Determination and speciation of anthropogenic tritium in the Loire River estuary (France).

O. Péron, Pastor L., C. Gégout, Elise Fourré, F. Siclet, Gilles F Montavon, Catherine Landesman

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O. PÉRON[1]*, L. PASTOR[2], C. GÉGOUT[1], E. FOURRÉ[3], F. SICLET[2], G. MONTAVON[1], C. LANDESMAN[1]
[1]SUBATECH, UMR 6457, 4 rue Alfred Kastler, 44307 Nantes, France (*corresponding: **olivier.peron@subatech.in2p3.fr**)
[2]EDF, LNHE, 6 quai Wattier 78400 Chatou Cedex, France
[3]LSCE, Orme des Merisers, CEN Saclay, 91191 Gif sur Yvette Cedex, France

Context & objectives

The aim of radioecology is to understand the transfer of radionuclides through the ecosystem. It relies strongly on field studies which can provide useful informations on the presence of radionuclides in the environment, and their origins (natural and anthropogenic). In this study, the radioactive isotope of hydrogen, *i.e.* tritium (^{3}H or T), is considered. Tritium is a beta emitter with a radioactive half life of 12.3 years. Environmental tritium occurs in three main forms: tritiated water (HTO or tissue free water), organically bound tritium (OBT) and tritiated gas (HT). Tritiated water is the most abundant chemical form of tritium in the aquatic and terrestrial environment. OBT can be subdivided in two fractions: the exchangeable OBT refers to tritium atoms that are easily exchanged (*e.g.* bound to nitrogen, oxygen or sulfur atoms), while the non-exchangeable OBT refers to the OBT covalently bound to carbon atoms. Because of its longer residence time in plants and living organisms, the radiological significance of non-exchangeable OBT is greater than that of HTO. In this study, mud and water samples from the Loire estuary, downstream from several nuclear power plants, were analyzed.

Location of the environmental samplings

Figure 1: Stations of the mud samplings in the Loire estuary

Figure 2: Transect of the surface water samplings in the Loire estuary

Figure 3: Nuclear power plants in France and location of the studied area

Samplings, treatments & analyses

Mud samples were recovered from the river floor with a Van Veen grab (Figure 1) and water samples were continuously pumped from the river surface during the transect (Figure 2). The environmental samples were freezed (-28°C) for an efficient storage.

In order to recover, free HTO and total OBT, mud samples were freeze-dryed (10^{-3} mbar, - 57°C) and burnt (combustion) respectively. In order to recover HTO, water samples were distilled.

The specific ^{3}H activities of the combustion and free waters were measured by liquid scintillation counting using Ultima Gold LLT cocktails (PerkinElmer Tri-carb 3170 TR/SL). The results were corrected for quenching effects. The overall uncertainty value is ± 15% (2σ).

Observations & results

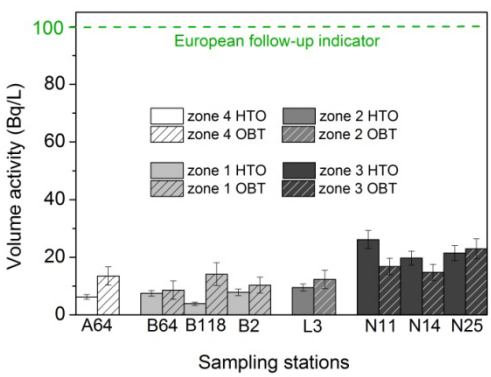

Figure 4: tritium volume activity in HTO and OBT forms in mud samples

HTO and total OBT activities from mud samples range from 4 to 26 Bq.L^{-1} and from 10 to 25 Bq.L^{-1}, respectively (Figure 4). The European follow-up indicator is 100 Bq.L^{-1}. The average ^{3}H volume activities in precipitation over the continental surface in the Northern hemisphere was estimated to be < 3 Bq.L^{-1} in 2012.

HTO activities of distilled surface water samples decrease with increasing salinity from 21 to 7 Bq.L^{-1} (Figure 5). These results are consistent with the values previously obtained by F. Siclet *et al.* (2001).

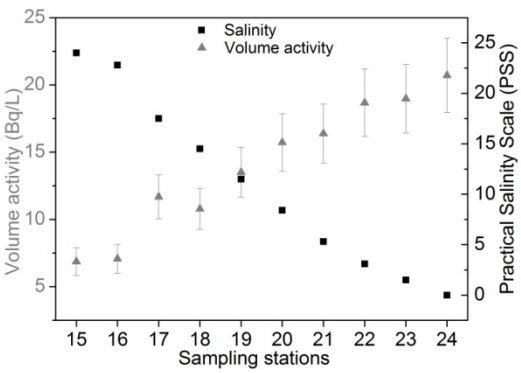

Figure 5: tritium volume activity and salinity of surface water samples

Perspectives & fundamental aspect

To estimate the non-exchangeable OBT activity in these samples (eq. 1), the exchangeable fraction of hydrogen (α coefficient) within the matrices has to be known (eq. 2). A dedicated experimental set up was thus developed. It consists in a temperature and humidity controlled glove box where different environmental matrices are exposed to specific atmospheres with fixed T/H vapour pressure ratios. The calibration phase of the method is being performed using a α-cellulose matrix with D/H (deuterium) vapour pressure ratios set by water from the station Concordia at Dome C in Antarctica.

$$\alpha \leftarrow \frac{\left(\frac{T}{H}\right)_{atmosphere}}{\left(\frac{T}{H}\right)_{sample}} \quad (eq.\,2)$$

$$\left(\frac{T}{H}\right)_{NE/OBT} = \frac{\left(\frac{T}{H}\right)_{tot/OBT} - \alpha \times \left(\frac{T}{H}\right)_{HTO}}{1-\alpha} \quad (eq.\,1)$$

F. Siclet et al., Hydroecol. Appl. 2001, Transfer in the Loire river basin down to the estuary of nuclear power plants radioactive discharges

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