Science of the Total Environment 548–549 (2016) 402–407

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Exposure Risk of Rural Residents to Copper in the Le'an River Basin, Jiangxi Province, China

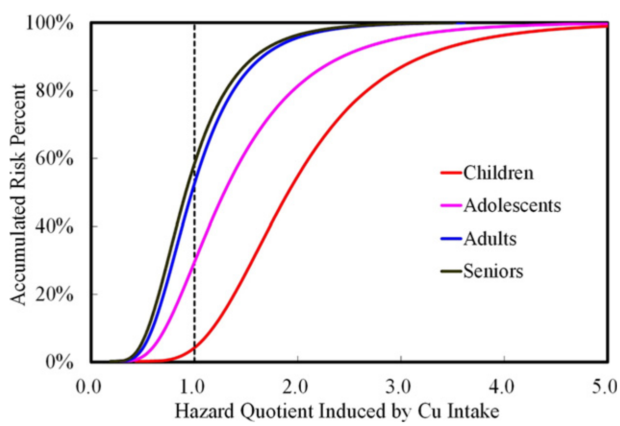
Yanxin Yu [a,*], Hui Wang [b], Qi Li [b], Bin Wang [c], Zhenghong Yan [b], Aizhong Ding [b]

[a] School of Environment, Beijing Normal University, Beijing 100875, P. R. China
[b] College of Water Science, Beijing Normal University, Beijing 100875, P. R. China
[c] Institute of Reproductive and Child Health/Ministry of Health Key Laboratory of Reproductive Health, School of Public Health, Peking University, Beijing 100191, P. R. China

HIGHLIGHTS

- Soils and food were studied in an area impacted by the largest copper mine in China.
- Cu content in various food types were affected by upstream Cu mining activities
- Cu intake decreased with increasing distance downstream from the Cu mining zone
- Vegetables and rice were the two major contributors to the total dietary intake of Cu
- Hazard Quotients indicated across the area >85% of children will be impacted

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 5 August 2015
Received in revised form 21 November 2015
Accepted 22 November 2015
Available online 21 January 2016

Editor: D. Barcelo

Keywords:
Mining
agricultural soil
food
chronic daily intake
CDI
exposure risk

ABSTRACT

The Dexing copper (Cu) mining zone in Jiangxi Province produces China's highest annual copper output, and the mineral waste residue and wastewater associated with ore processing are responsible for the Cu contamination of agricultural soil and food produced in the Le'an River Basin. We studied the dietary Cu intake from various foods, and the induced non-carcinogenic risk in rural residents from Dexing, Poyang, and Leping Counties situated along the Le'an River. Different food types based on the local dietary habits and agricultural soils were collected, and their Cu contents were analyzed. The Monte Carlo model was used to simulate the dietary chronic daily intake of Cu (CDI_{Cu}) and its non-carcinogenic risk in four subgroups (children, adolescents, adults, and seniors). A consistently decreasing trend in the Cu levels in agricultural soil and two local food types (vegetables and eggs) was found with increasing distance downstream from the mining zone from Dexing to Poyang, whereas this trend was not observed in other food types. The order of CDI_{Cu} among the three counties was Dexing > Leping > Poyang, and the order among the four subgroups was children > adolescents > adults = seniors. The two major contributors to the total CDI_{Cu} were vegetables and rice. Rural residents from Dexing County had the highest proportion of people with a hazard quotient (HQ) > 1 (i.e., 79%), followed by Leping (60%) and Poyang (48%). For Dexing, ~98% of the children living in rural areas displayed HQ > 1, compared with 97% in Leping and 85% in Poyang. Our results indicated the importance of the potential effects of Cu on the health of the local young population and the need to address such effects.

* Corresponding author.
E-mail address: yuhe_f@sina.com (Y. Yu).

http://dx.doi.org/10.1016/j.scitotenv.2015.11.107

1. Introduction

Mining activities and metal smelting are known to be the main sources of hazardous heavy metals transported to soils and rivers (El Hamiani et al., 2010; Rodriguez et al., 2009). Soil copper (Cu) content has been found to be higher than the global average level (30 mg kg^{-1}) near mining zones and smelting factories around the world (Cheng et al., 2011; Wcislo et al., 2002; Lim et al., 2008; Cao et al., 2014). Over the past several decades, the dramatic social and economic development taking place in China has stimulated a rapid rise in demand for Cu. Dexing Copper Mine in Jiangxi Province has China's highest annual copper ore production in Asia (Zhu et al., 2007)· of which the Fujiawu copper mine is the biggest with ~10,000 tons ore production per day. Cu in Dexing has been intensively exploited, and higher pollution around the mining zone has been reported in the previous studies (Zhu et al., 2007; Wang et al., 2008; Liu et al., 2013). The Le'an River, the primary river of the Le'an River Basin, flows through Dexing County to Poyang Lake, the largest freshwater lake in China. Mineral waste residue, wastewater, and dust containing heavy metals are discharged into the environment during the processes of mining and in mine tailings. Flooding, aerial deposition, and agricultural irrigation can additionally cause serious Cu contamination in nearby agricultural soil (Nan et al., 2002), resulting in elevated Cu levels in crops, livestock, and poultry (Gupta and Gupta, 1998). If agricultural soil, as an important sink of heavy metal pollution, is contaminated by Cu along the Le'an River downstream from the Dexing mining zone, it would be expected to be transferred to humans, with a particularly high risk for rural residents living along the Le'an River (Cheng et al., 2011; Teng et al., 2010). Therefore, it is important to investigate the Cu concentration of agricultural soil along the Le'an River downstream from the mining zone to assess the potential health effects of the Cu mining activities.

Three routes of exposure have been widely studied to assess the heavy metal exposure in humans: diet, inhalation, and skin contact (US Environmental Protection Agency, 1997). Results from previous studies suggest that diet was the major exposure route of heavy metals (Cao et al., 2014; Zheng et al., 2007). For example, the estimated risks induced by most heavy metals were mainly from the ingestion of local food, accounting for more than 85% of the three routes, and 98% for copper. Dietary habits, food sources, and the Cu concentrations of different food types can have a considerable influence on dietary Cu intake, and this intake can vary significantly with age group (Zhu et al., 2007). Hence, the selection of which foods to sample should be based on the dietary habits and food sources of the population in question. The spatial variance in the Cu concentrations of agricultural soil along the Le'an River can give rise to different contamination patterns of Cu in agricultural crops and foods. To control the levels of Cu exposure in the local population, knowledge of the relative contributions of different exposure sources to specific age groups is required.

Cu is an essential nutrient and a redox-active transition metal that can lead to oxidative stress damage in human body. Excessive intake of Cu can result in weakness, lethargy, and anorexia in the early stages (Winge and Mehra, 1990), as well as the development of acute gastrointestinal symptoms, hepatocellular necrosis in the liver, and acute tubular necrosis in the kidney (Barceloux, 1999). As Cu is a non-carcinogenic heavy metal, its non-carcinogenic risk is typically characterized by the hazard quotient (HQ), taking into consideration the amount of food consumption, the Cu content in various food types, body weight, and the reference exposure dose for humans (US Environmental Protection Agency, 1997). To include all of the influencing factors, the Monte Carlo method is usually used to simulate exposure from the multiple sources and provide a distribution of the exposure risk of the population (Cao et al., 2014).

The staple foods (e.g., vegetables, rice, and eggs) of rural residents living along the Le'an River downstream from the Dexing copper mining zone are produced mainly on local farmland. We measured the Cu contents of agricultural soil and various food types in rural areas along the Le'an River. The aims of this study were to investigate the following: 1) the Cu levels of agricultural soil and various food types in rural areas along the Le'an River downstream from the Dexing mining zone; 2) the chronic dietary intake of Cu from various food types in local residents living along the Le'an River and their relative contributions to their total Cu intake; and 3) the potential non-carcinogenic risk of Cu for these residents. Our hypothesis was that the dietary exposure risk of Cu for the rural residents along the Le'an River would decrease from the upstream mining zone to downstream and vary with population age groups.

2. Materials and Method

2.1. Study area and sampling

The location of the Le'an River and the main mining and smelter area in the Le'an River Basin in Jiangxi Province, China, are shown in **Figure S1** (Supporting Information)**.** The rural areas of Dexing County (upstream), Leping County (midstream), and Poyang County (downstream) along the Le'an River were chosen for our study.

Agricultural soil samples were collected from rural areas in the three counties, with 30 sampling zones in Dexing County, 13 in Leping County, and 10 in Poyang County. Each sampling zone (approximate size 200 × 200 m) was further divided into a grid of cells using a systematic grid sampling method with regularly spaced intervals (~40 m). Fifteen topsoil samples taken from depths of 0–20 cm were collected in each sampling zone using a random sampling method, and the 15 samples 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 rural villages of the three counties in 2013 based on the dietary habits of local residents. These comprised vegetables (giant radish, *brassica compestris L.* var. *purpurea Baileysh*, 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 as shown in Figure S1. 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). 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; Yılmaz et al., 2007). Briefly, rice samples were washed by tap water three times and further by ultrapure water three times, oven-dried to constant weight and ground with a stainless steel grinder to pass through a 100-mesh sieve. Muscle (edible parts) of fish were dissected using a stainless steel scissor. The tissue of fish and meat were frozen-dried at ~80°C to constant weight and ground into powder for further digestion. All food samples were dried at 60–100°C to achieve a constant weight and subsequently ground into powder prior to digestion.

2.2. Copper 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