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U isotopes distribution in the Lower Rhone River and its implication on radionuclides disequilibrium within the decay series

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ABSTRACT

The large rivers are main pathways for the delivery of suspended sediments into coastal environments, affecting the biogeochemical fluxes and the ecosystem functioning. The radionuclides from ^{238}U and ^{232}Th—series can be used to understand the dynamic processes affecting both catchment soil erosion and sediment delivery to oceans. Based on annual water discharge the Rhone River represents the largest river of the Mediterranean Sea. The Rhone valley also represents the largest concentration in nuclear power plants in Europe. A radioactive disequilibrium between particulate $^{226}Ra_{(p)}$ and $^{238}U_{(p)}$ was observed in the suspended sediment discharged by the Lower Rhone River (Eyrolle et al. 2012), and a fraction of particulate ^{234}Th was shown to derive from dissolved $^{238}U_{(d)}$ (Zebracki et al. 2013). This extensive study has investigated the dissolved U isotopes distribution in the Lower Rhone River and its implication on particulate radionuclides disequilibrium within the decay series. The suspended sediment and filtered river waters were collected at low and high water discharges. During the 4—months of the study, two flood events generated by the Rhone southern tributaries were monitored. In river waters, the total $U_{(d)}$ concentration and U isotopes distribution were obtained through Q-ICP-MS measurements. The Lower Rhone River has displayed non-conservative U—behavior, and the variations in $U_{(d)}$ concentration between southern tributaries were related to the differences in bedrock lithology. The artificially occurring ^{236}U was detected in the Rhone River at low water discharges, and was attributed to the liquid releases from nuclear industries located along the river. The $(^{235}U/^{238}U)_{(d)}$ activity ratio (=AR) in river waters was representative of the ^{235}U natural abundance on Earth. The $(^{226}Ra/^{238}U)_{(p)}$ AR in suspended sediment has indicated a radioactive disequilibrium (average 1.3 ± 0.1). The excess of ^{234}Th in suspended sediment $=(^{234}Th_{xs(p)})$ was apparent solely at low water discharges. The activity of $^{234}Th_{xs(p)}$ was calculated through gamma measurements and ranged from unquantifiable to 56 ± 14 Bq kg^{-1}. The possibility of using ^{234}Th as a tracer for the suspended sediment dynamics in large Mediterranean river was then discussed.

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1. Introduction

The large rivers are a major pathway for the delivery of suspended sediment into marine environments, affecting the biogeochemical fluxes and the ecosystem functioning at coastal zones (Syvitski et al., 2005; Meybeck, 2003; Walling and Fang, 2003). The quality of riverine suspended sediment is impacted by human activities in the river catchment and many particle-reactive contaminants are discharged at the river basin outlet (MerMex Group et al., 2011; Fohrer and Chicharo, 2011). As a growing issue, the management of riverine suspended particles requires comprehensive assessment of the suspended sediment dynamics at the scale of river catchment (Meybeck, 2003; Cardenas and Lick, 1996).

The radionuclides from ^{238}U and ^{232}Th—series can be used to understand dynamic processes, since radionuclides are continuously supplied and removed by physical or chemical processes as well as by radioactive decay (Vigier and Bourdon, 2011; Siddeeg, 2016; Porcelli et al., 2001).

Based on annual discharge the Rhone River represents the largest river of the Mediterranean Sea (about 1700 $m^3\ s^{-1}$), discharging together with the Po River almost one third of total freshwater input (Ludwig et al., 2009). The Rhone is the main sediment supplier, providing more than 80% of the total riverine input to the Gulf of Lions (Sempéré et al., 2000; Durrieu et al., 2000). The sediment and organisms at the Rhone outlet were shown to contain trace contaminants, as metals, PCB, pesticides and PAHs (MerMex Group et al., 2011). Besides, the Rhone valley represents the largest concentration in nuclear power plants in Europe. The Rhone River receives liquid effluents originating from four nuclear plants and one dismantling spent fuel reprocessing plant. Gamma-emitting liquid effluents from nuclear sites account for nearly 65% of the total discharged in French aquatic continental environment (Eyrolle-Boyer et al., 2015).

The suspended sediment of the Lower Rhone River was shown to display a radioactive disequilibrium between particulate ^{226}Ra and ^{238}U in ^{238}U—series (Eyrolle et al., 2012; Zebracki et al., 2013), and to contain a fraction of particulate ^{234}Th derived from the radioactive decay of dissolved ^{238}U (Zebracki et al., 2013). This extensive study has investigated the dissolved (=d) U isotopes distribution in the Lower Rhone River and its implication on particulate (=p) radionuclides disequilibrium within the decay series. Due to the influence of the southern tributaries on the chemical composition of waters discharged at the Rhone outlet (Ollivier et al., 2010), the investigations in U—origin and U—behavior were extended to the Lower Rhone basin, including two subcatchments displaying contrasted bedrock lithology.

In the Lower Rhone basin, samplings of river sediment and waters were conducted during contrasted periods of high and low water discharges. Naturally-occurring radionuclides were obtained through gamma measurements, and analyses in $U_{(d)}$ concentration and U isotopic composition were performed by Q-ICP-MS.

2. Material and methods

2.1. Study area

The Rhone River basin (about 98,000 km^2) displays heterogeneous geological features and is exposed to a wide variety of climate regimes (Pont et al., 2002; Pardé, 1925). The Rhone River basin consists of four mountainous sub-catchments displaying different bedrock lithology, *i.e.*, Alps, Jura, Vosges and Cevennes/Massif Central (Fig. 1). In the northern part of the basin, the Vosges and the Jura mountains are mainly calcareous. In the southeastern part, the Alps Mountains mostly consist of sedimentary, siliceous crystalline and metamorphic rocks. In the southwestern part, siliceous crystalline rocks dominate in the Cevennes/Massif Central Mountains.

Floods occurring in the Lower Rhone River are classified in four types, *i.e.*, Oceanic, Cevenol, Extensive Mediterranean, and Generalized, depending on the climatological regimes affecting the different sub-catchments (Pardé, 1925; Zebracki et al., 2015; Pont et al., 2002).

Forty kilometers from the mouth of the Rhone River, the river divides into the Petit Rhone River and the Grand Rhone River, which drains 90% of the water discharge (Ibanez et al., 1997), then flows to a delta of about 1500 km^2 into the northwestern Mediterranean Sea (Gulf of Lions). At the Beaucaire gauging station (*i.e.*, just upstream of the diffluence), the mean Rhone annual discharge was about 1700 $m^3\ s^{-1}$ for the period 1920–2012, and the annual suspended solid flux varied between 1.2 and 19.7 Mt y^{-1} for the period 1960–1996 (Antonelli, 2002; Pont et al., 2002). River discharges associated to 1—yr, 2—yr, 10—yr and 100—yr return