



Tritium and ^{14}C background levels in pristine aquatic systems and their potential sources of variability



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ABSTRACT

Tritium and ^{14}C are currently the two main radionuclides discharged by nuclear industry. Tritium integrates into and closely follows the water cycle and, as shown recently the carbon cycle, as does ^{14}C (Eyrolle-Boyer et al., 2014a, b). As a result, these two elements persist in both terrestrial and aquatic environments according to the recycling rates of organic matter. Although on average the organically bound tritium (OBT) activity of sediments in pristine rivers does not significantly differ today (2007–2012) from the mean tritiated water (HTO) content on record for rainwater (2.4 ± 0.6 Bq/L and 1.6 ± 0.4 Bq/L, respectively), regional differences are expected depending on the biomass inventories affected by atmospheric global fallout from nuclear testing and the recycling rate of organic matter within watersheds. The results obtained between 2007 and 2012 for ^{14}C show that the levels varied between 94.5 ± 1.5 and 234 ± 2.7 Bq/kg of C for the sediments in French rivers and across a slightly higher range of 199 ± 1.3 to 238 ± 3.1 Bq/kg of C for fish. This variation is most probably due to preferential uptake of some organic carbon compounds by fish restraining ^{14}C dilution with refractory organic carbon and/or with old carbonates both depleted in ^{14}C . Overall, most of these ranges of values are below the mean baseline value for the terrestrial environment (232.0 ± 1.8 Bq/kg of C in 2012, Roussel-Debet, 2014a) in relation to dilution by the carbonates and/or fossil organic carbon present in aquatic systems. This emphasises yet again the value of establishing regional baseline value ranges for these two radionuclides in order to account for palaeoclimatic and lithological variations. Besides, our results obtained from sedimentary archive investigation have confirmed the delayed contamination of aquatic sediments by tritium from the past nuclear tests atmospheric fallout, as recently demonstrated from data chronicles (Eyrolle-Boyer et al., 2014a,b). Thus Sedimentary archives can be successfully used to reconstruct past ^{14}C and OBT levels. Additionally, sediment repositories potentially represent significant storages of OBT that may account for in case of further remobilisation. We finally show that floods can significantly affect the OBT and ^{14}C levels within suspended particles or sediments depending on the origin of particles reinforcing the need to acquire baseline value range at a regional scale.

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1. Introduction

Tritium ($T_{1/2} = 12.3$ years) and ^{14}C ($T_{1/2} = 5730$ years) both have a naturally occurring cosmogenic source that is generated by the absorption of neutrons of different energies by nitrogen atoms at

various levels of the atmosphere. These radioactive isotopes were also artificially introduced into the environment during past nuclear testing and are still legally discharged by the nuclear industry in liquid and gaseous effluents. Surface nuclear tests conducted between 1945 and 1980 increased atmospheric tritium levels by three orders of magnitude (UNSCEAR, 2000) and those of ^{14}C by a factor of around two (Levin and Hesshaimer, 2000).

Tritium and ^{14}C are currently the two main radionuclides discharged by France's nuclear facilities (Gontier and Siclet, 2011). For

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example, more than 80% of the unbound ^3H activity present in the downstream waters of the Rhône River is believed to come from discharges from the nuclear industry (Eyrolle-Boyer et al., 2013).

Unlike most trace elements for which transfers within the environmental compartments are governed by balances at their interfaces (Kd, transfer coefficient), tritium and ^{14}C integrate into and closely follow the water and carbon cycles. As a result, ^{14}C and tritium are present in organic and inorganic form, as is the case with stable carbon and hydrogen.

The organic forms of tritium and ^{14}C produced during photosynthesis persist in living or detritic organic matter in both terrestrial and aquatic environments according to the growth and degradation process of the plant matter (biomass). Although tritiated water (HTO) and the inorganic forms of ^{14}C (associated with carbonates and hydrogen carbonates) follow the dynamics of water mass, the organic forms – in particular NE-OBT (non-exchangeable organically bound tritium) and ^{14}C integrated with living or detritic organic matter – are highly dependent on the biological cycles (Eyrolle-Boyer et al., 2014a,b).

Inorganic and organic compounds depend from one each others as abiotic and biological compartments are straight linked together. The inorganic forms contribute to the formation of plant mass during photosynthesis. These producers are eaten by primary consumers (members of fauna and microfauna), which themselves are eaten by upper-level predators, such as herbivorous, omnivorous and carnivorous fishes in the case of aquatic ecosystems. This is how these two radionuclides are ubiquitously present in the various abiotic and biological environmental components.

In order to be able to constrain the results of radiological assessments of the environmental impact of nuclear energy, it is essential to know the characteristic values of the baseline levels (geochemical backgrounds) of HTO, OBT and ^{14}C in both terrestrial and aquatic environments. This paper presents data sets obtained over the last decades in pristine French rivers where such background values can be acquired. These values are discussed in a historical and regional context to emphasize the variability factors. Additionally we present original data acquired on a sedimentary archive collected in a pristine area in the north eastern France that reinforce our data chronicles. Finally we present results obtained during a flood event at the lower course of nuclearised large river in order to underline the role of particle origins on OBT and ^{14}C activity levels.

2. Updating of current knowledge on tritium and ^{14}C behaviour in the environment

Tritium (^3H) is a radioisotope of hydrogen. By emitting a low-energy β -particle, it decays into stable helium-3 (He-3), with a radioactive half-life of 12.32 years. The behaviour of this radioactive isotope in the environment has been widely studied since the 1950s, both through laboratory experiments and, more recently, through field observations (e.g., Kirchmann et al., 1979; Belot, 1986; McCubbin et al., 2001; Kim et al., 2012; Fievet et al., 2013; Baglan et al., 2013; Jean-Baptiste and Fourné, 2013). Because it is an isotope of hydrogen, tritium has all the chemical properties of hydrogen. These properties determine its state and environmental fate as a component of the water molecule and a component of the organic molecules. As a result, ^3H is present in the environment mainly in the form of tritiated water (HTO) or incorporated into plant or animal organic matter (organically bound tritium, or OBT). The activity of tritium can be expressed in Bq/L of water in all environmental components, be they rainwater, surface water (rivers, lakes), ground water, seawater or water extracted from organic matter (plants and animals). Water extracted by calcination after freeze-drying of biological matrices corresponds to tissue

water (easily exchangeable with water bound, weak hydrogen bonds) and to atoms of hydrogen extracted from the organic structures of the matrices (non-exchangeable with bound water). The former is conventionally defined as exchangeable organically bound tritium (E-OBT) and the latter is conventionally defined as non-exchangeable organically bound tritium (NE-OBT). For example, in terrestrial environments, E-OBT is the form that will be exchanged during evapotranspiration. NE-OBT is initially generated during photosynthesis and persists in the environment by following the biodegradation cycle of organic matter, as is the case with ^{14}C . Today, it is analytically impossible to reproducibly measure these two species in the various environmental matrices (ASN, 2011). Lastly, tritium can be observed in more detail in a form associated with synthetic organic molecules (e.g., paint residues) known as technogenic tritium (Croudace et al., 2012). The organic matter present in the suspended or settled particles of continental aquatic systems (waterways, rivers and lakes) is brought in from watersheds by erosion and runoff of degradation products contained in the soils (allogenic matter) and the primary production developed in situ (autogenic matter). Tritium bound to allogenic organic matter comes from the degradation of the terrestrial biomass. This species thus provides a picture of the historical contamination levels of the terrestrial biomass. Provided the age of this organic matter and/or its recycling period within the watershed is known, it is possible to reconstruct past levels and vice versa. On the other hand, tritium bound to autogenic organic matter is in balance with the HTO of the waterway during its growth.

The allogenic organic matter within rivers transfers contamination from the terrestrial environment. Thus it is inert with regard to the uptake of HTO present in river freshwaters (detritic matter). By contrast, autogenic organic matter integrates the HTO in waterways besides that released in liquid effluents discharged from nuclear facilities. This is a rather theoretical scheme as allogenic OM (Organic Matter) will be colonized once in the water by aquatic organisms (microbes, invertebrates) which will contain autogenic OM, so the distinction is expected to become moot. Indeed, the two pools of organic matter (allogenic/autogenic) coexist in aquatic systems in varying proportions depending on their intrinsic typology and characteristics. This explains why waterways and rivers are primarily subject to inputs of organic matter from watersheds whereas canals and lakes generally have higher proportions of autogenic matter. Contamination of organic suspended particles and sediments in surface aquatic systems results from both the allogenic/autogenic ratio and the contamination level of these two pools of organic matter. Contaminated autochthonous compounds produced, for example, in rivers downstream of liquid radioactive releases, are expected to be largely and rapidly diluted by the allogenic non-contaminated organic compounds from the watershed (if not contaminated by recent or past atmospheric inputs), whether in the flow of suspended particles or in settled particles (sediments). The proportion of autogenic carbon is highly limited in most rivers. It generally decreases from upstream to downstream mainly because of the increase in the concentration of suspended particles.

Inversely, within watersheds affected by several decades of atmospheric releases, contaminated allogenic organic compounds progressively transfer tritium and ^{14}C contamination to rivers depending on the biomass degradation rate. The most persistent species (lignin, cellulosic compounds, refractory organic matter in soils, clay–humic complexes, etc.) will be degraded long after their contamination and their impact on rivers will persist for long time as recently demonstrated (Eyrolle-Boyer et al., 2014a,b).

According to this schema, efficient transfer of contaminated organic compounds by tritium and/or ^{14}C to living species is

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