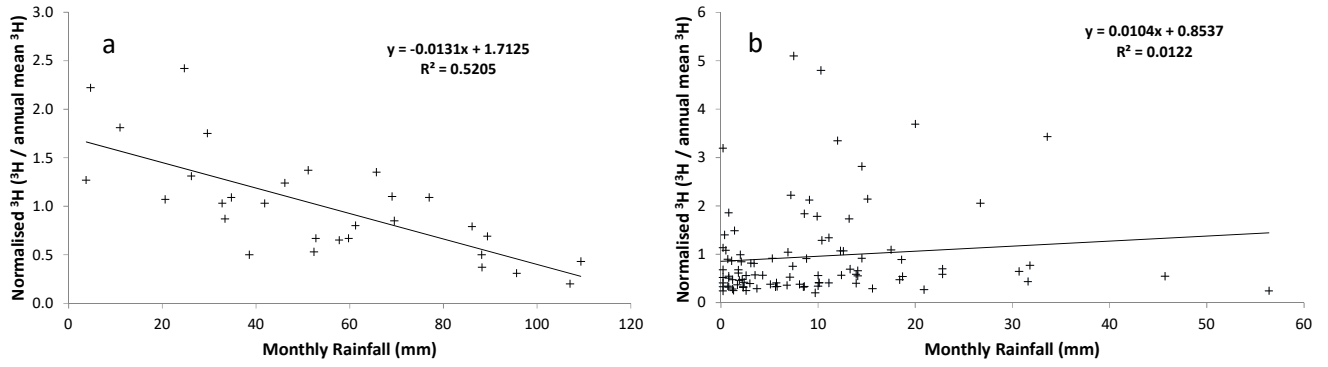


Figure 7 : Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) for IAEA-GNIP stations at Thonon Les Bains (France), Madrid (Spain), Brest (France), Valentia (Ireland), Punta Delgada (Portugal) for 1996-2016 period.

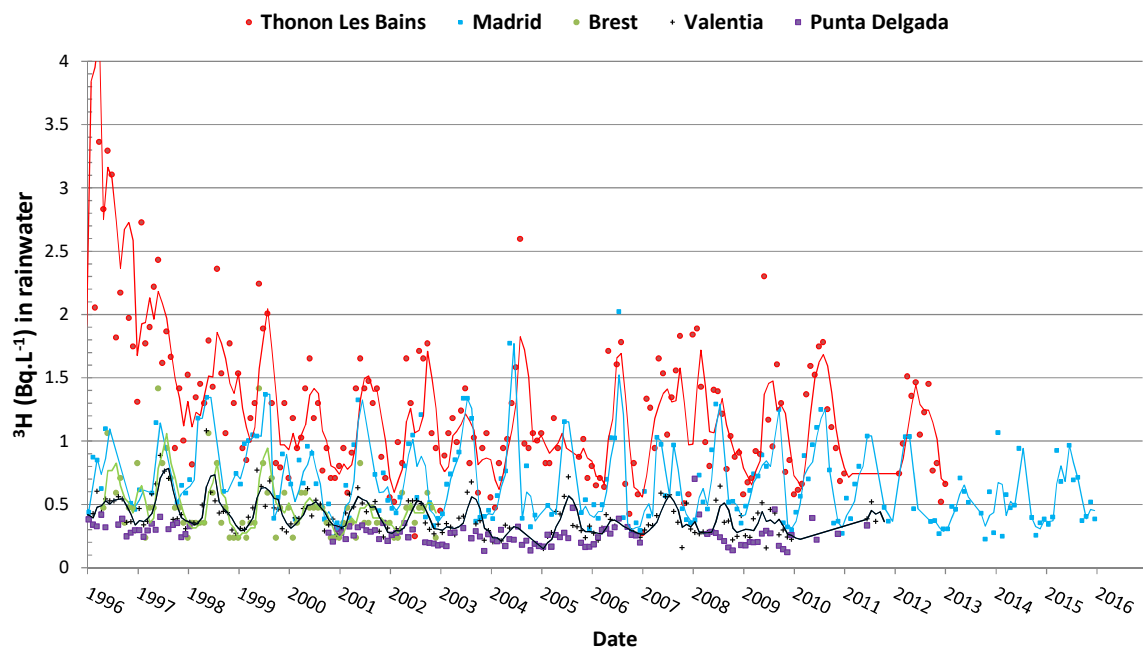


Figure 8: Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) for IAEA-GNIP stations at Thonon Les Bains (France), Brest (France), Valentia (Ireland) for 1996-2005 period.

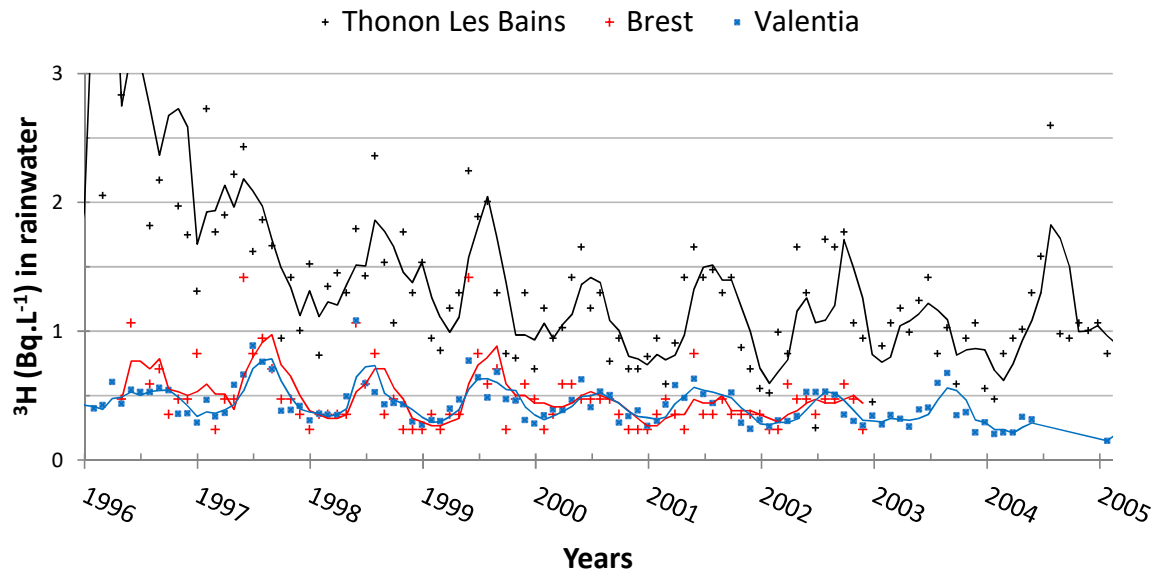


Figure 9 : Simulation by sine function of tritium activities in rainwater ( $\text{Bq}\cdot\text{L}^{-1}$ ) for IAEA-GNIP stations at Thonon Les Bains and Valentia between 2007 and 2020, with measurements at Valentia (until 2012), and our measurements in this work at Pontrieux (square), Cherbourg-LRC (black circles) and Lusignan (green circles) sites.

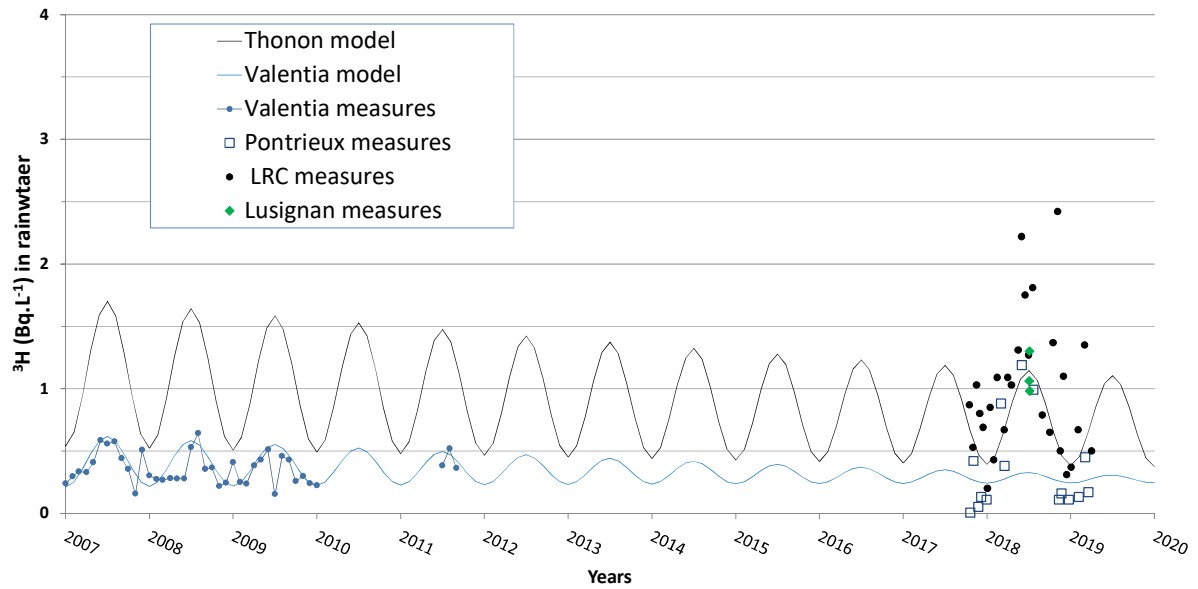




Table 1 : Tritium activities in rainwater ( $\text{Bq.L}^{-1} \pm 2\sigma$ ) at Pontrieux site, and origin of air masses during 72h before sampling (values lower than  $0.15 \text{ Bq.L}^{-1}$  are values  $< \text{SD}$  and indicate here to calculate average).

date start (TU)	date end (TU)	$^3\text{H Bq.L}^{-1}$		Air mass origin	remarks
19/10/2017 14:30	19/10/2017 17:15	0.01	$\pm 0.16$	SSW-Atlantic	< SD
03/11/2017 17:00	04/11/2017 07:00	0.42	$\pm 0.17$	SE-N-Channel	
23/11/2017 15:30	24/11/2017 18:15	0.05	$\pm 0.15$	W-Atlantic	< SD
06/12/2017 17:00	07/12/2017 11:00	0.13	$\pm 0.15$	SW-Atlantic	< SD
28/12/2017 18:00	29/12/2017 09:00	0.11	$\pm 0.16$	W-Atlantic	< SD
01/03/2018 17:00	02/03/2018 08:00	0.88	$\pm 0.17$	SE-land	
15/03/2018 17:00	17/03/2018 07:00	0.38	$\pm 0.16$	SE-land	
31/05/2018 12:00	31/05/2018 16:00	1.19	$\pm 0.16$	S-land	
21/07/2018 06:00	21/07/2018 10:00	0.99	$\pm 0.16$	W-S-land	
09/11/2018 17:00	10/11/2018 10:00	0.11	$\pm 0.15$	W-Atlantic	< SD
21/11/2018 07:30	22/11/2018 17:00	0.16	$\pm 0.15$	E-land	
22/12/2018 16:00	23/12/2018 09:00	0.11	$\pm 0.15$	SW-Atlantic	< SD
05/02/2019 14:00	06/02/2019 08:00	0.13	$\pm 0.15$	SW-Atlantic	< SD
03/03/2019 16:00	04/03/2019 09:00	0.45	$\pm 0.16$	NW-Channel	
13/03/2019 16:00	18/03/2019 09:00	0.17	$\pm 0.15$	N-Channel	

Table 2: Tritium activities in rainwater ( $\text{Bq.L}^{-1} \pm 2\sigma$ ) at Coutances, Saint-Lô and Cherbourg-Center town site (values lower than  $0.15 \text{ Bq.L}^{-1}$  are values  $< \text{SD}$  and indicate here to calculate average).

site	date start (TU)	date end (TU)	$^3\text{H Bq.L}^{-1}$		remarks
Coutances	09/10/2018 12:30	18/10/2018 13:30	1.05	$\pm$ 1.05	
	18/10/2018 15:00	08/11/2018 15:00	1.56	$\pm$ 0.19	
	08/11/2018 15:00	26/11/2018 11:15	0.13	$\pm$ 0.16	< SD
	26/11/2018 11:15	06/12/2018 13:15	0.16	$\pm$ 0.16	
	06/12/2018 13:15	17/12/2018 10:35	0.28	$\pm$ 0.14	
	17/12/2018 10:35	07/01/2019 10:30	0.43	$\pm$ 0.16	
	07/01/2019 10:30	22/01/2019 13:10	0.21	$\pm$ 0.16	
	22/01/2019 13:10	04/02/2019 08:25	< 1		
	04/02/2019 08:25	25/02/2019 10:00	< 1		
25/02/2019 10:00	25/03/2019 06:00	< 1			
Saint-Lô	11/10/2018 06:25	19/10/2018 10:50	1.36	$\pm$ 0.20	
	19/10/2018 15:00	06/11/2018 15:00	2.15	$\pm$ 0.20	
	06/11/2018 08:50	26/11/2018 07:15	0.19	$\pm$ 0.16	
	26/11/2018 07:00	10/12/2018 07:00	0.09	$\pm$ 0.15	< SD
	10/12/2018 07:00	21/12/2018 11:00	0.12	$\pm$ 0.15	< SD
	21/12/2018 11:00	08/01/2019 07:00	0.35	$\pm$ 0.16	
	08/01/2019 07:00	28/01/2019 09:00	0.25	$\pm$ 0.16	
28/01/2019 09:00	04/03/2019 08:00	< 1			
Cherbourg-Center town	25/09/2018 15:00	09/10/2018 14:00	2.08	$\pm$ 0.21	
	09/10/2018 14:00	23/10/2018 13:00	1.18	$\pm$ 0.19	
	23/10/2018 15:00	06/11/2018 15:00	3.02	$\pm$ 0.23	
	06/11/2018 15:00	20/11/2018 12:00	0.93	$\pm$ 0.17	
	20/11/2018 12:00	04/12/2018 15:00	0.73	$\pm$ 0.16	
	04/12/2018 15:00	18/12/2018 15:00	0.38	$\pm$ 0.15	
	18/12/2018 15:00	02/01/2019 09:00	0.56	$\pm$ 0.16	
	02/01/2019 09:00	15/01/2019 15:00	1.04	$\pm$ 1.00	
	15/01/2018 15:00	29/01/2019 15:00	0.60	$\pm$ 0.17	
29/01/2019 15:00	13/02/2019 08:30	1.38	$\pm$ 0.81		
24/02/2019 15:00	12/03/2019 15:00	1.24	$\pm$ 1.05		

Figure 1: (a) Location of the global network of isotopes precipitation (GNIP) stations (IAEA/WMO 2019) used in this work (yellow dots) and (b) location of local sampling site of our study (white and red dots). Positions of nuclear facilities were reported on (b).

Figure 2: (a) Pluviometer Eigenbrodt used at Cherbourg–LRC site, (b) Pluviometer used on others sites.

Figure 3: Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) at Cherbourg-LRC site (bi-Monthly measurements), at Pontrieux and Cherbourg Center Town sites (intermittent measurements). The dotted line represent the decision thresholds for our measurements (below its ‘uncensored’ values).

Figure 4: Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) at Omonville La Petite PTILH site (the black bar represent activities  $< 1 \text{ Bq.L}^{-1}$ ).

Figure 5: BoxPlots of  $^3\text{H}$  ( $\text{Bq.L}^{-1}$ ) in rainwater in our site of measurements. Cross represent median, The two ends of the rectangle represents 25-75% percentile, horizontal bar are average, circle marks min and max measurements (values of average are in italics, values of median are in bold).

Figure 6: Normalised  $^3\text{H}$  in function of rainfall (mm) at Cherbourg-LRC site (a) and PTILH site (b).

Figure 7: Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) for IAEA-GNIP stations at Thonon Les Bains (France), Madrid (Spain), Brest (France), Valentia (Ireland), Punta Delgada (Portugal) for 1996-2016 period.

Figure 8: Evolution of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) for IAEA-GNIP stations at Thonon Les Bains (France), Brest (France), Valentia (Ireland) for 1996-2005 period.

Figure 9: Simulation by sine function of tritium activities in rainwater ( $\text{Bq.L}^{-1}$ ) for IAEA-GNIP stations at Thonon Les Bains and Valentia between 2007 and 2020, with measurements at Valentia (until 2012), and our measurements in this work at Pontrieux (square), Cherbourg -LRC (black circles) and Lusignan (green circles) sites.

Table 1: Tritium activities in rainwater ( $\text{Bq.L}^{-1} \pm 2\sigma$ ) at Pontrieux site, and origin of air masses during 72h before sampling (values lower than  $0.15 \text{ Bq.L}^{-1}$  are values  $< \text{SD}$  and indicate here to calculate average).

Table 2: Tritium activities in rainwater ( $\text{Bq.L}^{-1} \pm 2\sigma$ ) at Coutances, Saint-Lô and Cherbourg-Center town site (values lower than  $0.15 \text{ Bq.L}^{-1}$  are values  $< \text{SD}$  and indicate here to calculate average).