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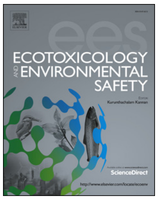

Multivariate linear regression model for source apportionment and health risk assessment of heavy metals from different environmental media

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ABSTRACT

The study evaluated source apportionment of heavy metals in vegetable samples from the potential sources of fertilizer, water and soil samples collected along the Changjiang River delta in China. The results showed that 25.72% of vegetable samples (*Brassica chinensis* L.) containing Pb, and Cd, Cu, Hg and Zn at relatively serious levels were from soil. Combined with principle component analysis (PCA) and cluster analysis (CA), the results of the spatial distribution of heavy metals in different environmental media indicated that fertilizer, water and soil were the main sources of heavy metals in vegetables. The results of multivariate linear regression (MLR) using partition indexes (P) showed that fertilizer contributed to 38.5%, 40.56%, 46.01%, 53.34% and 65.25% of As, Cd, Cu, Pb and Zn contents in vegetables, respectively. In contrast, 44.58% of As, 32.57% of Hg and 32.83% of Pb in vegetables came from soil and 42.78% of Cd and 66.97% of Hg contents in vegetables came from the irrigation water. The results of PCA and CA verified that MLR using P was suitable for determining source apportionment in a vegetable. A health risk assessment was performed; As, Cd and Pb contributed to more than 75% of the total hazard quotient (THQ) values and total carcinogenic risk values ($Risk_{total}$) for adults and children through oral ingestion. More than 70% of the estimated THQ and $Risk_{total}$ is contributed by water and fertilizer. Therefore, it is necessary to increase efforts in screening limits/levels of heavy metals in fertilizer and irrigation water and prioritize appropriate pollution management strategies.

1. Introduction

Heavy metal pollution has increased dramatically all over the world since the 1950s (Nriagu, 1996). Consequently, heavy metal pollution in plants and soils has become an important issue from the viewpoint of human health and well-being (Li et al., 2012, 2006; Zhuang et al., 2009). According to previous reports by International Agency for Research on Cancer (IARC), As has adverse effects on skin and respiratory and cardiovascular systems (IARC, 2004), while Cd, Pb and Cr can cause nervous system disorder, renal failure and increased carcinogenic risk (Bandara et al., 2008; IARC, 2004; Yang et al., 2004; Zhang et al., 2015). Besides soil, the largest reservoir of heavy metals (Aubert and Pinta, 1977; Z. Li et al., 2014), water and the atmosphere are fundamental parts of the heavy metal cycle in the natural environment (Fu et al., 2012; J. Li et al., 2014; MacKay et al., 2013). Heavy metals are present in various forms in the environment, where they are continuously migrating, transforming and increasing in the ecosystem. Direct human exposure to heavy metals through inhalation, dermal absorption and ingestion ultimately threatens human health (US EPA, 2011; Zhuang et al., 2009).

Abbreviations: PCA, Principle component analysis; CA, Cluster analysis; MLR, Multivariate linear regression; P, Partition index; THQ, Total hazard quotient; HQ, Hazard quotient; $Risk_i$, Carcinogenic risk posed by single heavy metal; $Risk_{Total}$, Total carcinogenic risk posed by all heavy metals of interest; ADI, Average daily intake; PC, Principal component

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Heavy metal pollution in soils poses potential threats and risk to human health through the ingestion of agriculture products grown in polluted soils. Agricultural products contaminated by heavy metals have become the most pressing global concern (Tóth et al., 2016). Much research has been done on rice and wheat, the most important staple crops worldwide (Adhikari and Rattan, 2000; Orun et al., 2008; Sayyad et al., 2010; Watanabe et al., 1996; Williams et al., 2007). Vegetables also play an important role in our daily diet, according to data from the World Bank (http://datatopics.worldbank.org/consumption/). However, besides cereal products, daily intake of vegetables contributes a lot of heavy metals to the human diet (Zheng et al., 2007a). Long-term exposure to heavy metals through food ingestion can cause acute and chronic adverse effects on human health. For example, exposure to As can cause cancer and skin disorders (Booth, 2009; Rahman et al., 2009). Previous researchers found that As and Cr potentially cause cancer through different pathways (Kim et al., 2015; Park et al., 2004). Besides cancer, arsenic is considered the most toxic heavy metal to human health worldwide, causing adverse health effects that may include dermal, respiratory, cardiovascular, gastrointestinal, renal, neurological, developmental, reproductive and immunological diseases (Booth, 2009; Kapaj et al., 2006). It was reported that Cd, Pb and Cr are also classified as elements which are probably carcinogenic to humans (IARC, 2017; Zhang et al., 2015).

Due to rapid industrialization, the use of chemical fertilizers, pesticides and sewage irrigation in agricultural production has increased, while the unreasonable disposal of domestic waste and the exploitation of nearby metal mines caused the accumulation of heavy metals in the vegetables (Cao et al., 2010; Sterrett et al., 1996; Zheng et al., 2007b; Zhuang et al., 2009). In developing countries, heavy metal pollution in agriculture is gradually increasing. In China, in particular, about 19.4% of arable land is polluted according to the report on a national general survey on soil contamination by the Ministry of Ecology and Environment (MEP) and Ministry of Natural Resources, PRC (MEP and MNR, 2014). However, in addition to soil, irrigation water, fertilizers and atmospheric deposition are potential sources of heavy metals in vegetables (He et al., 2004; Li et al., 2012; Liu et al., 2005; Mortvedt and Beaton, 1995; Sterrett et al., 1996; Wang et al., 2005). The transfer pathway of heavy metals from environmental media to vegetables is the most important link in pollution prevention and control. In the vicinity of industrial or e-waste processing sites, crop exposure to heavy metals could increase (Luo et al., 2011; Zheng et al., 2007a), due to soil contamination and, to some extent, atmospheric pollution (Feng et al., 2004; Li et al., 2012).

Various methods have been used to analyze heavy metal pollution. Contour maps can highlight hotspots of heavy metal pollution. Spatial distribution analysis can be used to determine locations of heavy metals contamination with high health risk values (Tóth et al., 2016). Application of isotopes can quantify levels of heavy metals. Principal component analysis (PCA) methods can be used to identify the pollution sources and characterize the transfer of heavy metals between different media through large-scale survey data (Dvonch et al., 1999; Song et al., 2011). However, isotopes methods are expensive and cumbersome due to stringent analysis conditions such as requiring high-quality sampling of media to avoid cross contamination between samples. The extraction of principal components is sometimes unsatisfactory when too many potential explanations are procured and thus limits the explanatory power of PCA. Moreover, PCA cannot provide quantitative information.

To further our understanding and quantitatively evaluate the transfer of heavy metals between multiple media, we collected samples of soil, irrigation water from the river, fertilizers and vegetables in southern Jiangsu, where intensive farming and industrial activities coexist in China. Amounts of heavy metals were partitioned among the four different media to characterize the transfer of heavy metals. In addition, a model established by multivariate linear regression (MLR) was used to evaluate the transfer of heavy metals from soil, irrigation water and fertilizer into vegetables. The relative contributions of heavy metals in vegetable samples, transferred from soil, irrigation water and fertilizer, were calculated and evaluated. Likewise, the contributions of soil, irrigation water and fertilizer to health risk of humans through consumption of heavy metal-contaminated vegetables was assessed.

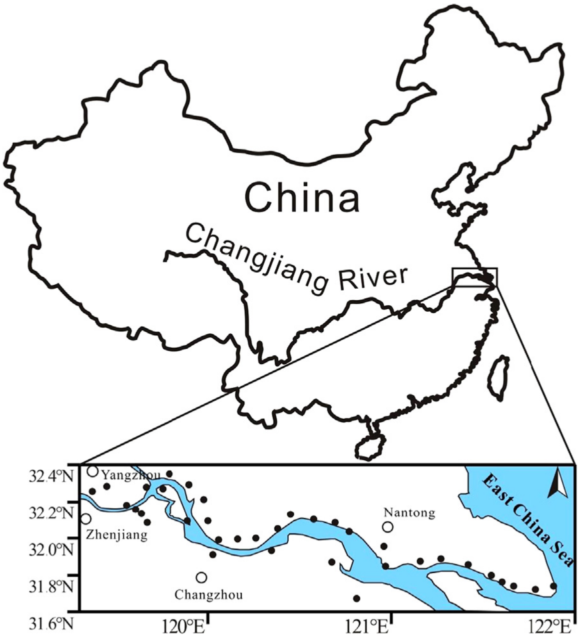

Fig. 1. Sampling locations distributed along the Changjiang River.

2. Materials and methods

2.1. Study area and sampling

As a part of a program of the Geochemical Survey in Changjiang River Basin, vegetable (*Brassica Chinensis* L.), irrigation water, fertilizer and soil samples were collected from 35 sampling sites in southern Jiangsu Province, China (31°30′–32°25′N; 119°00′–122°00′E, Fig. 1). All sampling sites were distributed at approximately 15 km intervals along the Changjiang River. Paired samples were selected in sampling sites at 15 km intervals. Each site was a 100 × 100 m area where four subsamples were collected randomly and mixed to obtain bulk samples of vegetable, water and soil. The vegetable samples were collected and sealed in plastic bags and taken to the laboratory. There, they were twice cleaned with distilled water to remove dust and impurities and cut into small pieces after removing the unusable parts. The samples were dried in an oven at 40 °C to a constant weight. The dried samples were ashed in a muffle furnace at 350–400 °C. For the water samples, dilute nitric acid-washed samplers/buckets were used to collect samples from small tributaries of the Changjiang River because they were the direct sources of irrigation water. The sampler was washed for 3–5 times with the respective irrigation water of each site before each sample collection. After filtration, the collected irrigation water samples were stored in acid-cleaned plastic bottles and taken to the laboratory and stored at 4 °C. In order to avoid metal impurities, plastic shovels were used to scrape the surface of the soil and dig through the soil to collect samples from a depth of 5–10 cm. Four soil samples were collected from each site and mixed into one representative sample. The collected soil samples were stored in plastic bags and taken to the laboratory to store at room temperature (~ 25 °C).

The fertilizer samples (urea is the primary fertilizer used in this area) were obtained from local farmers. The samples were subsequently dried and ashed before further chemical analysis.