



Assessment of baseline ecotoxicity of sediments from a prospective mining area enriched in light rare earth elements



A. Romero-Freire^{a,*}, L. Minguez^{a,1}, M. Pelletier^b, A. Cayer^b, C. Caillet^c, S. Devin^a, E.M. Gross^a, F. Guérol^a, S. Pain-Devin^a, D.A.L. Vignati^{a,2}, L. Giamberini^{a,2}

^a LIEC, CNRS, UMR 7360, Université de Lorraine, Campus Bridoux, Bâtiment IBISE, 8 rue du Général Delestraint, 57070 Metz, France

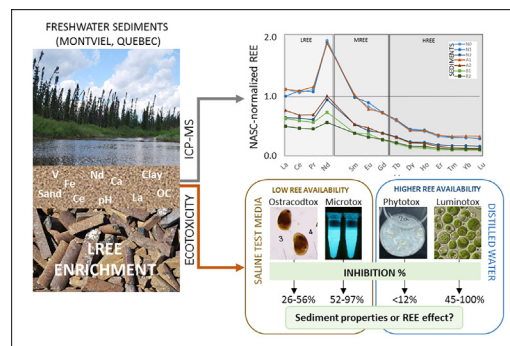
^b Ressources Géoméga, 75 Boulevard de Mortagne, Boucherville J4B 6Y4, Québec, Canada

^c LIEC, CNRS, UMR 7360, Université de Lorraine, 15 Avenue du Charmois, 54500 Vandœuvre-lès-Nancy, France

HIGHLIGHTS

- Sediments were collected in an area naturally enriched in LREEs.
- Microscale tests were used to assess the ecotoxicity of sediment.
- Ecotoxicological protocols varied in their ability to mobilize REEs from sediments.
- Different REE availability and toxicity depended on sediment composition.
- Sediment characteristics likely contributed to observed ecotoxicity.

GRAPHICAL ABSTRACT



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ABSTRACT

Rare earth elements (REEs) disperse from indigenous rocks to the environment, thus making sediments one of the major sinks and sources of metal pollution. The emerging use of REEs and the subsequent opening of new mining areas may contribute to their release into surrounding ecosystems. For this reason, this study was performed in a natural area with geological material abundant in ferrocarnatites and light REE. The aim of this work was to assess the natural REE availability and (eco)toxicity in freshwater sediments. Sediments showed high REE concentrations in samples with fine grain size fractions, and low in organic-rich sediments. The enrichment in LREE was mostly from rocks and the obtained enrichment factors (EF) confirmed that the sediments are not anthropogenically polluted. To assess REE availability and ecotoxicity, four toxicity tests were performed. REEs measured as the dissolved concentration in the test media were very low compared to the potentially available total REE in sediments and showed positive or negative correlations with fine or coarse grain sizes, respectively, and positive correlations with the content in Mg, Fe and Al. In tests performed in media supplemented with salts, the availability of REEs decreased considerably. Only some toxic effects could be linked to the REE contents in the ostracodtox and luminotox tests. However, measurement of toxicity could be influenced by the sediment properties and lead to a potential overestimation of ecotoxicity if only REE are regarded. Our study reveals that the physicochemical properties of sediments are a key factor controlling both REE availability and toxicity, whereas the determination of REE effects from toxicity tests using liquid media with salt addition will decrease

* Corresponding author.

E-mail address: ana.romero-freire@univ-lorraine.fr (A. Romero-Freire).

¹ These authors have contributed equally as first authors.

² These authors have contributed equally as last authors.

REE availability and could mask toxic effects. Our findings provide new knowledge about REE behaviour in sediments and are a starting point for understanding potential REE pollution around prospective mining areas.

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1. Introduction

Rare earth elements (REEs) have become a highly valuable commodity due to their increased use in emerging technology applications (Lambert and Ledrich, 2014). They encompass the 15 lanthanides (LNs) plus scandium and yttrium. The future exploitation of areas with high REE contents will play a strategic geopolitical role around the world (Gholz, 2014). Five REEs have been identified as economically critical for energy production (Dy, Tb, Eu, Nd and Y) and two additional ones as nearly critical (Ce and La) (US DOE, 2012), with three of them belonging to the group of light rare earth elements (LREE) (La, Ce and Nd). The progressing activity in opening new mining areas will contribute to increased REE mobility and will influence the fate of these elements in the environment over the next decades (Davranche et al., 2015). Establishing baseline (eco)toxicity in prospective mining areas will be crucial to monitoring of the contribution of mining activity to a potential REE enrichment in the surrounding environments during and after mining activities, and their subsequent rehabilitation at the end of the life cycle of exploitation.

In recent years, the environmental behaviour of REEs has continued to attract attention, especially the study of their geochemistry (Aksentov and Sattarova, 2016; Armand et al., 2015; Silva et al., 2014; Zhang et al., 2014), along with an increased interest in their ecotoxicity (Herrmann et al., 2016; González et al., 2014). Insoluble forms of LNs have shown low toxicity, however high toxicity was reported for soluble ones (Lambert and Ledrich, 2014), thus calling for further studies of their potential to cause environmental damage. Even if all REEs usually show similar behaviour during natural processes owing to their similar atomic and chemical properties (Aksentov and Sattarova, 2016), there is no consensus about a predictable pattern of toxicity of REEs across the lanthanide series (González et al., 2014). This warrants further investigation on REE (eco)toxicity. However, until today, only few publications on soil and waters deal with their toxicity (González et al., 2014), and studies in aquatic sediments are scarce, although they are recognised as a major sink (and potential secondary source) for many contaminants characterized by low solubility and/or strong affinity to environmental particles, such as REEs (Watts et al., 2017). Herrmann et al. (2016) identified five studies dealing with toxicity in sediments, performed with lanthanum and/or Phoslock® (a lanthanum modified clay), four in artificial sediments and one in natural sediment. Other studies dealing with REE toxicity in natural sediments evaluated the contamination by using geochemical indices calculation based on the content in other reference materials and/or metal backgrounds, but without parallel ecotoxicological assessments (e.g. Atibu et al., 2016; Watts et al., 2017). In addition, available ecotoxicity data have been obtained by exposure to only one element, with most studies focussing on Ce, La and Gd (Pagano et al., 2015); whereas other REEs remains relatively scarce (Pagano et al., 2016), and studies on REE mixture toxicity are almost absent.

For this study, a LREE enriched geological site has been selected (Montviel, Quebec). The most special feature of the selected area is that the main deposit presents an intrusion with abundant ferrocarnatites, differentiating this area from the others around the World (Nadeau et al., 2015). Moreover, the area is located in a pristine area, still unexploited. The owner company of the area (GéoMégA Resources) has estimated the inferred mineral resource as 266.6 Mt., with a total rare earth oxide grade of 1.43%. Many LNs are known to possess highly adsorptive properties, causing them to bind to mineral particles, colloids and organic matter, thus influencing their availability (Tan et al., 2014). The ecotoxicity assessment of natural sediments, as they are occurring in rivers or small

streams, integrates physicochemical effects of sediment properties that might change REE availability and therefore potential REE toxicity. The most common way to assess the ecotoxicity of a pollutant is by obtaining direct responses of living organisms exposed to these compounds, however, the effects of the pollutants on organisms will greatly differ due to different uptake pathways, which asks for the use of a set of organisms for a more realistic ecotoxicity assessment.

The general objective of the present study is to assess the baseline ecotoxicity of freshwater sediments in a prospective mining area with high LREEs content using aquatic organisms representative of the major groups (decomposers, primary producers and consumers) and taking into account the different physicochemical properties of sediments.

2. Materials and methods

2.1. Study area and sampling procedure

Seven sediment sampling sites were selected in Montviel (49° 49' 28.7"N, 76° 31' 46.4"W, Northern Québec, Canada) according to their parent materials and the existence of a watercourse. In total, three watercourses were sampled, the main river crossing the studied area (N, Nomans River), and two unnamed rivers hereinafter referred to as stream A and stream B. Three sites were directly sampled in the upper part of the carbonatite rock material: B1 (ferrocarbonatite), B2 (silicocarbonatite) and N2 (polygenic breccia). Four further sites, among them two sites above pyroxenite material (A2 and N1), one above tonalite material (A1), and one far from the carbonatite intrusion, was sampled in the limit of tonalite and basalt material, upstream of Nomans River (N0) (Fig. 1). Sediment material was collected at each sampling site by a strainer at 0–10 cm depth from the river bank. Samples were stored in polyethylene bags, previously rinsed with distilled water. Bags were filled to the brim to reduce oxygen exposure and overlying water was removed by gentle manual pressure. Samples were stored in a dark cool box for their transport.

2.2. Physicochemical measurements

Once in laboratory, pH was directly determined in sediments (as they contained sufficient water) by a microprocessor pH-meter pH 3000 (WTW). Loss-on-ignition (%LOI), corresponding to the combustion of organic matter, was measured by weight difference of 2 mm-sieved dried sediment and ignited to 525 °C during 4.5 h in a Thermolyne 6000 Furnace (Sybron). The equivalent calcium carbonate content (%) (inorganic carbon equivalent, ICe) was determined by gas volumetry with a Bernard calcimeter (Barahona and Iriarte, 1984). Particle size analyses were performed with a laser diffraction granulometer (SympaTEC Helos BF) in wet conditions on sediments previously sieved at 500 µm and dispersed in ultra-pure water by ultrasonication. Size classification was expressed in volume percentage: particles of size ≤4.5 µm are considered to be mostly clay, fine silt between 4.5 and 21.5 µm, coarse silt in range 21.5–215 µm and sand fraction from 215 to 500 µm. Basic cations were extracted using CH₃COONH₄ (1 N, pH 7.0) and CEC (cmol_c kg⁻¹) was determined by the use of CH₃COONa (1 N, pH 8.2) (USDA-SCS, 1972). Extracted elements (Ca, Na, Mg and K) were measured by atomic absorption spectrophotometry (AAS) (Perkin Elmer AAnalyst5100) and base saturation (%V) was calculated from the obtained results.

Acid digestion of the finely ground sediment fraction in a ratio of 1:20 (w:v sediment:extractant) was performed with a mixture of

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