



Key parameters influencing metallic element mobility associated with sediments in a daily-managed reservoir



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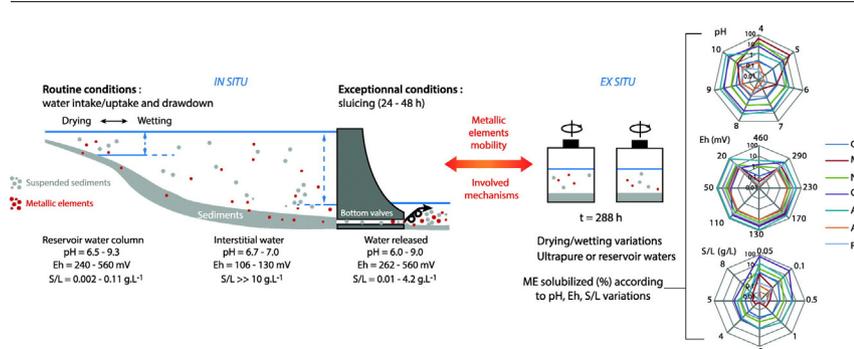
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HIGHLIGHTS

- Dam sediments are subjected to natural or human-induced physicochemical changes.
- Changes in water pH can lead to enhance dissolved metal concentration by a factor 5.
- Decrease in S/L ratio during resuspension event promotes metals solubilization.
- Eh induces metals mobility through bearing phases dissolution/precipitation.
- Sediment drying pretreatment affects metallic elements speciation and mobility.

GRAPHICAL ABSTRACT



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ABSTRACT

In a hydroelectric reservoir, sediments are subject to remobilization events, water-level fluctuations and physicochemical changes. Depending on their associated metallic content, surficial oxic to suboxic sediments could constitute a major source of metals. To identify the key parameters that control metallic elements in terms of their mobility and sensitivity to reservoir management, sediments were subject to resuspension and drying/wetting cycle experiments over a wide range of pH values, solid/liquid ratios (S/L) and redox (Eh) conditions. During these tests, special attention was also paid to the influence of pretreatments on samples, i.e., drying, aeration and the leachate composition (ultrapure water vs. natural water); on the preservation of the sediment characteristics; and especially on metallic element release. The results of this study show that the pH, S/L ratio and Eh parameters are key variables in metal solubilization; the pH influences metal mobility primarily through sorption-desorption phenomena as well as the dissolution of metallic-bearing phases, the S/L ratio modifies the sorption-desorption equilibria, and the Eh primarily affects the reducible sensitive phases and associated metallic elements through dissolution-precipitation processes. Under environmental conditions, evolution of these parameters can lead to a >20% solubilization of the most mobile elements, i.e., As and Cd. These results are influenced by the sample pretreatment and experimental conditions. In fact, even if the solubilization patterns show no significant differences between dry and wet sediment depending on the physicochemical conditions, the magnitude of their release is significantly affected. Drying pretreatment induces changes in metal speciation, notably altering the distribution of the most weakly bound elements; there is almost half the amount of metallic elements

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associated with the exchangeable fraction in dry compared to wet sediments. The solubilization percentages were higher in the ultrapure phase than in reservoir water primarily due to the low pH, which influenced the sorption equilibria.

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1. Introduction

Sediments act as a major sink for metallic elements (Viers et al., 2009; Zhang et al., 2014). This is the case for rivers equipped with dams in which low water flow favours particle sedimentation and metal scavenging (Zhao et al., 2013). Nevertheless, sediment retention is not definitive, and the associated metal concentrations and their partitioning are largely influenced by (i) the sediment characteristics; (ii) the intrinsic properties of the metallic elements; and (iii) external factors such as the physicochemical and hydrological conditions as well as the water column composition (e.g., Eggleton and Thomas, 2004; Alves Martins et al., 2015). As a result, the fate of trace metals is complex, and similar contents within sediments can be more or less mobile and bioavailable to biota.

The analysis of binding forms is often performed through operationally defined partitioning protocols, such as solubilization tests and selective extractions (e.g., Tessier et al., 1979). The combination of these protocols, along with physical approaches such as X-ray Diffraction (XRD) and Scanning Electron Microscopy (SEM-EDX), is necessary because they are subjected to inherent limitations that are due to the lack of selectivity of chemical extractants, as extensively reported in the scientific literature (e.g., Bacon and Davidson, 2008). They are also limited by the heterogeneous composition of the sediments, which dictates their associated contaminant sensitivities under experimental conditions (Peltier et al., 2005). Thus, specific attention should be paid to the physicochemical conditions during handling and the experimental treatment of sediments, notably regarding aeration and drying phenomena, which can occur during the pretreatments prior to extraction. Some authors demonstrated that they could lead to modifications in the solid speciation and bearing phase alterations, therefore promoting an evolution in the metallic solubilization process (Baeyens et al., 2003; Bordas and Bourg, 1998). These pretreatments may notably influence the evolution of the pH and Eh parameters, which are two of the primary factors controlling metal mobility in aquatic systems (Tack et al., 1996). If the effects of these changes on metal mobility have been studied in highly contaminated and/or anoxic sediments (Bordas and Bourg, 1998; Kersten and Förstner, 1987; Kersten and Förstner, 1986; Larner et al., 2008), then they have received little attention with respect to daily-managed reservoirs characterized by naturally contaminated oxic to suboxic sediments. In fact, in a dam context, similar changes in the physicochemical conditions could be generated during intensive managing operations, leading to the induction of modifications in the circulation and level of the water column, a high and rapid evolution in the oxygenation status and therefore an evolution in the metallic element mobility. Top sediments can also be subject to resuspension events, fast successions of inundation and desiccation as well as physical changes, especially during water release/intake. These changes can include the compaction and aggregation of surficial fine particles, as well as changes in the pH, Eh and rapid changes in the solid/liquid ratio (S/L).

For this study, a methodological approach was thus selected to gain new insights into the potential effect of dam exploitation on the mobility of metals and, consequently, on the water quality. For that purpose, the importance of changes in the primary variables within the reservoir (the pH, redox conditions, S/L ratio and contact time) that dictate the mobility of metals was examined through independent laboratory sequential extractions and solubilization tests. The results of these tests were compared to the in situ environmental variation range as

measured for 19 months during routine and exceptional exploitations. The influence of the pretreatment modalities (drying) and water composition (synthetic versus water reservoir) were also tested and discussed.

2. Material and methods

2.1. Hydroelectric complex description

Depending on the season and energy demand, the Vaussaire reservoir (Rhue watershed, Cantal, France) is subjected to daily water intake/uptake, leading to water level fluctuations that can reach up to 5 m (April 2014, personal observation). Moreover, to preserve its water storage capacity, this reservoir has undergone specific annual (or biennial) sluicing operations since 2003, i.e., the opening of the dam's bottom valves during high flows to release the accumulated sediments in the downstream river. The reservoir (1.5 hm³) is thus entirely emptied and refilled on a 24-hour scale. This management limits the formation of massive sediment stocks so that the sediments in the reservoir are fresh, and they have been deposited over the year or two years preceding the previous sluicing event (Frémion et al., 2016b). Because of these intensive daily, seasonal and exceptional variations, the Rhue water column exhibited a wide range of pH values [6.0–9.3], S/L [0.002–4.2 g·L⁻¹], O₂ saturation [80–180%] and Eh [240–560 mV] values. These variations lead to partial to complete aeration and drying, which may also help to change the metal mobility and speciation.

2.2. Sediments and interstitial water sampling and pretreatments

Top sediment samples (from the top layer at the 0 to 5 cm depth) were collected in December 2013 and April 2014 using an Ekman grab sampler (Eijkkelkamp Soil & Water, Giesbeek, The Netherlands). All the samples were introduced into sealed plastic bags and kept in a cooling box during transportation. In the laboratory, dry (D) sediments were prepared by oven-drying them at 40 °C, and they were sieved to 2 mm and 63 µm sizes (Nylon meshes). Wet (W) sediments were also quickly sieved at 2 mm and 63 µm (the most reactive and easily remobilized fraction) in the laboratory the day after sampling and then immediately stored at 4 °C in different sealed containers, and the containers were filled as much as possible to squeeze out air before further analysis.

The reservoir water was sampled, filtered (0.2 µm, Nylon mesh) and conserved at 4 °C for further solubilization tests. Interstitial water (IW) was sampled in situ immediately after sediment collection using a Rhizon device (Rhizosphere, Rhizon MOM pore diameter 0.1 µm). The water physicochemical parameter measurements (pH, Eh, conductivity, and dissolved oxygen) were performed in situ immediately after sample collection using a previously calibrated portable WTW multi-parameter.

2.3. Sediment characterization and mineralization

The water contents of all the samples were determined by oven-drying them to a constant weight (~1 g) at 105 °C. All the subsequent analyses were calculated according to these dry weight measurements. In the case of experiments performed with wet sediments, a dry weight correction using the water content was performed to maintain the same S/L ratio (dry mass). The loss on ignition (LOI) values were used to

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