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Isotopic ratio based source apportionment of children's blood lead around coking plant area

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ABSTRACT

Lead exposure in the environment is a major hazard affecting human health, particularly for children. The blood lead levels in the local children living around the largest coking area in China were measured, and the source of blood lead and the main pathways of lead exposure were investigated based on lead isotopic ratios ($^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$) in blood and in a variety of media, including food, airborne particulate matter, soil, dust and drinking water. The children's blood lead level was 5.25 (1.59 to 34.36 as range) μg dL^{-1}, lower than the threshold in the current criteria of China defined by the US Centers for Disease Control (10 μg dL^{-1}). The isotopic ratios in the blood were 2.111 ± 0.018 for $^{208}Pb/^{206}Pb$ and 0.864 ± 0.005 for $^{207}Pb/^{206}Pb$, similar to those of vegetables, wheat, drinking water, airborne particulate matter, but different from those of vehicle emission and soil/dust, suggesting that the formers were the main pathway of lead exposure among the children. The exposure pathway analysis based on the isotopic ratios and the human health risk assessment showed that dietary intake of food and drinking water contributed 93.67% of total exposed lead. The study further indicated that the coal used in the coking plant is the dominant pollution source of lead in children's blood.

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1. Introduction

Lead (Pb) is a well-established environmental pollutant that exerts neurotoxic effects on human health (Huang et al., 2012). It is reported that lead exposure is one of the important 67 risk factors contributing to the global burden of disease (Lim et al., 2012). High exposure to lead can seriously damage the kidney, liver, central nervous and hematologic systems (Counter et al., 2008; Sun et al., 2006). The US Centers for Disease Control (CDC) reported that there was no safe level of exposure to lead (Koller et al., 2004; Lanphear et al., 2005). Recent studies have also found negative health effects of Pb after low dose exposure (ATSDR, 2007; Grandjean, 2010; Jusko et al., 2008; Lanphear et al., 2005). Children in particular are more vulnerable to Pb contamination (Fulton et al., 1987; Jain and Hu, 2006; Safi et al., 2006; Winneke et al., 1990), due to their frequent hand-to-mouth behavior, higher intake rate and undeveloped neural system (He et al., 2009). In China, Pb contamination is also of widespread concern, particularly with the increased number of Pb poisoning incidents on children.

Dietary ingestion and inhalation are two main exposure pathways of Pb after its release into the environment (Díaz-Somoano et al., 2009), and the predominant exposure media include dust/soil, food, drinking water, and inhaled aerosol particles (USEPA, 2006). It is accepted that the principal sources of lead pollution are anthropogenic lead emissions, derived primarily from industrial activity and traffic emission (Li et al., 2012; Wang et al., 2000). Vehicle emission was one large source of anthropogenic Pb before the leaded gasoline was phased out (Ewing et al., 2010; Niisoe et al., 2010). After the phase out of leaded gasoline, it was established that coal combustion has become the principle source of atmospheric Pb emissions (Li et al., 2012). In some regions of China, lead in children's blood is derived mainly from coal-fired ash (Liang et al., 2010). Therefore, the lead contamination of children living around coal-related activities and its potential pollution sources should be a focus of research. However, most previous studies regarding coal-related industries have focused on the PAH pollution and heavy metals in air or soil, and few surveys on children's blood lead level (BLL) and the possible pollution sources have been conducted.

For lead toxicity monitoring and the identification of endogenous and exogenous lead exposure, combining with environment media

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samples, matrixes including deciduous teeth (Gulson et al., 1997, 2004), bone (Gulson and Gillings, 1997; Gulson et al., 2003), urine (Gulson et al., 2000; Manton et al., 2000), and blood (Gulson et al., 2001, 2006) were studied. Considering the feasibility of sample collection and analysis in environmental health investigation, whole blood lead level is regarded as the most representative indicator of the current environmental and body lead contamination level (Barbosa et al., 2005; Batariova et al., 2006; Gil et al., 2011; Liu et al., 2009; Yan et al., 2013). Lead isotopes are not measurably fractionated in chemical and biological processes after geological formation, allowing lead isotope ratios (LIRs) to be used as a reliable index to trace lead contamination and pollution sources (Komárek et al., 2008; Widory et al., 2004; Zheng et al., 2004). Therefore, lead content and its isotope composition provide an ideal tool for identifying the sources and pathways of pollution (Ault et al., 1970; Graney et al., 1995). Gulson (2008) and Gulson et al. (2012) have summarized the applications of lead isotopes in environmental health, such as source apportionment, pathways of lead in diverse environments from urban to mining communities, as well as various applications associated with pregnancy, the contribution of bone lead to blood lead in different periods, which confirm the successful applications of LIRs with emphasis on human investigation.

The coal industry is well represented in Shanxi Province of China, with numerous coal-related enterprises, especially coking plants. Alongside the economic development, the industry has generated substantial environmental lead pollution, and was ranked as the third largest contributor to atmospheric lead emissions over the period of 2005–2009 (Li et al., 2012), this pollution threatened the health of local children (Shi et al., 2003). To investigate the level of lead poisoning in children living in the coking area, we selected a town in which the largest coking plant in Shanxi Province is situated as a typical model for assessment of BLLs in children. We then attempted to identify the primary source of lead exposure to children by analyzing the relationships of LIRs found in children's blood, atmospheric particles, dusts/soil, drinking water and food consumed by the children, and in the coal used in the coking plant.

2. Materials and methods

2.1. Study area

Shanxi Province is the largest coking coal resource base in China, the proven reserves of coking coal resources is 124.59 billion tons, accounting for 51.76% of the national resources and 57.60% of the proven reserves in the province coal resources (Hu et al., 2006). Coking industry is the pillar industry for economy in Shanxi Province. The coking production in Shanxi Province accounted for 40.00% of the national output in 2005, accounting for 20.00% of the world. The coking exports in Shanxi Province accounted for 80.00% of the national total exports, accounting for 48.00% of the world (Zhao, 2005). The studied coking plant is the largest one of those in China, and is located in a town in the southwest of Shanxi Province. The historically state-owned coking industry in China plays an important role in the economy. The clean coal used as raw material in the coking plant, is coming from the local coking coal resources in Shanxi Province. With 4.26 million tons of coking coal consumed in the coking plant, 3.60 million tons of coke are produced per year (http://www.sxcc.com.cn/html/xwdt/jtxs/index.html). The coking area is surrounded by a residential district, a primary school, and farmland. Except for the coking plant, there is a well-known scenic area in the town, in which neither large enterprises nor large highway carrying motor vehicle are present.

2.2. Sample collection and preparation

2.2.1. Biological samples

Before the survey and sampling, written informed consents to participate in the investigation were obtained from 72 subjects who were native-born and aged from 7 to 12 years. For each child, 4 ml of cubital fossa venous blood were taken from the arms and drawn into vacutainers containing sodium heparin anticoagulant. After being thoroughly shaken, a 1 ml portion of the whole blood sample was placed into an acid-cleaned Teflon tank and subjected to cold digestion in 2 ml of 65% concentrated HNO_3 for 30 min, followed by digestion in 1 ml H_2O_2 for a further 10 min. The cold-digested solutions were then digested using the following three-stage program: Stage 1, heating to 150 °C over 10 min with 50% power; Stage 2, heating to 180 °C over 20 min with 80% power; and Stage 3, cooling to 100 °C over 10 min with 40% power. Digestion solutions were then evaporated to near dryness after being transferred into acid-cleaned quartz digestion cups, and finally dissolved and diluted to 20 ml with ultrapure water (MilliQ A13, Millipore, U.S.). The solutions were stored in a fridge at 4 °C prior to analysis.

2.2.2. Soil and dust

Soil samples (500 g) were collected from the upper soil layer (0–20 cm) of five sub-locations within a 20 × 20 cm grid at 12 sites, located around the coking plant. Sampling points were selected in undisturbed locations. After being air-dried at room temperature and ground in an agate mortar and sieved through a nylon sieve into 100-mesh powders, the soil samples were prepared with an acid digestion according to the procedure described by Zhang et al. (2010). The digested solutions were then dissolved and diluted to 50 ml for analysis. For dust samples, because the local children spent half of their day at school and at home, two samples were collected inside their homes and two from the floor inside the classroom and on the stairs of the primary school. And another one dust sample was collected inside the coking plant to identify the effect of coal combustion to the dust. Each dust sample was mixed with four or five random sub-samples. The preparation procedures for the dust samples were identical to those used for soil samples.

2.2.3. Drinking water

There is only one company supplying drinking water in the study area, and the water originates from an ancient spring. Therefore, tap water samples were randomly collected from 11 houses of children participating in the study. After adding two drops of 65% concentrated HNO_3, a portion of each sample was stored in a refrigerator at 4 °C prior to analysis. A further portion of each sample was transferred into Teflon vial, evaporated to dryness at 110 °C, and dissolved with ultrapure water to 20 ml for the determination of isotopic compositions.

2.2.4. Airborne particulate matter

Samples of total suspended particle (TSP), fine particulate matter (PM2.5) and particulate matter (PM10) were collected at locations in the school, residential area, coking plant and commercial district. The coking plant was considered to represent the experimental group and the others were set as a control group. Samples of TSP were collected on pre-combusted (500 °C, 6 h) quartz microfiber filters (90 mm, Munktell Inc., Sweden) using a large-volume air sampler (Laoshan Instruments Co. Ltd., Qingdao, China) with a flow rate of 100 L min^{-1}. Samples of PM2.5 and PM10 were collected on pre-combusted quartz microfiber filters (37 mm) using a low-volume sampler (Buck Libra Plus, AP Buck Inc., U.K.) with a flow rate of 2 L min^{-1}. Each quartz microfiber filter was cut into fragments and then digested using the established procedures (Chen et al., 2005).

2.2.5. Food

Seventeen locally-produced vegetable species (1 kg) were selected from three farmer's markets, and each variety of vegetables was integrated into one sample. Four wheat samples (the staple food) were also collected. After being thoroughly washed, the edible parts of food samples were cut into small pieces and subjected to vacuum freeze drying at −75 °C for 2 d. The freeze-dried samples were then ground

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