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Atmospheric tritium concentrations under influence of AREVA NC La Hague reprocessing plant (France) and background levels

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ABSTRACT

In-air tritium measurements were conducted around the AREVA NC La Hague reprocessing plant, as well as on other sites that are not impacted by the nuclear industry in northwest of France. The results indicate that the dominant tritium form around the AREVA site is HT (86%). HT and HTO levels are lower than 5 and 1 Bq. m^{-3} for hourly samples taken in the plume. No tritiated organic molecules (TOM) were detected. 26 measurement campaigns were performed and links were established between near-field ^{85}Kr, HT and HTO activities. Environmental measurements are in line with those taken at the discharge stack, and tend to demonstrate that there are no rapid changes in the tritium forms released. Out of the influence of any nuclear activities, the levels measured were below 13 $mBq.m^{-3}$ for HT and 5 $mBq.m^{-3}$ for HTO (<0.5 Bq. L^{-1}). HTO level in air seems to be influenced by HTO activities in surrounding seawater.

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1. Introduction

Tritium (^3H) is a low energy β-emitter radionuclide with a half-life of 12.3 years. Tritium production originated from naturally-occurring processes from the interaction of gases and cosmic radiation lead to a global inventory of 2.65 kg y^{-1} which corresponds to 9.6 10^5 TBq.y^{-1} (UNSCEAR, 2000). ^3H is mainly released into the environment by nuclear power plants, military nuclear facilities and nuclear reprocessing plants (CNSC, 2009; ASN, 2010). Atmospheric tests of nuclear weapons conducted by different states between 1945 and 1980 led to estimated release of 668 kg, which corresponds to 242,10^6 TBq during this period (UNSCEAR, 2000; Okada and Momoshima, 1993). Most of the ^3H released by these nuclear explosions were in hydrogen gas (HT) and methyl tritium gas (CH_3T) forms, which oxidized into HTO before being extracted from the atmosphere through precipitation. At a global level, current gaseous release for all nuclear reactors is estimated at approximately 6000 TBq.y^{-1}, with reprocessing plants representing approximately 300 TBq.y^{-1}. In France, gaseous ^3H release for the nuclear power plant is estimated at between 23 and 50 TBq.y^{-1} (IRSN, 2016). In 2014, the AREVA NC fuel reprocessing plant released 78 TBq into the atmosphere (AREVA, 2015), representing approximately 25% of French release. The remaining different industries and research centers in France release approximately 400 TBq.y^{-1}, nearly 65% of which is attributed to the Valduc and Marcoule centers (IRSN, 2016). As a comparison, at the end of the 2000s, a country such as Canada is estimated to have released 2700 TBq.y^{-1} of gaseous tritium, nearly 2000 of which was attributed to CANDU nuclear power plants (CNSC, 2009). In the future, the construction of new nuclear facilities (e.g. European Pressurized Reactor, International Thermonuclear Experimental Reactor ITER) as well as changes in nuclear fuel management may lead to an increase of ^3H releases from the nuclear industry in France. For example, ITER could release between 220 and 900 TBq.y^{-1} (Cortes, 2015) and we cannot exclude, on a global scale, the possibility of HT releases related to tritium production and underground tests by countries that are actively developing nuclear weapons programs.

Tritium can be readily exchanged with hydrogen atoms in the environment, thus entering the hydrological cycle, and then can enter in human/biological body through ingestion and inhalation (Belot, 1996). It is of fundamental interest to have the best possible knowledge on speciation and behavior in the environment for radiation dose assessment and transfers to terrestrial ecosystems (Maro et al., 2017). Recently, studies into tritium levels near nuclear areas were published, principally for Asiatic region (Kim et al.,

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1998; Momoshima et al., 2007; Fujita et al., 2007; Akata et al., 2011).

This study on atmospheric tritium levels (2014–2017) can be divided into four parts: The first important step involved obtaining improved data on background concentrations in France, which are not directly affected by industrial sites. Tritium is generally measured within the framework of regulatory monitoring and few very low level measurements are available. Even on a world scale, very few studies present very low level airborne tritium measurements excluding the influence of nuclear activities, with the exception of Japan (Okai and Takashima, 1991; Uda et al., 2008a, 2005; Okai et al., 1999; Momoshima et al., 2007; Akata et al., 2011). In a second phase, we worked closely with the AREVA NC nuclear fuel reprocessing plant, located in northwest of France, which is the largest reprocessing plant in the world. During shearing and dissolution activities, tritium is mainly released into the sea in liquid form, but is also released in gaseous form into the atmosphere. The total tritium-in-air activity measured around the plant by the operator (weekly average of <0.39 Bq. m^{-3}, AREVA, 2015) remains low compared with the authorized standards (weekly average of 8 Bq. m^{-3}, ministerial ruling of 10/01/2003, amended on 08/01/2007, AREVA, 2015) and the dose impact related to gaseous tritium releases is also extremely low (less than 1% of the dose linked to the whole gaseous release). Tritium concentration in the marine environment has already been measured in the vicinity of this site (e.g. Bailly du Bois et al., 2002, 2012), and it is now considered helpful to understand tritium behavior (forms, activity levels) in the atmosphere around this site, which is used as a research area for studies into transfers to ecosystems (Maro et al., 2017). The atmosphere is recognized as a leading input channel in transfer studies and as such it must be studied further (Eyrolle-Boyer et al., 2014; Mihok et al., 2016). The third and fourth parts of this project are not addressed in this paper: they will focus on possible changes in airborne HT forms and the long-distance dispersal of tritium and the resulting levels that are influenced by the plant over several hundred km.

In this paper, we discuss levels and ratios of the different forms, water vapor (HTO), hydrogen gas (HT), Tritium Organic Molecules (TOM) and methyl tritium (CH_3T) in the atmosphere measured in 2014–2016 in two background sites and near AREVA NC reprocessing plant.

2. Experimental

2.1. Sampling site for HT, HTO, CH_3T background levels

Two sites in northwest France were studied (Damgan (DA) and Aire de La Chaine (AC)), outside any zone of influence of nuclear facilities (Fig. 1a and Table 1). These sites are located at more than 100 km from the nearest nuclear facilities. DA is a coastal site and AC is a continental site. Samples were taken on three occasions (Table 1) with westerly winds from the Atlantic ocean (Fig. 2). The back trajectories in Fig. 2 indicate that the winds were in a west/south west direction for several days before the sample dates, thus providing an air mass that can be taken as an air background level. For comparison purposes, in June 2015 samples were also taken around a third site, Santec (SA), located in proximity to the English Channel (Fig. 1a, Table 1, Fig. 2d), during an outage period at the La Hague AREVA NC plant.

2.2. Sampling site near the AREVA NC La Hague nuclear reprocessing plant

20 sampling campaigns to measure HT, HTO, CH_3T under the influence of the AREVA NC La Hague plant were performed between May 2014 and June 2016 (Fig. 1b, Table 2). The nuclear fuel reprocessing plant, AREVA NC La Hague, is located on the La Hague peninsula in northwestern France. The industrial site is 2 km long by 500 m wide and is characterized by a dense tangle of buildings as much as a few tens of meters high. It has two production units (known as UP3 and UP2-800) located 200 m apart. Each unit is equipped with a 100-m-high stack. During nuclear fuel reprocessing, tritium is released into the atmosphere at the same time as ^{85}Kr for successive periods of 30–45 min. Other facilities at the plant may occasionally release tritium, but during our study, the samples were intentionally taken downwind from the main facilities UP2-800 and UP3. To perform sampling in the plume of the UP2-800 and/or UP3 stacks during the various shearing phases, ^{85}Kr measurements were performed in real-time (Connan et al., 2014). The objective of this work was to define measurement points in the stack plumes, using ^{85}Kr as a tracer, and take samples during activity peaks to measure tritium.

2.3. Sample collection

2.3.1. Sampling constraints

Campaigns with the objective of sampling air over the DA and AC sites for background levels necessitated checking weather forecasts to ensure the samples were taken in dry weather, with no risk of rain, and with westerly/south-westerly winds (from the Atlantic) prevailing over several days, which remove any influence coming from France. The samples were taken on the same day at both sites: firstly at the DA site, then the same air mass was sampled at AC a few hours later. For campaigns in the near-field AREVA NC plume, dry weather was also required, and it was even more important to ensure that one of the two UP3 or UP2-800

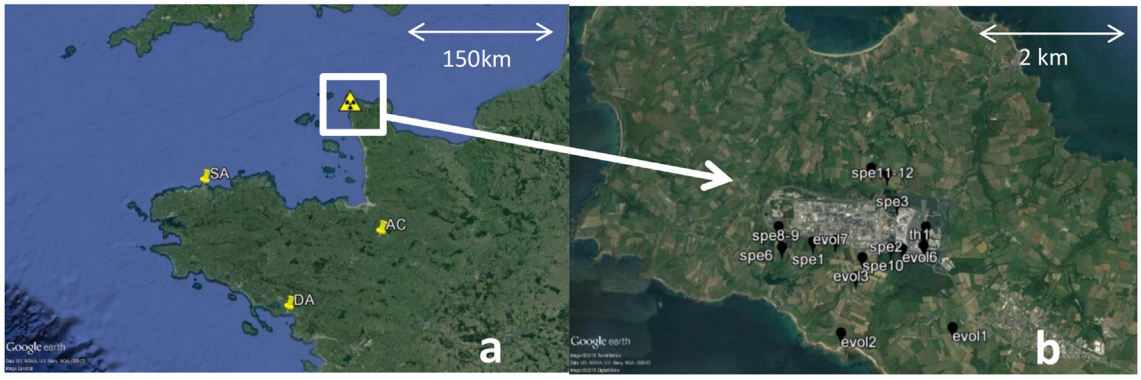

Fig. 1. Experimental area: (a) sampling site (AC, DA, SA) for background measurements in yellow mark, (b) sampling site near AREVA NC reprocessing plant (black dot). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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