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Exposure risk of local residents to copper near the largest flash copper smelter in China

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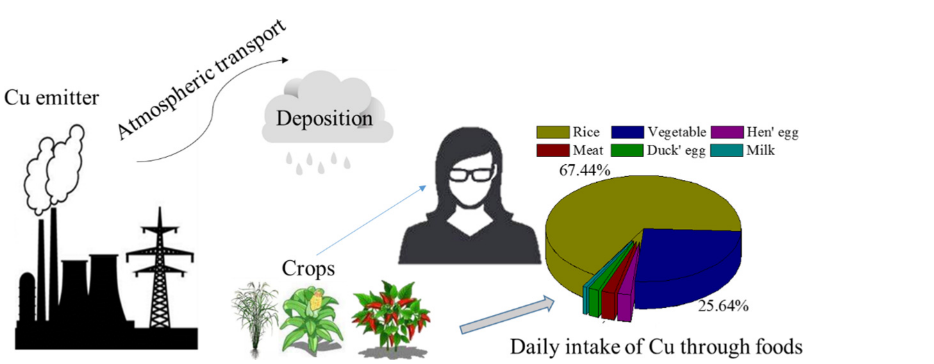
HIGHLIGHTS

- Copper health risks of food intake were studied in residents near a Cu smelter.
- Atmospheric deposition was an important source of Cu in local environment.
- Rice and vegetables were the two major pathways of the dietary intake of Cu.
- Hazard Quotients and bioindicators showed Cu exposure was highest to children.
- Copper smelter even pose a health risk to residents lived 7 km away.

GRAPHICAL ABSTRACT



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ABSTRACT

Copper (Cu) smelting released large amounts of Cu and contaminated the environment. However, few studies have investigated the Cu exposure risks for people located near Cu smelters. In this study, atmospheric bulk deposition, food from local families, drinking water and biological samples (hair and urine) were collected in three villages near the largest flash Cu smelter in China. The objective of the current study was to investigate how nonferrous metals smelting affect the human health. Total atmospheric Cu depositions (56–767 μg m^{-2} yr^{-1}) were one or two orders of magnitude greater than that of unpolluted rural areas. The Cu concentrations in locally grown vegetables and dietary chronic daily intake (CDI) of local residents showed a consistently decreasing trend with atmospheric Cu depositions. Dietary intake of vegetables and rice were the two major pathways of total CDI, which accounted for >93% totally. The Cu exposure showed higher potential non-carcinogenic risk to human health of local residents, especially children living around the Cu smelter through food consumptions. Health impact monitoring data revealed that mean Cu concentrations in hair and urine samples were ranged from 5.13 to 28.85 mg kg^{-1} and 19.90 to 54.61 μg L^{-1} in the three villages, respectively. Significant correlation between hair Cu concentrations and the CDI of Cu indicated food ingestion had adverse effects on the health of the local residents. The result suggested that nonferrous metal smelter should be away from residential area and locally produced crops became unsuitable for consumption. Therefore, effective measures on Cu pollution management and control in the surrounding area should be formulated and implemented.

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1. Introduction

Copper (Cu) is an essential trace element to biology, but large amounts can threaten human health and have adverse impacts on the growth of animals and plants (Bost et al., 2016; He et al., 2013; Khazaee et al., 2016). Therefore, Cu contaminations have been a growing concern for the environment in recent years. In general, mining and smelting are the two major anthropogenic sources of Cu pollution in the environment (Vorobeichik and Kaigorodova, 2017). Some other sources also contribute, such as using Cu-containing pesticide or fertilizer, automobile exhaust, coal combustion, sintering, metal processing, machinery manufacturing, iron and steel production (Vorobeichik and Kaigorodova, 2017; Wei and Yang, 2010). The emission of smelting dust is a major source of Cu pollution in the atmosphere. A large proportion of Cu in atmospheric particulates is deposited to the agriculture soils by wet and dry depositions, because smelters are usually located in the suburban and rural areas. The atmospheric deposition of the toxic metal has resulted in its accumulation in water, soil and crops, and then affects human health through food chains (Fernández-Olmo et al., 2014; Khazaee et al., 2016; Song et al., 2012; Vorobeichik and Kaigorodova, 2017; Zhou et al., 2016; Zhou et al., 2015b). However, due to lack of precise information of Cu status in the environment, it is usually difficult to estimate the impact on human health.

The most of the Cu in the human body is relevant to enzyme prosthetic groups or bound to proteins and Cu homeostasis is intimately regulated by a complex system of Cu transporters and chaperone proteins (Gaetke et al., 2014; Khazaee et al., 2016). Although the mechanisms of intestinal Cu absorption are not fully clear, it is absorbed mainly in the small intestine by a mechanism of free diffusion. After absorption, Cu is carried to the plasma and transported into the liver via the portal vein (Bost et al., 2016). The Cu concentrations in the whole blood, plasma, hair and urine are used as biological markers of Cu nutritional status of recent intake. Recent studies also suggested that Cu toxicity could induce changes in cellular activity, such as regulation of lipid metabolism, gene expression, neuronal activity, resistance of tumor cells to chemotherapeutic drugs, and temporal and spatial distribution of Cu in hepatocytes (Gaetke et al., 2014).

Increasing evidence suggests that Cu exposure may be more prevalent than previously thought in polluted areas and the occurrence of mild Cu deficiency or excess Cu exposure is not easily recognized (Gaetke et al., 2014). Due to lack of sensitive and specifically biological indicator, blood, urine, and hair analysis are used to detect Cu toxicity. The Cu in urine can reflect the short-term exposure, as the absorbed Cu is basically excreted in urine with several days in human bodies (Turnlund, 1983). Analysis of hair reveals a period beyond that offered by urine and blood and would provide a temporal reflection of Cu exposure, because hair grows at a rate of approximately 1 cm per month. Additionally, human hair incorporates xenobiotics from the human body, primarily the blood supply (Kempson et al., 2007). Therefore, the study of xenobiotics in hair and urine have promised to develop an alternative means by which to monitor nutrition, health status, and exposure to toxins and pollutants with respect to inorganic elements (Gaetke et al., 2014; Kempson et al., 2007; Ohashi et al., 2006; Pan et al., 2015).

As the economy grows rapidly, the demand of Cu has rapidly raised in China. The Guixi Copper Smelter is the largest flash Cu smelter in China, which produced 7.5×10^5 tons of Cu annually (Xiao et al., 2011) and resulted in serious Cu contamination in surrounding areas (Xu et al., 2017). After the ore was smelted, huge amounts of slag were stored in the open ground outside the plant and without any treatments. During the rainy season, the heavy rain washed off the stacked slag into the surrounding pond, stream and farmland (Jiang et al., 2010; Xiao et al., 2011; Xu et al., 2017). The heavy rain has also resulted in the overflow of acid sewage drainages, which flooded the surrounding farmlands and caused large amount of heavy metals accumulation in the surrounding agriculture soils. Additionally, the other two major pathways of atmospheric deposition and sewage irrigation have also caused serious Cu pollution in nearby farmlands, resulting in higher Cu concentrations in crops, livestock and poultry (Jiang et al., 2010; Xiao et al., 2011). Therefore, agricultural soil around the Cu smelter would act as large sinks of Cu pollution, and the Cu in the agricultural soils would be expected to be transferred to local residents, with particularly high risks to people residing around the smelter. It is urgently needed to assess the potential effects of the Cu smelting activities on local residents' health around the smelter.

In this study, the Cu concentrations in the bulk deposition, food (rice, vegetable, chicken's egg, duck's egg, meat and milk), drinking water, hair and urine were investigated near a largest flash Cu smelter. To the best of our knowledge, the current study is the first to investigate Cu pollution and potential health risk from Cu smelting areas in China. The primary goals of this paper are (1) to investigate the atmospheric Cu depositions; (2) to study Cu concentrations of various food types and potable water, and dietary intake of Cu from food and drinking water in local residents; (3) to assess the potential health risks associated with food consumption by hair and urine analysis among people resided in three villages near the Guixi Copper Smelter. We hypothesized that atmospheric deposition was one of the most important sources of Cu contamination in local environment and the exposure risk of Cu varied with population age groups.

2. Materials and methods

2.1. Study area

The Guixi Copper Smelter (28°19′30.1″ N, 117°14′28.9″ E) is the largest flash Cu smelter in China (Fig. 1). The smelter is located within 5 km north of the center of Guixi City and the smelter was set up in 1979. The climate in this area is mainly controlled by the subtropical monsoon, characterized by a wet and hot summer, and a dry and cool winter, with a mean annual temperature of 18.3 °C and rainfall of 1905 mm. The production capacity of the smelter is about 7.5 $\times 10^5$ tons of Cu in 2007. Some other accessory products were also produced simultaneously, such as 1.65×10^5 tons of H_2SO_4, 1.4×10^3 tons of As_2O_3, 1.3×10 tons of gold (Au), and 3.5×10^2 tons of silver (Ag) per year (Xiao et al., 2011). Due to the large-scale smelting, the local farmland was seriously contaminated by the irrigation of discharged wastewater, atmospheric deposition of flue gas and accumulation of waste residue. Over 130 hm^2 of farmland around the smelter was suffering from issues with heavy metal contaminations, resulting in heavy metal concentrations in crops exceeding the acceptable level (Xu et al., 2017). Three villages were selected to evaluate the Cu contamination and exposure to local residents, which was located to 1.0 km (Jiuniugang, JNG) and 7 km (Jiangnancun, JNC) to the west of the smelter and 1.5 km (Bingjiangxiaoqu, BJXQ) to the east of the smelter (Fig. 1).

2.2. Sampling

The total atmospheric Cu deposition, integrating both wet and dry depositions, was collected from the three study sites (Fig. 1). At each sampling site, the total atmospheric deposition was collected by a polyethylene terephthalate (PET) bottle (10 L in volume, 15 cm in diameter), which was placed on the rooftop of local house, which were about 5 m above the ground and 1.5 m above the rooftop to avoid the contamination of re-suspended soil particles. The samples were collected monthly and the sampling period was one year from August 2016 to July 2017.

Totally, 50 volunteers participated the sampling campaign in August 2017 living at the three villages and they were native-born with the age ranges of 5–81 years old. All the participants were local farmers and students. The age, sex, height, weight, resident time and diet of each participant were asked and recorded during the campaign. Hair samples were cut with stainless steel scissors from the occipital region of the scalp and