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Atmospheric thorium pollution and inhalation exposure in the largest rare earth mining and smelting area in China

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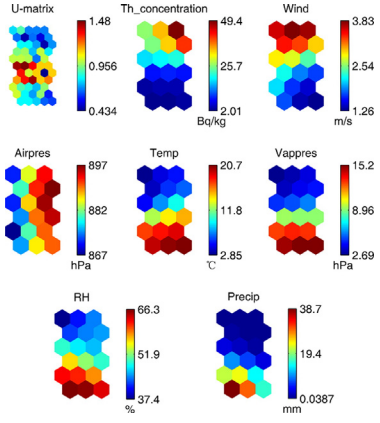
HIGHLIGHTS

- Atmospheric thorium pollution was investigated around the Bayan Obo rare earth mine.
- ^{232}Th concentrations were significantly higher than the world reference of 0.5 μBq m^{-3}.
- A self-organizing map was used to identify spatiotemporal pattern of airborne thorium.
- The inhalation exposure results show a high radioactive risk for local dwellers.

GRAPHICAL ABSTRACT



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ABSTRACT

Exposure to radionuclide thorium (Th) has generated widespread public concerns, mainly because of its radiological effects on human health. Activity levels of airborne ^{232}Th in total suspended particulate (TSP) were measured in the vicinity of the largest rare earth mine in China in August 2012 and March 2013. The mean activity concentrations of ^{232}Th in TSP ranged from 820 μBq m^{-3} in a mining area in August 2012 to 39,720 μBq m^{-3} in a smelting area in March 2013, much higher than the world reference of 0.5 μBq m^{-3}. Multistatistical analysis and Kohonen's self-organizing maps suggested that ^{232}Th in TSP was mainly derived from rare earth mining and smelting practices. In addition, personal inhalation exposures to ^{232}Th associated with respirable particulate (PM_{10}) were also measured among local dwellers via personal monitoring. The mean dose values for different age groups in the smelting and mining areas ranged from 97.86 to 417 μSv year^{-1} and from 101.03 to 430.83 μSv year^{-1}, respectively. These results indicate that people living in the study areas are exposed to high levels of widespread ^{232}Th.

1. Introduction

Thorium (Th) is a primordial radioactive element which is ubiquitous in the environment at varying concentrations. >99% of natural thorium exist in the form of ^{232}Th, which has a long half-life

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(1.39×10^{10} year) and low specific activity (4.1 kBq g^{-1}) (Höllriegl et al., 2007). Numerous investigations over the past decade have focused on the ^{232}Th concentration in rocks, soils, water bodies and sediments and the potential external radiation dose to the public (Cowart and Burnett, 1994; Kritsananuwat et al., 2015). However, research on airborne ^{232}Th activity concentrations is still limited because of analytical limitations.

Thorium can be released into the atmosphere from both natural and anthropogenic sources that elevate its level over the background, including substances in Earth's crust, cosmic rays, volcano eruption, nuclear fuel plants, coal combustion, and ore mining and refining (Cowart and Burnett, 1994; Alvarado et al., 2014). Atmospheric particulate matter (PM) contaminated with ^{232}Th are harmful to human health due to increased radiotoxicity and chemotoxicity. Studies revealed that ^{232}Th if inhaled as a dust or accumulated in human body increased the risk of lung and liver diseases, lung and pancreatic cancer, adverse effect on blood and changes in human genetic material (Soudek et al., 2013). It is estimated that more than half of total radioactive dose received by world's population is associated with inhalation of radioactive aerosol particles (United Nations Scientific Committee on Effects of Atomic Radiation, UNSCEAR, 2000). Therefore, it is of importance to investigate the activity level of Thorium in aerosols, particularly the respirable PM (PM_{10}), which contributes significantly to internal radiation dose and may subsequently cause adverse health effects.

Nearly all rare earth ores contain the radioactive elements thorium and uranium (Binnemans and Jones, 2015). The Bayan Obo deposit in North China contains the largest rare earth resources in the world. Its rare earth mineral is concomitant with natural radiothorium in the ThO_2 with a mean concentration of 0.04% by weight (Chen et al., 2005). Since the discovery of the main ore body in 1927, mining activities had been carried out over 90 years. Intensive exploitation activities of rare earth resources have caused the substantial increases of ^{232}Th contents in the environment (Li et al., 2014). Thorium was released into the atmosphere as aerosol particles from mining, milling, processing operations of rare earth minerals, and particle resuspension from tailing piles. Residents living around the mining and smelting areas are likely exposed to the radioactive aerosols with ^{232}Th through inhalation, ingestion of food and water contaminated by the fallout of particles, and dermal contact with fallout.

Kohonen's self-organizing maps (SOM), which is based on an unsupervised neural network model, can serve as an important tool for pattern recognition and classification without preliminary knowledge of the process (Mukherjee, 1997; García and González, 2004). SOM potentially outperforms other multivariate techniques such as principle component analysis for its visualizing interpretation, powerful clustering capability, and ability to deal with nonlinear problems (Hong et al., 2003; Aguado et al., 2008). SOM has been frequently used for pollutants pattern recognition in various environmental media (Lee and Scholz, 2006; Nadal et al., 2006; Gulson et al., 2007; Mari et al., 2010). In the present study, aerosol samples of total suspended particulate (TSP) and PM_{10} were collected from two sampling sites in the largest rare earth industry city, one in the Bayan Obo rare earth mining area and the other in the nearby smelting areas. We used the SOM model to identify the most impacted zones and to assess the potential correlations between thorium in aerosols and meteorological factors. Such research will provide a basis for understanding the impact of thorium emissions on the atmospheric environment and human health due to the rare earth smelting and mining activities.

2. Materials and methods

2.1. Study area

Baotou city (Fig. 1, 40°14′56″–42°43′49″N, 109°15′12″–111°26′25″E) is the largest rare earth industrial base and accounts for >70% of total rare earth production in China (Zhu et al., 2015). This region has a typical arid continental climate, characterized by windy and dry winter and spring, and warm and comparatively wet summer followed by short and cool autumn (Li et al., 2002). Mean annual temperature is around 6.5 °C, and the lowest and highest monthly mean temperatures are −11.1 °C in January and 23.3 °C in July, respectively. The average annual precipitation is about 240–400 mm, of which over 67% falls in the summer months (June–August). Mean annual evaporation is around 1938–2342 mm, several times greater than the annual precipitation. Prevailing wind directions are northwest in winter and spring, and southwest to south in summer and autumn. On a 50-year average, the annual wind speed is 3.1 m s^{-1} and the number of days with strong wind (≥17 m s^{-1}), floating dust, and the dust storm are about 46, 26 and 43 per year, respectively. After the cold and dry winter, aeolian sand activity is strong and frequent in spring (March to May) because of high air pressure gradients between the Siberian mainland and East Asia.

The giant Bayan Obo Fe–RE–Nb deposit comprises >170 different minerals. Its total reserves have been reported as at least 1.5 billion tonnes of iron (average grade 35%), 48 million tonnes of rare earth oxides (average grade 6%), and about 1 million tonnes of niobium (average grade 0.13%) (Wu, 2008). The concentrates after ore beneficiation used for rare earth production usually contain about 50–60% REO and 0.18–0.3% ThO_2 (Zhu et al., 2015). It is estimated that refining one tonne of rare earth oxide can potentially produce 6×10^4 m^3 of waste gas, 200 m^3 of acidic water and 1.4 tonnes of radioactive waste (Hao et al., 2015). Reports indicate that 96%–98% of Bayan Obo's thorium ends up in solid waste, 0.1%–0.5% is emitted as exhaust gas, and 0.6%–2.0% goes to liquid effluents (Ault et al., 2015). Long-term mining activities have produced a large amount of tailings which discharged into the reservoir through open slots by circulating water. The openly dumped tailing covers an area of 11.5 km^2 with thorium-radioactive tailings of 1.5×10^8 tonnes. After water is evaporated from the tailing pond, part of the tailing area is exposed to the air. The tailing powder was easily spread to the surrounding environment with strong wind. This process will inevitably result in elevated levels of ^{232}Th in the surrounding areas and cause adverse effects on human health (Chen et al., 2004). Therefore, it is necessary to characterize and quantify the natural radioactive content in TSP and PM_{10} to evaluate the associated environmental and human health risks.

2.2. Sampling and measurement

TSP samples were collected from 5 sites (Y01–Y05) in the rare earth mining area and 13 sites (B01–B13) in the smelting area in Baotou (Fig. 1). The intelligent medium-volume air sampler (TH-150C, Wuhan Tianhong Instrument Co., Ltd., China) and 90-mm quartz microfiber filters were used to collect TSP samples. The sampling flow rate was fixed at 100 L min^{-1}. At each site, 12-h samples were collected for three days from 8:00 am to 8:00 pm in 14th–20th August 2012 and 16th–22th March 2013, respectively. The sampling periods were chosen to avoid rainy days and most of the sampling at different sites was performed concurrently. A total of 104 TSP samples were collected from three consecutive days at both sampling sites.

To evaluate the radiation risk of inhalation exposure, 10 volunteers from the Kuangshan community near the mining area and 9 volunteers from the Wulanji community around the smelting area were randomly selected to wear a personal inhalation exposure sampler with a 10-μm cut-size (Omni5000IS, Beijing Jinkesantong Instrument Co., Ltd., China) to collect PM_{10} samples. Each sampler (flow rate 3 L min^{-1}) was placed at the breathing zone of the volunteers for 24 h from 8:00 am to 8:00 am the next day. Three samples were obtained from each volunteer for three consecutive days. The PM_{10} samples were collected on 37-mm quartz microfiber filters.

Prior to use, the filters of both Φ90 mm and Φ37 mm (Pore size = 0.2 μm, retention >99%, MK360, Munktell, Sweden) were pre-baked in a muffle furnace at 450 °C for at least 4 h and then maintained in a refrigerator (2 °C) for over 24 h and weighed before and after sampling. The mass