



## Short communication

## Accumulation and fractionation of rare earth elements in atmospheric particulates around a mine tailing in Baotou, China



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## ARTICLE INFO

## Article history:

Received 16 October 2013

Received in revised form

29 January 2014

Accepted 30 January 2014

Available online 1 February 2014

## Keywords:

Rare earth elements

Mine tailing

Atmospheric particles

TSP

PM<sub>10</sub>

## ABSTRACT

Rare earth elements (REEs) have been increasingly emitted into the atmosphere with a worldwide increase in use of these metals. However, the research on REEs in atmospheric particulates is fairly limited. In this paper, atmospheric particulates including total suspended particulate (TSP) matter and particles with an equivalent aerodynamic diameter less than 10  $\mu\text{m}$  (PM<sub>10</sub>) were collected around a rare earth mine tailing in Baotou, the largest rare earth industrial base in China, in August 2012 and March 2013, for the analyses of REE levels and distributions. The total concentrations of REEs for TSP were 172.91 and 297.49  $\text{ng}/\text{m}^3$ , and those for PM<sub>10</sub> were 63.23 and 105.52  $\text{ng}/\text{m}^3$ , in August 2012 and March 2013, respectively. Enrichment factors for all 14 analyzed REEs in the TSP and PM<sub>10</sub> indicated that the REE enrichment in atmosphere particulates was caused by anthropogenic sources and influenced by the strong wind in spring season. The spatial distribution of REEs in TSP showed a strong gradient in the prevailing wind direction. The chondrite-normalized patterns of REEs in TSP and PM<sub>10</sub> were similar with the conspicuous fractionation between light REEs and heavy REEs.

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## 1. Introduction

Rare earth elements (REEs) are located at the bottom of Mendeleev's Periodic Table, including fifteen lanthanides, from La to Lu, Sc and Y (Liang et al., 2008). REEs are divided into heavy REEs (HREEs) and light REEs (LREEs) depending on their atomic masses and radii. LREEs include La, Ce, Pr, Nd, Pm, Sm and Eu, while HREEs include Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu (Zhang et al., 2001; Wang et al., 2011). These elements form a coherent group of elements with similar chemical properties and are often found together in mineral deposits. REEs have become vital and indispensable components of many high-tech products. It is reported that with a complete technological system in mining, dressing, smelting and separating of rare earth ores, China can produce over 400 varieties of rare earth products in more than 1000 specifications (Chen, 2011). In 2011, China has produced over 90% of the world's rare earth supply with only 23 percent of the world total reserves (Anonymous, 2012). The large-scale exploitation and application of rare earth resources will inevitably result in substantial increases of REE concentration levels in soil, water and air.

During the past decades, considerable research has focused on the concentration distribution of REEs in soils and water bodies (Zhang et al., 2001; Wang et al., 2008; Liang et al., 2008; Li et al., 2010). However, research on their concentrations in the air is fairly limited because of analytical limitations. Previous studies have investigated the long haul transport of REEs in atmospheric dust from continents to oceans (Lee et al., 2010; Csavina et al., 2012) and levels of REEs in the airborne particulate matter in urban areas (Wang et al., 2001; Kulkarni et al., 2006; Moreno et al., 2010; Shaltout et al., 2013). Air pollution caused by REEs in forms of ambient particles has been found during the combustion of oil in the vicinity of refinery factories (Kitto et al., 1992; Pacyna and Pacyna, 2001) and a steel plant (Geagea et al., 2007), because petroleum-cracking catalyst and products are highly enriched in LREEs (La, Ce and Nd).

Though being the non-essential elements for living organisms (Pang et al., 2001), REEs have positive effects on the growth of farming animals and various plant species (Pang et al., 2001; Hu et al., 2004). Consequently, REEs had been used at low concentrations as feed additives and fertilizers in China for decades (Wang et al., 2008). However, the environmental safety of the application of REEs has always been controversial and human health risk caused by rare earth intake has drawn increasing attention. Many researches have shown that REEs can enter human bodies and the presence of significant amounts of REEs in human bone, blood,

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teeth, liver, kidney and lymph nodes has been observed (Koeberl and Bayer, 1992; Zhang et al., 2000). Chen et al. (2001) reported that REEs could enter the cell and cell organelles and mainly be bound with biological macromolecules. The long-term consumption of food contaminated with REEs may cause chronic poisoning (Hirano and Suzuki, 1996; Jiang et al., 2012). Nevertheless, little information is available about the dose intake and potential health effects of exposure to REEs on human beings living in the rare earth mining and tailing areas.

The harmful effects of atmospheric particulates including total suspended particulate (TSP) matter and particles with an equivalent aerodynamic diameter less than 10  $\mu\text{m}$  (PM<sub>10</sub>) on human health are drawing more attention nowadays (Kulkarni et al., 2006; Moreno et al., 2010; Cheng et al., 2013). Previous studies have demonstrated an association between atmospheric pollutants and morbidity and adverse health outcomes such as hospital admissions for cardiovascular and respiratory diseases, urgent care visits, asthma attacks, acute bronchitis and restrictions in activities (Anenberg et al., 2010; Cheng et al., 2013). Atmospheric particulates contain a mixture of solid particles with organic and inorganic pollutants (Lee et al., 2010; Cheng et al., 2013) which are more harmful to human health than the particulates themselves. Along with a worldwide increase in the use of rare earths, these elements have increasingly been emitted into atmosphere. Therefore, it is of importance to analyze and evaluate the levels and distribution of REEs in the air.

Baotou REE tailing was produced by Baotou Iron and Steel Company, China's largest rare earths producer. It contains high levels of REEs and heavy metals from the Bayan Obo mine, the largest light rare earth deposit ever found in the world. Since the tailing is powdery and composed of various mineral matters, it is one of the major pollution sources for the surrounding environment (Guo et al., 2013). A large amount of tailing powders with high REE concentrations can enter the atmosphere easily due to the strong wind occurring frequently all year around in this region. The presence and accumulation of REEs in the atmospheric particulates may have serious consequences on the surrounding ecosystems as well as human health. To assess the potential environmental risk of REEs, in this study, we focused on the concentration and distribution of REEs in the atmospheric particulates in a representative open-air rare earth tailing region of northern China. TSP and PM<sub>10</sub> samples were collected around the mine tailing in August 2012 and March 2013, representing the warm and wet season and the cold and dry season in this area, respectively.

## 2. Materials and methods

### 2.1. Study area

Baotou (40°14'56"–42°43'49"N, 109°15'12"–111°26'25"E) is the capital city of Inner Mongolia which has been known as the largest rare earth industrial base in China since the discovery of the large Bayan Obo Nb–REE–Fe deposit in 1927 (Xu et al., 2008). It is located in the central part of Inner Mongolia, on the Tumochuan and Hetao Plain, with the Yellow River to the south and Mongolia to the north. It has a semi-arid, temperate, continental monsoon climate. The mean annual temperature is 6.5 °C. The average annual precipitation is about 240–400 mm, with an evaporation of 1938–2342 mm. The soil type is mainly chestnut soil. The prevailing wind direction is northwest. Because of the frequent exchange of warm and cold air, dust storms frequently occur in this area from March to May every year. The average wind speed is 3 m/s and the average numbers of days with strong wind, floating dust and the dust storm are about 46 days, 25.9 days and 43.3 days per year, respectively.

The REE ore was mined from the Bayan Obo deposit and transported about 150 km by railroad to milling facilities in Baotou (Xu et al., 2008). About 90% of the mined deposit was stored in the tailing of Baotou Iron and Steel Company, 12 km away from Baotou city for future use. The tailing powders were discharged into the reservoir through open slots by circulating water. The mine tailing was established in 1965 with poor supportive capacity (Guo et al., 2013). It covers an area of 11.5 km<sup>2</sup>, with tailings of  $1.5 \times 10^8$  t, of which about  $9.3 \times 10^6$  t are REE tailings (Li et al., 2010). Compared with the raw ore, the average grade of REEs in the tailings had been increased from 6.8% to 8.85%. After evaporation of water in the tailing reservoir, part of the tailing area is exposed to the air. The particle size of mineral powder was fairly fine and would easily spread to the surrounding environment with the strong wind. This process will inevitably result in accumulation of REEs in the surrounding environment and might affect human health through the food chain (see Fig. 1).

### 2.2. Sampling and measurement

A total of 13 sampling sites were chosen around the rare earth tailings to assess the impact on the surrounding environment. A global positioning system (GPS) was used to record the locations of the sampling sites. The TSP samples were collected for three days from 8:00 am to 8:00 pm during the periods of August 14th–20th, 2012 and March 14th–20th, 2013. The rainy days were avoided, and the sampling periods were chosen after 2 or 3 continuous sunny days. The TSP sampling was conducted using a mid-volume aerosol sampler (TH-150C, Wuhan Tianhong Instrument Co., Ltd, China) at the sampling rate of 100 L min<sup>-1</sup>. The sampling height was about 1.5 m. The filters used were quartz fiber filters ( $\varnothing$  90 mm) with low weight and low blank levels for REEs. Quartz filters were pre-heated (450 °C for 4 h) and pre-weighted (after conditioning for 24 h at 20–25 °C) prior to sample collection, and were stored in a refrigerator (2 °C) prior to sample collection. The filters were weighed (0.1 mg accuracy) before and after sampling to determine the total mass of collected TSP. The filters were sealed in the filter holder with a cover during transport to the sampling sites or back to the laboratory in order to avoid contamination. Meteorological parameters such as wind speed, wind direction, temperature and humidity were also simultaneously measured during the sampling period.

To measure the participants' PM<sub>10</sub> exposures during normal activities, 24-h personal PM<sub>10</sub> samples were simultaneously collected in a village Wulanji near the tailing reservoir during the same sampling period as the TSP. The personal air sampling-pump (Omni5000IS, Beijing Jinkesantong Instrument Co., Ltd, China) was used to collect PM<sub>10</sub> samples on 37-mm quartz fiber filters around the tailings. The 37-mm filters were pretreated (heating and weighting) in line with the 90-mm filters. At a nominal flow rate of 3 L min<sup>-1</sup>, approximately 4.32 m<sup>3</sup> of air was collected during each 24-h sampling period. Sixteen adult participants living in the Wulanji village were invited to wear the personal air sampling-pump to collect PM<sub>10</sub> samples for 24 h from 8:00 am first day to 8:00 am next day. The sampling inlet was positioned in the breathing zone of each participant at the midline of their chest.

After sampling, all the filters were weighed after equilibrated in a desiccator for 48 h and the net weight of the sampled TSP and PM<sub>10</sub> can be obtained by subtraction. The filters loading samples were cut into small pieces using plastic scissors and subsequently leached for 4 h with 43% HNO<sub>3</sub> (super pure). The leachate was filtered and transferred to a flask and diluted to 100 ml with de-ionized water. The concentrations of REEs and Al were analyzed by inductively coupled plasma mass spectrometry (ELAN DRC-e, Perkin Elmer SCIEX) (Wang et al., 2011). Each measurement was

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