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Pollution level and inhalation exposure of ambient aerosol fluoride as affected by polymetallic rare earth mining and smelting in Baotou, north China

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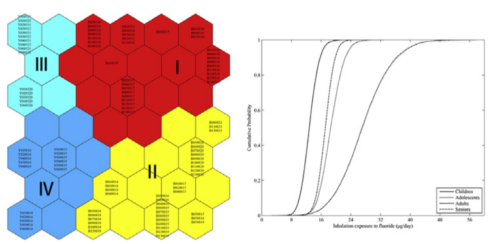
HIGHLIGHTS

- F^- concentrations bound to TSP were higher in smelting than in mining area.
- Self-organizing maps analyze link between F^- contents and meteorological factors.
- Four clusters of samples are identified on the basis of sampling periods and sites.
- The exposure level of fluoride inhaled for population were relatively high.

GRAPHICAL ABSTRACT



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ABSTRACT

Airborne fluoride associated with total suspended particles (TSP) and respirable particulate (PM_{10}) in the rare earth mining and smelting areas were analyzed during August 2012 and March 2013. In March, average concentrations of fluoride bound to TSP in the mining and smelting areas were 0.598 ± 0.626 μg/m^3 and 3.615 ± 4.267 μg/m^3, respectively, whereas that in August were 0.699 ± 0.801 μg/m^3 and 1.917 ± 2.233 μg/m^3, respectively. TSP samples were classified into four categories by different sampling periods and locations using Kohonen's self-organizing map, which demonstrates that high airborne fluoride concentrations in March in the smelting area were probably attributed to industrial emissions from smelting activities and wind-blown dust from tailings pond, influenced by meteorologic parameters such as temperature, relative humidity, precipitation and wind speed. The mean daily amount of fluoride inhaled in the mining and smelting areas were estimated to be in the range of 2.77–57.61 μg/day and 3.39–64.32 μg/day, respectively. These results indicate the high potential exposure level of fluoride inhaled for local residents in the polymetallic mining and smelting areas.
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1. Introduction

Fluorine is the lightest member of the halogen group and it is an extremely reactive electronegative element that can form compounds with almost any other element (Ozsvath, 2009; Pyle and Mather, 2009; Jelenko and Pokorny, 2010; Yasmin et al., 2011).

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Fluorine is widely distributed in the environment and is the 13th most abundant element in the earth crust with a mean concentration of ca. 550 ppm (Pyle and Mather, 2009; Pagariya et al., 2013). Fluorine always occurs in combined form (fluoride, F^-) in minerals including fluorite (CaF_2), sellaite (MgF_2), fluorapatite ($Ca_5(PO_4)_3F$), cryolite (Na_3AlF_6), villiaumite (NaF), and topaz ($Al_2(SiO_4)F_2$) (Brahman et al., 2013; Pettenati et al., 2013; Rafique et al., 2015). In addition to natural processes such as volcanic emission, fluoride can be released into the atmospheric environment from numerous industrial processes including mining, refining and smelting (Mezghani et al., 2005; Franzaring et al., 2006; Jelenko and Pokorny, 2010; Cao et al., 2013; Kierdorf et al., 2016). Atmospheric fluoride may be in gaseous (such as hydrogen fluoride, sulfur hexafluoride and silicon tetrafluoride) or particulate forms (such as sodium aluminum fluoride, calcium phosphate fluoride and aluminum fluoride) (Feng et al., 2003; Ahmad et al., 2012; Ozbek et al., 2016).

Although fluorine has beneficial effects on the mineralization of bones and the formation of enamel in teeth at low concentrations, this element is generally not considered as essential for human and animals (Oliver and Gregory, 2015). The prolonged exposure to fluoride at high levels can lead to fluorosis, a considerable worldwide health problem (Amini et al., 2008; Ozsvath, 2009; Varol et al., 2010; Brahman et al., 2013; Rafique et al., 2015). China has more than 40 million dental fluorosis patients and 2.6 million skeletal fluorosis patients, much more cases of fluorosis than any other country (Dai and Ren, 2006; Chen et al., 2012; Liu et al., 2015). The epidemiological and experimental work confirm that excessive fluoride intake also has a detrimental effect on the gastrointestinal tract, kidneys, and liver, as well as the nervous, reproductive, and immune systems (Xiong et al., 2007; Amini et al., 2008; Brahman et al., 2013; Taghipour et al., 2016; Wei et al., 2016).

The dispersion of fluoride in the environment continues to be of great concern because of its hazardous effect on ecosystems and public health (Pan et al., 2013; Rango et al., 2014; An et al., 2015; Choi et al., 2015). Many studies focused on chronic fluoride exposure from drinking water, in which it is considered beneficial at levels below 0.7 mg L^{-1} but hazardous if it exceeds 1.5 mg L^{-1} (Brahman et al., 2013; Augustsson and Berger, 2014; Craig et al., 2015). Atmospheric fluoride pollution is potentially another pathway of human exposure, especially in the historic mining and smelting areas. Fluoride in aerosols can be transported over large distances by wind or as a result of atmospheric turbulence (Lewandowska et al., 2013).

Bayan Obo is a giant polymetallic rare earth element (REE)-Fe-Nb ore deposit of hydrothermal origin in Baotou in north China. The fluorite is predominantly found in the Main and East orebodies, while most other orebodies contain 1–10% fluorite (Yao et al., 2013). The processing and refining in multiple steps produce large amounts of fluoride after calcining bastnasite (Navarro and Zhao, 2014; Zhu et al., 2015). Most previous studies that examined fluoride levels have focused on hydrogeochemical characteristics (Huang et al., 2016). Massive release of fluoride to the environment has been an issue of considerable public concern in this region (Wu et al., 2011). The estimated anthropogenic emissions of fluoride in Baotou urban area amount to 2657 tons/yr, which could threaten the health of surrounding habitants (Yao et al., 2013). In this study, the fluoride concentrations bound to the total suspended particles (TSP) and personal exposure to respirable particulates (PM_{10}) were concurrently monitored by first time in Baotou. A Kohonen's self-organizing map (SOM) enables pattern recognition and classification of aerosol samples by interpreting correlation patterns among fluoride concentrations in aerosols and meteorological conditions. The second objective was to make a preliminary evaluation of the magnitude of human exposure to fluoride via inhalation. The results from this study will provide a better knowledge on the fluoride pollution sources in order to improve air quality polices, which may further pose a public health concern in this region.

2. Materials and methods

2.1. Study area

Baotou is a large REE-Nb-Fe mining and smelting region, located on the Tumochuan and Hetao plain in north China (Fig. 1). This region has a continental semi-arid monsoon climate, which is characterized by windy and dry winter and spring, and warm and relatively rainy summer followed by a short and cool fall (Li et al., 2002). The annual temperature variations are high with an annual mean of 6.5 °C. The annual rainfall amount is approximately 378 mm, decreasing from the southeast to the northwest of Baotou (Wu et al., 2013). The dominant wind comes from the north-west and, to a lesser extent, from the north and the northwest. The annual wind speed is 3.1 m s^{-1}, meanwhile seasonal wind weather like strong wind ($\geq$17 m s^{-1}), floating dust, and dust storm frequently occurs from March to May in the study area.

The prevalence of endemic fluorosis is found in Baotou, probably attributed to the long-term exposure to high-fluoride drinking water and inhalation of fluoride borne aerosols. Fluoride is a particular air pollutant in Baotou, which was mainly emitted from industrial sources, including steel plant, aluminum plants, rare earth production and thermal power plants. The gaseous and particulate fluoride is released into the atmosphere in the processes of beneficiation and smelting for iron ore and of electrolytic extraction for aluminum ore (Jin, 1995; Kalinić et al., 1997). The fluoride emissions from the steel plant and aluminum production, accounting for 50.6% and 26.3% of total industrial emissions, respectively, subsequently fall out and create a considerable pollution problem in this region. Therefore, it is necessary to characterize and quantify the fluoride concentrations in TSP and PM_{10} to evaluate the associated environmental and human health risks.

2.2. Sampling and measurement

Sampling of fluoride associated with total suspended particles was conducted at two locations in Baotou (Fig. 1). These locations were chosen to represent two different environmental characteristics: (1) the smelting area located in the west of Baotou urban area, where the fluoride borne aerosol is impacted by various emission sources originated from steel plant, electrolytic aluminum production, and rare earth smelting. In addition, the tailing powders of polymetallic ores smelting are usually discharged into the open dumps covering an area of 12 km^2. Since the tailing is powdery and its quantity is large, it can be easily blown away by wind storms and it is one of the major sources for atmospheric particulate matter pollution, especially during the dust outbreak (March to May) in this region; (2) The mining area, is situated in Bayan Obo deposit, at about 150 km north from Baotou urban area. TSP samples were collected at six sites in the mining area and thirteen sites in the smelting area during 14th~20th August 2012 and 16th~22nd March 2013, respectively. Sample device was a TH-150C type TSP medium-volume aerosol sampler (Wuhan Tianhong Instrument Co., Ltd, China), with the flow rate of 100 L min^{-1}. Samples for TSP and fluoride analysis were collected on 90 mm diameter, 0.2-μm pore size Quartz fiber filters. Sampling at each of the 19 sites was done mostly concurrently between three consecutive days from 8:00 a.m. to 8:00 p.m. in both August and March.

To estimate inhalation exposure to fluoride, personal PM_{10} samples were simultaneously collected during TSP sampling. A total of 19 adult volunteers (10 males and 9 females) ranging from

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