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Exposure risk of young population to lead: A case study in Le'an River Basin in Jiangxi Province, China

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ABSTRACT

Blood lead (Pb) level of children has widely been attracting public concern in China, particularly in the sites near mining or industrial areas. However, the policies about how to efficiently reduce the Pb intake of children are still under discussion. We collected six food types based on the local dietary habits and soils from Dexing, Leping, and Poyang Counties situated along the Le'an River Basin from upstream to downstream, and their Pb contents were analyzed. A Monte Carlo model was used to simulate the dietary chronic daily intake of Pb (CDI_{Pb}) from various foods and ingested soil by hand-to-mouth activities and its non-carcinogenic risk to children indicated by hazard quotient (HQ). Only in the rural area of Dexing, its soil and vegetables both had higher Pb content than the national tolerance limits of China, resulting its the highest CDI_{Pb} among all the areas. The Pb contents of the six food types and soils in other sites were overall below the limits. Vegetables and rice accounted for from 63% (Leping, urban) to 85% (Dexing, rural) of the total CDI_{Pb} and ingested soil overall took up ~6%. In the rural area, Dexing had the highest proportion (82.8%) of children with HQ > 1, followed by Leping (36.1%) and Poyang (27.7%). Different order was found in the urban areas, i.e. Dexing (46.7%) > Poyang (41.0%) > Leping (26.4%). Vegetables and rice were overall the two major contributors to the total CDI of Pb, which should be focused on to control the Pb intake by the local children, especially for those living in the rural area of Dexing County.

1. Introduction

The lead (Pb) poisoning has been attracting wide public concern in China due to higher portion, i.e. 23.9% (3.2%–80.7%), of Chinese children with blood lead level (BLL) above the tolerance limit (100 μg L^{-1}) than U.S.A, Canada and other developed countries (He et al., 2009; Wang and Zhang, 2006). Pb, as an important environmental toxicant, may decrease intelligence, growth and hearing, and cause anemia, attention deficits and behavioral problems for children, particularly for pre-school aged children (Koller et al., 2004). Mining and metal smelting are regarded as the two most important anthropogenic Pb emission sources (Kabala and Singh, 2001; Rodriguez et al., 2009). Le'an River Basin is one of the most important mining zones in China with high productions of heavy metals, like copper, lead, zinc, gold, and silver (Liu et al., 2013a). The annual emission of Pb from various industries were about 6500 tons, of which metal smelting accounted for ~85%, following by Pb–Zn mining (~12%) and other manufacturing industries (~4%) estimated by the Environmental Protection Department of Jiangxi Province. Le'an River, as the primary river of the Le'an River Basin, flows through Dexing, Leping, and Poyang counties to Poyang Lake, the largest freshwater lake in China. During the processes of mining and metal smelting, mineral waste residue, wastewater, and dust containing Pb are discharged into the environment. The agricultural soil close to mining tailings and smelting factories, especially along the downstream of the industrial areas, is vulnerable to be polluted by flooding or irrigation (Cao et al., 2014; Fu et al., 2008; Liu et al., 2013a; Zhao et al., 2012), may resulting in the elevation of Pb in the agricultural plants, and further livestock, poultry, and human that consume contaminated plants (Gupta and Gupta, 1998). It was reported that the Pb content in the scalp hair, indicating population accumulated internal exposure level of Pb, of

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residents living close to Dexing Copper Mine area was significantly higher than those free from any mining and other industrial pollution (Ni et al., 2011). Therefore, it's very necessary to investigate characteristics of the exposure risk by Pb of children living in the Le'an River Basin, which can be referred for other mining areas to protect children's health.

The main exposure routes to Pb for children include diet, aerosol inhalation, hand-to-mouth activities, and dermal contact. A case study in Shanxi Province, China showed that the dietary foods and ingested soil by hand-to-month activities accounted for >90% of the total Pb intake and others for a small portion (Cao et al., 2014). In Europe, the relative contributions of the diet exposure routes to blood Pb levels in pre-school children were ~70% (Bierkens et al., 2012). For the children in U.S.A, it was estimated that diet route is a predominant source of Pb exposure, followed by consumption of beverages and tap water (USEPA, 2006). Generally, the ingestion of vegetables and cereals take up a majority of Pb intake from diet, but their contribution patterns varied with local dietary habit (Li et al., 2012). For example, vegetables and corn accounted for ~75% of the total Pb intake from diet in Huludao City, China (Zheng et al., 2007), while vegetables and wheat for about 99% (Cao et al., 2014) for children. It was reported that great differences of diet habit existed between the rural and urban area (Zhai and Yang, 2006). The children living in the rural area consume more rice and vegetables, while less fish, meat, eggs and milk than in the urban area. Hence, the collection of which foods to indicate the Pb exposure risk should be based on the dietary habits and food sources of the concerned population. The spatial variance in the Pb concentrations of agricultural soil along the Le'an River can give rise to different contamination patterns of Pb in agricultural crops and foods. To control the levels of Pb exposure in the local population, knowledge of the relative contributions of different exposure sources to children is required.

For the local young population living in Le'an River Basin, the aims of this study were to investigate: 1) the Pb levels of various food types and soil; 2) the relative contributions to chronic dietary intake of Pb from various food categories and ingested soil by hand-to-mouth activities; 3) the non-carcinogenic health risk induced by Pb intake.

2. Materials and methods

2.1. Study area and sampling

The locations of the main mining zone and metal smelting factories in the Le'an River Basin in Jiangxi Province, China, are shown in Fig. S1 (Supplementary Materials). Both of the rural and urban areas of Dexing County (upstream), Leping County (midstream), and Poyang County (downstream) along the Le'an River were chosen for our study.

Soil samples of agricultural cropland were collected, of which 30, 13, and 10 sampling sites in the rural areas of Dexing, Leping, and Poyang Counties, respectively. Three soil samples in the city park were collected in each county as the typical urban soil sample. Each sampling site (approximate size 200 × 200 m) was further divided into a grid of cells using a systematic grid sampling method with regularly spaced intervals. Fifteen topsoil samples taken from depths of 0–20 cm were collected in each sampling zone using a random sampling method, and they were mixed thoroughly to form a composite sample. The soil samples were air-dried at room temperature and then pulverized using an agate mortar and sieved through a 0.15-mm polyamide sieve.

Six types of food samples were collected in the three counties in 2013 based on the dietary habits of local residents. These comprised vegetables (giant radish, brassica compestris L. var. purpurea Bailey, potato, green pepper, cucumber, eggplant, tomato, spinach, bok choy, small rape, and lactuca sativa L.), fish (grass carp, yellow-headed catfish, ricefield eel, crucian carp, and chub), meat (pork and mutton), rice, eggs (laid by local hens or purchased from the local market), and milk (liquid milk of six brands purchased locally). Eggs from local hens, homegrown vegetables, and rice were collected from farmers' homes, and the other food types were purchased from local markets, which were the main places where local residents obtained their food. For each food sub-type, five samples were collected and pooled into one bulk sample. For example, five carp were collected from the local markets and pooled as one representative carp sample. Vegetables, rice, eggs, and milk samples were stored and transferred at 4 °C, and meat and fish at −18 °C. Vegetables were washed with tap water to remove dust or soil and then further rinsed three times with deionized water, as in previous studies (Cao et al., 2014; Gebrekidan et al., 2013). The washed vegetables were subsequently dried at room temperature. The pretreatment of rice, fish, and meat was conducted according to previous studies (Gebrekidan et al., 2013; Shahbaz et al., 2013). All food samples were dried at 60–100 °C to achieve a constant weight and subsequently ground into powder prior to digestion.

2.2. Lead analysis

Approximately 0.1 g of each soil sample or 1 g of each food sample was taken from the composite sample, transferred to a Teflon® tube, and digested in a mixture of HNO_3 (65% GR, 5 mL), HF (48% GR, 2 mL), and $HClO_4$ (70% GR, 1 mL) in an automatic graphite digestion instrument (Model: ST60, Beijing Polytech Instruments, Ltd., China). After cooling the digestion mixture to room temperature, the mixture of digested soil was diluted to 25 mL and that of digested food to 10 mL with deionized water. The Pb concentration was quantified using inductively coupled plasma–atomic emission spectrometry (SPECTRO Analytical Instruments GmbH, Germany) for the soil samples, and inductively coupled plasma–mass spectrometry (NexION300x, PerkinElmer Instruments Co., Ltd, USA). Standard samples of pork (GBW08552), spinach [GBW10014(GSB-6)], and rice [GBW10014(GSB–1)] from the National Standard Material Center of China were used for quality control. Each sample was analyzed in triplicate. Three operation blanks were set for each batch of measurements. When the measured Pb concentration was lower than its corresponding operation blank, the Pb concentration was set to be half of the limit of detection (LOD). The LOD of Pb in the six food matrices and soil were $1.0–4.0 \times 10^{-3}$ μg g^{-1}. The average recovery of spiked Pb standards in the six food categories was 97.1 ± 7.5%.

2.3. Chronic dietary intake and risk assessment

The chronic daily intake (CDI, μg kg^{-1} d^{-1}) from foods and soil was used to indicate chronic exposure to Pb by reference to the US Environmental Protection Agency (USEPA) guidelines (USEPA, 1989), expressed as:

$$CDI = (C \times IR)/BW \tag{1}$$

where C (μg g^{-1}) is the Pb concentration in foods or soil, IR (g d^{-1}) is the ingested rate of the food and soil, and BW (kg) is body weight. The amounts of consumption of various food categories and body weights of the local population were obtained from the 2002 Chinese National Health and Nutrition Survey (CNNS) (Zhai and Yang, 2006). The soil ingestion amount was 50 mg d^{-1} recommended by US Environmental Protection Agency (USEPA, 2011).

The non-carcinogenic risk of Pb is typically characterized by the