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Tracing the origin of suspended sediment in a large Mediterranean river by combining continuous river monitoring and measurement of artificial and natural radionuclides

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HIGHLIGHTS

- The majority of sediment flux from the Rhone River is exported during floods.
- Classification of floods recorded between 2000 and 2012 at the outlet was provided.
- Natural and artificial radionuclides were used as fingerprints of sediment origin.
- Discrimination was achieved between Pre-alpine, Cevenol and Upstream inputs.
- Results showed that Pre-Alpine tributaries were the main sediment supplier.

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ABSTRACT

Delivery of suspended sediment from large rivers to marine environments has important environmental impacts on coastal zones. In France, the Rhone River (catchment area of 98,000 km^2) is by far the main supplier of sediment to the Mediterranean Sea and its annual solid discharge is largely controlled by flood events. This study investigates the relevance of alternative and original fingerprinting techniques based on the relative abundances of a series of radionuclides measured routinely at the Rhone River outlet to quantify the relative contribution of sediment supplied by the main tributaries during floods. Floods were classified according to the relative contribution of the main subcatchments (i.e., Oceanic, Cevenol, extensive Mediterranean and generalised). Between 2000 and 2012, 221 samples of suspended sediment were collected at the outlet and were shown to be representative of all flood types that occurred during the last decade. Three geogenic radionuclides (i.e., ^{238}U, ^{232}Th and ^{40}K) were used as fingerprints in a multivariate mixing model in order to estimate the relative contribution of the main subcatchment sources—characterised by different lithologies—in sediment samples collected at the outlet. Results showed that total sediment supply originating from Pre-Alpine, Upstream, and Cevenol sources amounted to 10, 7 and 2.10^6 tons, respectively. These results highlight the role of Pre-Alpine tributaries as the main sediment supplier (53%) to the Rhone River during floods. Other fingerprinting approaches based on artificial radionuclide activity ratios (i.e., $^{137}Cs/^{239+240}Pu$ and $^{238}Pu/^{239+240}Pu$) were tested and provided a way to quantify sediment remobilisation or the relative contributions of the southern tributaries. In the future, fingerprinting methods based on natural radionuclides should be further applied to catchments with heterogeneous lithologies. Methods based on artificial radionuclides should be further applied to catchments characterised by heterogeneous post-Chernobyl ^{137}Cs deposition or by specific releases of radioactive effluents.

1. Introduction

Delivery of suspended sediments from large rivers to marine environments has important environmental impacts on coastal zones,

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where it modifies water quality, estuarine geomorphology and biogeochemical cycles (Syvitski et al., 2005; Meybeck and Vörösmarty, 2005; Meybeck et al., 2007; Durrieu de Madron et al., 2011). Furthermore, the quality of riverine suspended sediment is impacted by human activities in the river catchment, and sediments may transport various particle-reactive contaminants from their sources within the catchment and convey them into marine environments (Durrieu de Madron et al., 2011; Fohrer and Chicharo, 2011).

The magnitude of this impact is controlled by the sediment discharge of rivers that is strongly variable throughout time (Meybeck et al., 2003). In France, the Rhone River is by far the main supplier of sediment to the marine environment (Delmas et al., 2012).

The Rhone River supplies almost two-thirds of the total river discharge into the western Mediterranean Sea (Ludwig et al., 2009), delivering together with the Po River the most important input of suspended sediment to the Mediterranean Sea (Syvitski and Kettner, 2007). It delivers more than 80% of the particulate inputs to the Gulf of Lions (Raimbault and Durrieu de Madron, 2003) and exerts thereby a major ecological influence by enhancing primary productivity (Bosc et al., 2004).

In the Rhone River, export of annual suspended solid loads is concentrated during floods (Sempéré et al., 2000; Ollivier et al., 2011) with significant inputs from southern tributaries (Pont et al., 2002). For instance, the large flood that occurred in December 2003 exported 83% of the total annual sediment load (Antonelli et al., 2008; Ollivier et al., 2010). As it covers a large drainage area (98,000 km^2) characterised by strong variations in climate and geological conditions, the relative contribution of the main Rhone River tributaries to its sediment discharge may vary throughout time (Pardé, 1925; Pont et al., 2002; Antonelli et al., 2008). Sediment conveyed by the Rhone River was documented to contain large concentrations in contaminants, such as organic pollutants (Sicre et al., 2008; Desmet et al., 2012; Mourier et al., 2014) and metals (Radakovitch et al., 2008). In addition, the Rhone valley represents Europe's largest concentration of nuclear power plants, and the river receives radioactive liquid effluents originating from four nuclear plants and a spent fuel reprocessing plant currently under dismantlement. As a consequence, sedimentary archives were shown to reflect significant enrichments in artificial radionuclides in the lower Rhone River sections (Ferrand et al., 2012). It is therefore crucial to better constrain those sources as marine sediments have the capacity to store these contaminants in continental shelf areas and in abyssal plains (Charmasson et al., 1998; Radakovitch et al., 1999; Lee et al., 2003; Garcia-Orellana et al., 2009).

In this context, quantifying the sources supplying sediments to the Rhone River and eventually to the Mediterranean Sea represents a crucial prerequisite for better understanding the riverine transfer and its potential role in global biogeochemical cycles, and for implementing effective control strategies to improve water and sediment quality (Walling and Collins, 2008). The relative contribution of sediments originating from the main tributaries during the floods recorded in the lower sections of the Rhone River was shown to reflect lithological differences and to imprint the geochemical and mineralogical properties of the main upstream sediment sources (Pont et al., 2002; Ollivier et al., 2010; Zebracki et al., 2013a). We therefore propose to use these sediment characteristics to fingerprint the origin of sediment transported in the lower Rhone River.

Exports of suspended sediments from the Rhone River have been continuously monitored since 2000 at the Rhone River Observatory Station in Arles (SORA), which is located 40 km upstream of its mouth. Furthermore, the suspended sediment content in natural and artificial radionuclides has been continuously analysed in the framework of legal radioecological surveillance (Eyrolle et al., 2012).

This study therefore proposes to provide alternative and original fingerprinting techniques based on radionuclide properties measured routinely to quantify the proportion of sediments supplied by the different tributaries to the Rhone River outlet during floods. Variations in geogenic ^{238}U, ^{232}Th and ^{40}K radionuclide activities may reflect the contribution of source areas with different lithologies (Olley et al., 1993; Yeager and Santschi, 2003). In addition, amongst artificial radionuclides, spatial variations in ^{137}Cs and plutonium isotope activities (i.e., ^{238}Pu and $^{239+240}$Pu) may provide powerful tracers of the sediment origin. During Rhone River floods, ^{137}Cs was shown to originate mainly from erosion of the soils contaminated by global atmospheric fallout and Chernobyl accident (Antonelli et al., 2008), and to display an East-West decreasing gradient of contamination across the catchment (Renaud et al., 2003; Roussel-Debel et al., 2007). In contrast, and in the particular case of the lower Rhone River, Pu isotopes may either originate from erosion of the catchment soils contaminated by global atmospheric fallout or from remobilisation of sediment labelled by liquid effluents released by the Marcoule spent fuel reprocessing plant (from 1960s and decommissioned since 1997).

A method based on ^{238}Pu/$^{239+240}$Pu activity ratio (PuAR) measurements was developed to estimate the fraction of the Pu isotopes that originated from the Rhone River in marine deposits (Thomas, 1997; Lansard et al., 2007). This method was applied to distinguish between Pu supply through soil erosion across the Rhone catchment and remobilisation of sediment stored in the river channel downstream of Marcoule, and to quantify their relative contribution to Pu fluxes recorded at Arles (Rolland, 2006; Eyrolle et al., 2012).

After identifying the origin of floods, fingerprinting techniques presented above were applied to quantify the relative contribution of the main sources delivering sediments to the Rhone River, based on the continuous measurement of natural and artificial radionuclides. They were applied to suspended sediments collected at Arles outlet (SORA observatory station) between October 2000 and June 2012.

2. Study area

The Rhone River basin area consists of four mountainous subcatchments, i.e., Alps, Jura, Cevennes/Massif Central, and Vosges. In the northern part of the basin, the Jura and Vosges mountains (drained by the Saone River) are mainly calcareous. When moving to the South and to the East, the Alpine mountains (drained by the Upper Rhone, Isere and Durance rivers) mostly consist of sedimentary rocks and of siliceous crystalline and metamorphic rocks. Finally, in the southwestern part of the basin, crystalline siliceous rocks dominate in the Cevennes Mountains (drained by the Ardeche, Ceze and Gard rivers). Details on those geological substrate variations are given in Ollivier et al. (2010) and Ollivier et al. (2011). In addition to this geological heterogeneity, the Rhone basin is exposed to a wide variety of climate conditions (Pont et al., 2002). The tributaries of the Rhone are used to be organised in three main groups characterised by distinct hydrographic features (Fig. 1): northern tributaries (Ain, Fier, Isere, Saone rivers) and southern tributaries, which may in turn be distinguished as Cevenol tributaries (Eyrieux, Ardeche, Ceze, Gard rivers) and southern Pre-Alpine tributaries (Durance, Drome, Aigues, and Ouveze rivers).

Forty kilometres upstream of its mouth the Rhone River subdivides into the Grand Rhone River and the Petit Rhone River, and flows into a delta of 1500 km^2 (Fig. 1). The sampling station in Arles is located on the Grand Rhone River, which drains about 90% of the water discharge. The SORA observatory in Arles is an automatic sampling station operated by the French Institute for Radioprotection and Nuclear Safety (IRSN/LERCM).

At the Beaucaire gauging station (i.e., 8 km upstream of the Rhone difluence, and 14 km upstream of the SORA sampling station in Arles), the Rhone mean annual discharge was about 1700 m^3 s^{-1} for the period 1920–2012, whilst the annual suspended solid load varied from 1.2 to 19.7 Mt yr^{-1} for the period 1961–1996 (Antonelli, 2002; Pont et al., 2002). River discharges associated with the 1-yr, 2-yr, 10-yr, and 100-yr return periods amount to 4000, 5000, 8400 and 11,200 m^3 s^{-1}, respectively.