Journal of Environmental Radioactivity xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Tritium and radiocarbon levels in the Rhône river delta and along the French Mediterranean coastline

P. Jean-Baptiste[a,*], M. Fontugne[a], E. Fourré[a], L. Marang[b], C. Antonelli[c], S. Charmasson[d], F. Siclet[b]

[a] CEA-CNRS-UVSQ, Laboratoire des Sciences du Climat et de l'Environnement, Centre de Saclay, 91191, Gif-sur-Yvette, France
[b] EDF-R&D, Laboratoire National Hydraulique et Environnement, 6 Quai Wattier, 78401, Chatou, France
[c] IRSN, PSE-ENV, Laboratoire d'Études Radioécologiques en milieu Continental et Marin, SESURE/LERCM, 13115, Saint Paul Lez Durance, France
[d] IRSN, PSE-ENV/SRTE, Laboratoire de Recherche sur les Transferts des radionucléides au sein des écosystèmes Aquatiques (LRTA), Centre IFREMER, 83507, La Seyne-sur-mer, France

ARTICLE INFO

Keywords:
Tritium
Radiocarbon
River Rhône
Mediterranean coastline

ABSTRACT

The Rhône is characterised by a heavy concentration of nuclear-based industries including nuclear power stations and nuclear sites housing civilian and military facilities. Here, we report the results of a four-year survey (2010–2013) of tritium and radiocarbon levels in a variety of matrices within the Rhône delta and along the French Mediterranean coastline. The aim of the study is to create a spatial reference framework of environmental levels of these two radionuclides, which are the most prevalent in radioactive effluents from nuclear power stations. Although both tritium and radiocarbon levels in the samples analysed are very low and can only be detected using ultra-sensitive analytical techniques, they clearly show the influence of the tritium and radiocarbon discharges carried by the Rhône plume along the Mediterranean coast. The tritium content of suspended matter and sediments of the Rhône is a special case, which shows elevated tritium values not seen in other French rivers with similar nuclear facilities. The north-south spatial distribution of this tritium anomaly shows that these trace values are at their highest in the upper Rhône, close to the Swiss border and upstream of Creys Malville, the northernmost nuclear power station on the Rhône. This points to a legacy of past tritium releases by the watchmaking industry. A dedicated study would be needed to clearly identify the source and the exact nature of this contamination.

1. Introduction

The Rhône is the main river flowing into the western Mediterranean. Its catchment is characterised by a heavy concentration of nuclear-based industries, including four nuclear power stations with a total of 14 reactors operated by Electricité de France (EDF), and two major nuclear sites housing civilian and military facilities (Fig. 1). Unlike most radionuclides, for which there are effective industrial methods of capture in operation, there is no satisfactory technology for removing tritium and radiocarbon from effluents and their discharge is therefore not limited. As a result, these two radionuclides are the most prevalent in liquid radioactive effluent from nuclear power stations (UNSCEAR, 2017).

Although the environmental impact associated with these discharges is extremely low, it is nevertheless important to monitor the levels observed in the Rhône and in the marine environment. Such monitoring allows us to put the tritium and radiocarbon releases associated with the operation of the power stations in context and compare them to other contributions (either natural or due to human activity), and to consolidate the estimation of the doses associated with these discharges, whether for ecosystems or humans.

Here, we report the result of a four-year survey (2010–2013) of water, suspended matter, surface sediments and various macro-organisms, aimed at creating a multi-matrix spatial reference framework of levels of tritium and radiocarbon in the Rhône delta and along the French Mediterranean coastline.

2. Characteristics of the survey zone

2.1. The Rhône

With a length of 812 km and a catchment area of around 100,000 km^2, the Rhône is the main river flowing into the western Mediterranean basin. It rises in the Swiss Alps at an altitude of 2200 m and ends in the Camargue delta before flowing into the Gulf of Lions.

The Rhône's hydrological profile is characterised by significant

* Corresponding author.
E-mail address: pjb@lsce.ipsl.fr (P. Jean-Baptiste).

https://doi.org/10.1016/j.jenvrad.2018.01.031
Received 27 October 2017; Received in revised form 25 January 2018; Accepted 31 January 2018

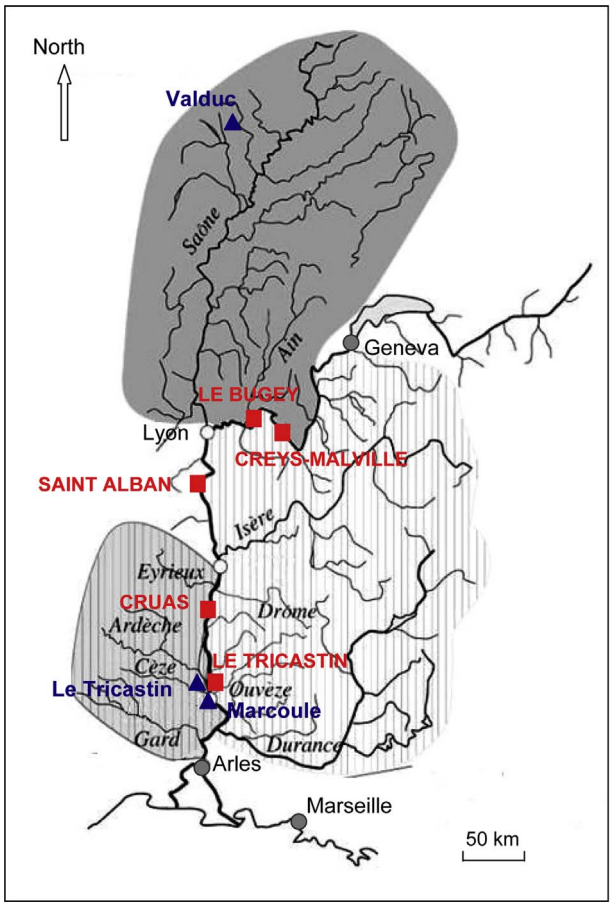

Fig. 1. Location of main nuclear activities in the Rhône catchment: nuclear power stations (red squares) operated by Electricité de France (EDF) and nuclear facilities (blue triangles) operated by the French Atomic Energy Commission (CEA) and the AREVA company. Note that Creys-Malville was decommissioned in 1997 and is currently being dismantled.

seasonal and interannual variations. Flow rates are irregular with a low-flow period in summer and periods of high flow at the start of autumn and the end of winter. At Beaucaire, located 20 km to the north of Arles at the entrance to the Rhône delta, the mean annual flow rate of the river is 1700 $m^3 s^{-1}$. Nevertheless, with exceptional meteorological events (drought or floods originating in the Cevennes or Alps mountain ranges), the Rhône's flow rates can vary widely with periods of low flow at less than 500 $m^3 s^{-1}$ and periods of exceptionally high flow reaching more than 10,000 $m^3 s^{-1}$ (Pont et al., 2002). The annual floods are generally characterised by a threshold flow rate of around 3000 $m^3 s^{-1}$. The most recent exceptional, one-in-a-hundred year, flood occurred on 8 January 1994, with a flow rate of 11,000 $m^3 s^{-1}$ at Beaucaire (Météo-France -http://pluiesextremes.meteo.fr).

The runoff from the Rhône's catchment transports more than 80% of the sediments entering the Gulf of Lions (Monaco et al., 1990). This transport plays a major role in determining the dynamics of the ecosystems of the Gulf of Lions (Cruzado and Velasquez, 1990). The much smaller catchment areas in the Pyrenees and Languedoc, provide the rest of the runoff. The variability observed with river flow rates is also reflected in the seasonal dynamics of the solid transport, the Alpine tributaries (Arve, Fier, Isère, Durance) and the Saône being the main providers: overall on the order of 2–8 Mt yr^{-1}, although they can reach up to 26.5 Mt yr^{-1} (Pont et al., 2002). Around 80% of solid material is mobilised during flooding, contributing to high fluxes of suspended sediments and radionuclides to the Mediterranean Sea (Antonelli et al., 2008).

Fig. 2. Satellite image of the Rhône plume (NASA/Goddard Space Flight Center).

2.2. The Rhône delta

The Camargue, formed by the Rhône delta, is a huge sedimentary complex bordered by the Petit Rhône to the west and the main Rhône to the east, which provides 90% of the total river flow. At the mouth of the Rhône the fresh water spreads out across the denser seawater forming a dilution plume that can be easily identified by its turbidity (Fig. 2). The material suspended in the plume settles quickly under the effect of flocculation and aggregation of organic minerals (Thill et al., 2001). This accumulation of particles leads to the creation of a sedimentary prism (Got et al., 1985). This prodelta plays a central role in the sedimentary dynamics of the Gulf of Lions because it receives solid runoff directly from the Rhône and it ensures continuity with the continental shelf. This sedimentary accumulation leads to the capture of a substantial proportion of the pollutants carried by the Rhône (Charmasson et al., 1998; Roussiez et al., 2005; Lansard et al., 2007; Mourier et al., 2014; Cossa et al., 2018).

2.3. Coastal lagoons

The Mediterranean lagoons (‘étangs’ in French) are water bodies separated from the sea by a strip of coastline and supplied with fresh water by the runoff from the catchment area. Created in the main from a body of seawater being isolated by the build up of sand and pebbles carried by major and minor rivers more than 2000 years ago, Mediterranean lagoons are generally shallow (about 1 m deep). Apart from these sedimentary lagoons, others may have tectonic origins and these are characterised by greater depths (e.g., 4.5 m for the Etang de Thau – Fig. 6). Located between the land and sea, these lagoons receive various types of runoff from the catchment area and are connected to the sea by means of a 'grau' (Occitan word meaning 'channel').

The ‘Palavasian’ lagoons extend for about 20 km around the cities of Sète and Montpellier. This lagoon complex comprises nine shallow (average 0.6 m) water bodies (la Peyrade, les Mouettes, Ingril, Vic, Pierre-Blanche, Arnel, Prévost, Méjean-Pérols and Grec). Further south, the lagoons of Leucate, Sigean and Canet are also shallow, with an average depth of 2 m, 2.5 m and 0.5 m, respectively. To the east, the lagoons of the Petite Camargue complex (Etang du Cabri and Etang des Launes) are not only influenced by both the sea and the land, but also by the floods from the Petit Rhône.

2.4. General circulation in the Gulf of Lions and dynamics of the Rhône plume

The Gulf of Lions is situated in the northwestern part of the western Mediterranean basin. With a surface area of around 6700 km^2, the Gulf of Lions is characterised by one of the most significant continental shelves in the Mediterranean. Water masses circulating in the Gulf are influenced by a general cyclonic-type circulation characterised by the existence of the Northern Current (also known as the 'Ligure Current'). This current is between 30 and 50 km wide and runs along the Gulf of