



# Dependence of samarium-soil interaction on samarium concentration: Implications for environmental risk assessment<sup>☆</sup>

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## ABSTRACT

The sorption and desorption behaviour of samarium (Sm), an emerging contaminant, was examined in soil samples at varying Sm concentrations. The obtained sorption and desorption parameters revealed that soil possessed a high Sm retention capacity (sorption was higher than 99% and desorption lower than 2%) at low Sm concentrations, whereas at high Sm concentrations, the sorption-desorption behaviour varied among the soil samples tested. The fractionation of the Sm sorbed in soils, obtained by sequential extractions, allowed to suggest the soil properties (pH and organic matter solubility) and phases (organic matter, carbonates and clay minerals) governing the Sm-soil interaction. The sorption models constructed in the present work along with the sorption behaviour of Sm explained in terms of soil main characteristics will allow properly assessing the Sm-soil interaction depending on the contamination scenario under study. Moreover, the sorption and desorption  $K_d$  values of radiosamarium in soils were strongly correlated with those of stable Sm at low concentrations ( $r = 0.98$ ); indicating that the mobility of Sm radioisotopes and, thus, the risk of radioactive Sm contamination can be predicted using data from low concentrations of stable Sm.

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## 1. Introduction

Rare earth elements (REEs), especially lanthanides, have become important in agriculture and industrial activities because of their unique physicochemical properties (Awual et al., 2013). Increased anthropogenic inputs of REE in the environment through mining (Jinxia et al., 2010; Li et al., 2013; Liang et al., 2014), the discharge of industrial effluents (Ali, 2014; He et al., 2010) and the extensive application of REE-enriched fertilisers (Cao et al., 2001; Pang et al., 2001; Tyler, 2004; Zhang et al., 2000) have led to the contamination of watersheds and soil.

Trivalent lanthanide samarium (Sm) is a REE emerging contaminant that has been found in wastes resulting from agricultural and industrial activities (He et al., 2010; Pang et al., 2001). There are many evidences of watersheds that, due to contamination by industrial wastes, specially mine tailings, contain Sm at concentration levels several orders of magnitude higher than the water background value in Chinese rivers (around  $10^{-4} \mu\text{g L}^{-1}$ , that is,  $\approx 10^{-9} \text{meq L}^{-1}$ ) (Meng and Ji, 2008), such as the case of Baotou Area

containing up to  $130 \mu\text{g Sm L}^{-1}$  ( $2.4 \times 10^{-3} \text{meq L}^{-1}$ ) (Lu et al., 1995). Besides, some works have also reported that the Sm concentration in lands treated with REE-rich fertilisers or irrigated by the abovementioned contaminated liquid sources is abnormally higher, e.g.,  $25 \text{mg kg}^{-1}$  in Jiangxi area, China (Zhu et al., 2002) or  $492 \text{mg kg}^{-1}$  in Bayan Obo, China (Jinxia et al., 2010), compared to the soil background value in China ( $5 \text{mg kg}^{-1}$ ) (Wei et al., 1991) and the world average ( $1 \text{mg kg}^{-1}$ ) (Wang et al., 1989). This concomitant soil contamination is also of special concern since soils can act as sink of Sm that can be further incorporated in the food chain or reintroduced into water sources due to irrigation and raining processes (Liang et al., 2005). Consequently, there is increasing concern about the potential harmful effects of trivalent REE contamination on non-human biota and humans (Ali, 2014; Humsa and Srivastava, 2015; Jinxia et al., 2010; Li et al., 2013; Zhao et al., 2017; Zhu et al., 1997). Although trivalent REE present limited toxicity for humans, human exposure to high dose of these compounds can cause adverse effects such as pneumonitis, whereas long-term exposure can cause more harmful effects such as pneumoconiosis (Hirano and Suzuki, 1996) or abnormal neuro-behavioral development (Feng et al., 2005).

Furthermore, Sm radioisotopes are present in spent nuclear fuel and radioactive wastes (GRS, 2012), as the beta-emitting  $^{151}\text{Sm}$

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radioisotope with a relatively long half-life ( $T_{1/2} = 90$  years), thus being an element of concern in the mid-term (100–500 years) management of low- and intermediate-level radioactive wastes (Rego et al., 2011).

The interaction of trivalent lanthanides (Ln (III)) with geological media is mediated by the formation of surface complexes with clay minerals, metal oxides and organic matter (OM) solid particles, which depend on factors affecting Ln (III) aqueous speciation and sorption site availability, such as pH, organic matter solubility and mineralogy (Fan et al., 2010; McCarthy et al., 1998; Ramírez-Guinart et al., 2017; Takahashi et al., 1998; Ye et al., 2014). Ln (III) sorption by solid mineral particles increases with increasing pH due to the increasingly negative net surface charge and the predominating  $\text{Ln}^{3+}$  and cationic hydroxyl complexes (Fan et al., 2010; Geckeis and Rabung, 2008; Wenming et al., 2001). Although some studies have indicated a high affinity of Ln (III) for OM particles (Pang et al., 2001; Pourret and Martinez, 2009; Shan et al., 2002) and carbonate minerals like calcite or aragonite (Sutton, 2009), the presence of large amounts of dissolved humate compounds or carbonates may decrease Ln (III) sorption as Ln (III) can form soluble anionic humate and carbonate complexes (Dupré et al., 1999; Shan et al., 2002; Wenming et al., 2001; Xiangke et al., 2000; Zhong and Mucci, 1995). Moreover, the mobility of Ln (III) in a terrestrial ecosystem depends on its concentration (Galunin et al., 2009, 2010) and soil properties (Jones, 1997; Zhu et al., 1993).

There are few studies examining the effect of geochemical factors on the interaction of REEs with soil and sediments (Cao et al., 2000, 2001; Li et al., 2000, 2001; Ramírez-Guinart et al., 2017; Shan et al., 2002; Wang et al., 2011). To the best of our knowledge, none have simultaneously evaluated the effect of soil properties and REE concentration on their sorption-desorption behaviour. The objective of this study is to examine the interaction between Sm and soil samples with contrasting edaphic properties as a function of Sm concentration, assessing five types of soils to account for different contamination scenarios. The dependency of Sm sorption and desorption parameters on Sm concentration was examined by constructing Sm sorption isotherms, while the soil phases and mechanisms involved in Sm sorption were assessed by a sequential extraction procedure. Finally, the sorption and desorption parameters here obtained for low concentrations of stable Sm were compared with those gathered for Sm radioisotopes in a previous work from the same soils tested here (Ramírez-Guinart et al., 2017).

## 2. Materials and methods

### 2.1. Soil samples

For the present study five soil samples presenting contrasting

properties (see Table 1) were selected from a collection of soils for which the interaction of Sm radioisotopes was extensively studied in a previous work (Ramírez-Guinart et al., 2017). All samples were taken from the surface layer (0–10 cm), air-dried, sieved through a 2-mm mesh, homogenised with a roller table and stored in plastic bottles until analysis. The following physical-chemical characterisation was performed. Particle size distribution (clay, silt and sand percentages) was determined by the pipette method (Burt, 2004), except for DUBLIN soil, where the clay percentage was determined by X-ray diffraction, due to the high organic matter content of this soil. The carbonate ( $\text{CaCO}_3$ ) content was determined by using the calcimeter Bernard method (Mueller and Gastner, 1971). The loss on ignition (LOI) content was determined as the loss of soil weight by ashing 2 g of each soil sample (previously oven-dried overnight at 110 °C) at 450 °C for 16 h in a muffle furnace. The cation exchange capacity (CEC) was determined as the sum of exchangeable bases plus the exchangeable acidity obtained by displacement with  $\text{BaCl}_2$ -triethanolamine solution buffered at pH 8.2 (Burt, 2004) and the specific surface area of soils (SSA) was determined by  $\text{N}_2$  adsorption after degasification at 100 °C (Fagerlund, 1973). In addition, the solutions obtained after equilibrating the soil samples with deionised water under the same conditions as those of the sorption tests (see Section 2.2.) were also characterised in terms of pH, dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC), and concentration of major cations (Ca, Mg, K and Na). The DOC content was measured using a Total Organic Carbon analyser (Shimadzu TOC-5000 A) (Shimadzu, Japan) with a previous acidification with HCl to a pH of 3 to remove the carbonates in solution. The DIC content was calculated as the difference between the total carbon content, determined as the DOC but without the acidification step, and the DOC content. The concentration of major cations was determined by inductively coupled plasma optical emission spectrometry (ICP-OES) (Thermo-Jarrell Ash 25 and Perkin Elmer Optima 3200 RL, USA). Details about the methodologies used for the physical-chemical characterisation of soil samples are described in Ramírez-Guinart et al. (2016).

Most of the soil properties (see Table 1) varied within wide ranges, thus being representative of different environmental scenarios. CABRIL and ASCO are mineral soils with low amounts of organic matter (OM), the latter one being a calcareous soil with high carbonate content and a basic pH. ANDCOR and DUBLIN are organic soils with an acidic pH and high amounts of soluble organic matter, here represented with dissolved organic carbon content (DOC). Finally, DELTA2 has a basic pH and significant organic matter content, but moderate amounts of soluble organic matter. It had the highest contents of carbonate and clay (fraction of particles with size < 0.002 mm) among all the soil samples tested.

**Table 1**  
Summary of soil characteristics.

Soil sample	Solid phase							Liquid phase <sup>a</sup>						
	Clay (%wt)	Silt (%wt)	Sand (%wt)	$\text{CaCO}_3$ (%wt)	LOI (%wt)	CEC ( $\text{cmol}_c \text{ kg}^{-1}$ )	SSA ( $\text{m}^2 \text{ g}^{-1}$ )	pH	DIC ( $\text{mg L}^{-1}$ )	DOC ( $\text{mg L}^{-1}$ )	Ca ( $\text{mmol L}^{-1}$ )	Mg ( $\text{mmol L}^{-1}$ )	K ( $\text{mmol L}^{-1}$ )	Na ( $\text{mmol L}^{-1}$ )
DELTA2	33.5	31.3	12.2	51	23	87.3	6.5	8.0	32	39	2.0	0.70	0.13	4.0
DUBLIN	1.3 <sup>b</sup>	na	na	2	78	140	0.6	5.7	29	290	1.4	0.37	0.15	0.19
ANDCOR	18.3	23.6	39.1	2	19	54.2	4.5	4.9	1	78	0.10	0.37	0.08	0.13
CABRIL	20.0	16.5	57.5	2	6.0	19.3	9.5	6.4	3	10	0.12	0.12	0.05	0.05
ASCO	16.9	63.0	18.5	38	1.6	40.8	11.0	8.4	5	5	3.4	0.16	0.13	0.03

LOI, loss on ignition; CEC, cation exchange capacity; SSA, specific surface area; DIC, dissolved inorganic carbon; DOC, dissolved organic carbon.

Clay, silt and sand percentages are referred to the whole soil.

na: not analysed.

<sup>a</sup> Liquid phase refers to the supernatant obtained from sorption blank assays (see section 2.2).

<sup>b</sup> Clay content was obtained by X-ray diffraction.

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