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Linkage between human population and trace elements in soils of the Pearl River Delta: Implications for source identification and risk assessment

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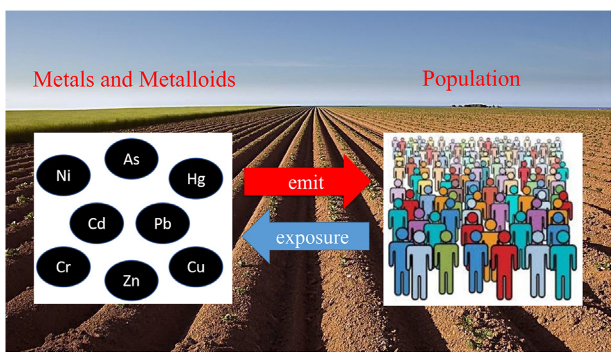
HIGHLIGHTS

- Population's relationship with metals and their sources in PRD was investigated.
- Adults showed stronger correlations with industrial metals than other age groups.
- Only seniors were significantly correlated with agricultural and natural metals.
- Population-weighted exposure to metals is higher than directly-calculated in PRD.

GRAPHICAL ABSTRACT



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ABSTRACT

The human population is both an emitter and receptor of metals. This study aims to clarify how the relationship of metals and metalloids to human populations influences their source characterization and health risk, based on metal concentrations in 298 soil samples in the Pearl River Delta (PRD) and the corresponding zip-code level population. Nickel (Ni), copper (Cu), zinc (Zn), cadmium (Cd), mercury (Hg), and lead (Pb), but not chromium (Cr) and arsenic (As), were significantly correlated with population ($p < 0.01$), suggesting potential anthropogenic sources. A principal component analysis (PCA) revealed three factors (i.e., F1, F2, and F3) contributing to metal levels in the PRD: (1) metal transport from rivers (F1), which explained the high levels of Cr, Ni, Cu, Zn, As, and Cd in downstream areas; (2) industrial sources (F2), mainly contributing to Ni, Cu, Zn, Cd, Hg, and Pb; and (3) natural and agricultural sources (F3), mainly contributing to As and Pb. F2 was significantly correlated with population, while F3 was not, indicating that an analysis of the correlation with population could be used to identify industrial sources of metals. Compared with directly calculated risks, the population-weighted non-carcinogenic and carcinogenic risks were increased by 4.2–4.9% and 7.7–9.2%, respectively. A unit increase in the concentration of industrial metals led to higher extra risks than a corresponding increase in natural metals due to the proximity to human populations.

1. Introduction

Soils interface with the lithosphere, hydrosphere, biosphere, and atmosphere and are critical for the viability of its inhabitants. In recent

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years, soils in China have been severely contaminated with metals and metalloids such as chromium (Cr), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) (Chen et al., 2014, 2015). Metals and metalloids originate from both natural processes and anthropogenic activities (Wuana and Okieimen, 2011). However, anthropogenic sources, such as metal mining, industrial waste, and sewage irrigation, have been proven to be the major causes of soil pollution.

Human exposure to metals in soils occurs either through food chains or direct intake (i.e., oral ingestion, dermal contact, and inhalation) (Watt et al., 1993; Tóth et al., 2016). There are substantial evidences that the levels of metals in human blood are closely related to their levels in soils, especially among children (Watt et al., 1993; Davies et al., 1990). Although some metals (e.g., Cr, Cu, and Zn) are essential nutrients, an excessive intake of some metals will lead to a variety of adverse effects and diseases, including cancers (Tchounwou et al., 2012). As one of the mechanisms of metal toxicity, metals can bond to proteins and disturb their normal functioning (Tchounwou et al., 2012). In this regard, children and seniors are more vulnerable to metal exposure because they are still developing or experiencing physiological decline, respectively, and are therefore less resistant to disturbance.

The Pearl River Delta (PRD) is one of the most developed regions in China. It holds 4% of the total population and contributes to >10% of the national gross domestic product (GDP), but only occupies <0.5% of the Chinese national area (GDPBS, 2014). Unfortunately, the PRD is also one of the most polluted regions of the country, with elevated levels of soil contamination. Several studies have investigated the level and health risk of metals and metalloids in soils in the PRD from the perspective of administrative districts and landscape variables (Hu et al., 2013; Liu et al., 2016; Wu et al., 2015; Zhang et al., 2015; Li et al., 2015). However, the relationship between population and soil metals in this region is unclear, but might be important. On the one hand, the relationship could reveal the impact of anthropogenic activities on regional metal pollution. On the other hand, metals and metalloids may pose more of a threat if their spatial distribution overlaps with the population. In this study, we measured the concentration of eight metals or metalloids in 298 soil samples collected in the PRD and obtained the zip-code level population data of each site from the 6th National Population Census (NSB, 2012). The aim of the study was: (1) to characterize the levels and sources of metals in the soils of the PRD and investigate their relationship with the population; and (2) quantify the additional risk caused by the overlap between population and metals in soils.

2. Methods

2.1. Sample collection

From August 2011 to March 2012, a total of 298 surface (0–20 cm) agricultural soil samples were collected from across the PRD to represent the whole area (Fig. 1). At each sampling site, five subsamples were taken from the same area (approximately 100 m^2) and pooled together to form one composite sample.

All samples were air-dried for one week at room temperature in a storage room; sieved into 250 μm mesh size particles after removing stones, residual roots, and other unwanted materials; and then sealed in brown glass bottles and conserved in a refrigerator at −4 °C until analysis. Samples were analyzed following the United States Environmental Protection Agency (US EPA) methods 3050B and 6010C (USEPA, 1996a, 1996b). In brief, approximately 10 g of sample was air-dried, gently ground with an agate pestle, and passed through a 100-mesh nylon sieve. One gram of sample was placed into Anton PVC digestion vessels along with 9 mL concentrated nitric acid (HNO_3) and 3 mL hydrogen peroxide (H_2O_2), then sealed and heated at 180 °C for 15 min. The concentrations of metals, Cr, Ni, Cu, Zn, As, Cd, Hg, and Pb, in the digested solution were determined using inductively coupled plasma mass spectrometry (ICP-MS, POEMS3, Thermo Fisher Scientific, Waltham, MA, USA).

2.2. Quality assurance and quality control

Soil standard reference material (GBW, GSS21, 22, 25) was used for quality assurance and quality control. Accepted recoveries ranged from 83% to 112%. Analysis methods were evaluated in blank (n = 30) and duplicate samples (n = 30) for each set of samples. The relative deviation of the duplicate samples was <7% for all batch treatments. The data mention above was listed in Table S1 and Table S2 of the Supplementary material.

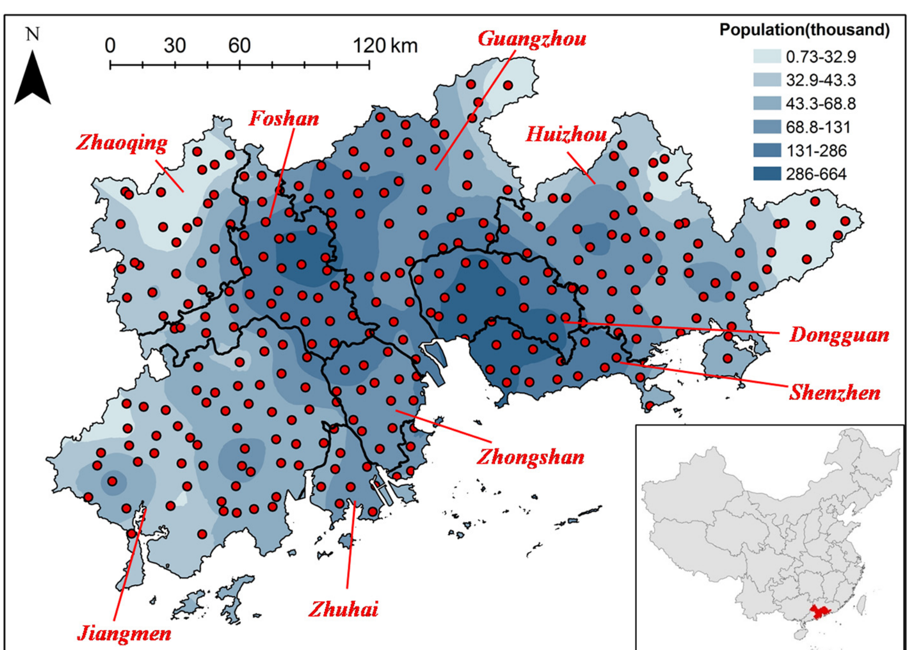

Fig. 1. Map of sampling sites and the spatial distribution of population in PRD.