



Heavy metal contents, distribution, and prediction in a regional soil–wheat system



Jing Ran^{a,b}, Dejian Wang^{a,*}, Can Wang^a, Gang Zhang^a, Hailin Zhang^c

^a Institute of Soil Science, Chinese Academy of Sciences, Nanjing 210008, China

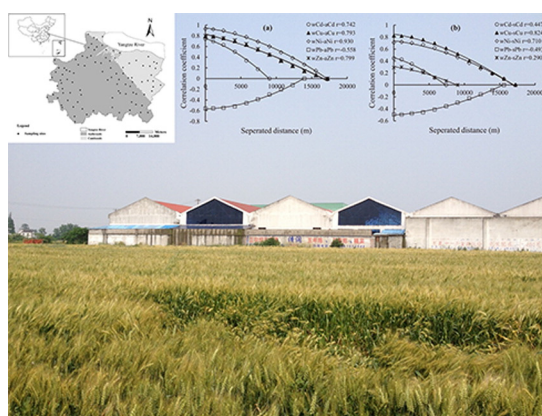
^b University of Chinese Academy of Sciences, Beijing 100049, China

^c Department of Plant and Soil Sciences, Oklahoma State University, Stillwater, OK 74078, USA

HIGHLIGHTS

- Accumulation of heavy metals was found in soils and wheat grains, especially Cd.
- Cd, Cu, Ni, and Zn had similar distribution patterns, while Pb had opposite trends.
- Heavy metals in wheat grains were spatially correlated with most soil properties.
- pH was varied from 5.0 to 8.6, and was negatively correlated with wheat grain Cd.
- Models for wheat Cd were established with a maximum predictive success of 65%.

GRAPHICAL ABSTRACT



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ABSTRACT

The entry of heavy metals into the food chain is of concern for potential health risks. To investigate the spatial relationships of heavy metals in a regional soil–wheat system, 99 pairs of surface soil (0–15 cm) and wheat grain samples were collected from Changshu, China, a typical county in the Yangtze Delta region. Both soil and wheat grain samples were analyzed for total Cd, Cu, Ni, Pb, and Zn. DTPA-extractable metals and major physico-chemical properties were also determined for soil samples. Moderate accumulation of heavy metals was found in soils and wheat grains, especially Cd. However, the levels were within the target hazard quotients (THQ) safe values with respect to non-carcinogenic risks, but more attention should be paid to Cd. Spatially, Cd, Cu, Ni, and Zn in wheat grains and soils had similar geographical patterns, whereas Pb showed opposite trends. Cross-correlograms further quantitatively confirmed the spatial relationships of heavy metals in wheat grains and soils. In addition, heavy metals in wheat grains were significantly spatially correlated with most soil physico-chemical properties. Particularly, a set of regression models for Cd in wheat grains were established with a maximum predictive success of 65%. These models can be used to predict Cd in wheat grains, and thus allows farmers to decrease the threat by certain farming practices such as ameliorating soil pH or growing a less metal-accumulating cultivar.

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* Corresponding author.

E-mail address: djwang@issas.ac.cn (D. Wang).

1. Introduction

Accumulation and contamination of heavy metals in agricultural soils have aroused a worldwide attention for their toxicity, non-biodegradability, and persistence (Alloway, 2012). In China, heavy metals in contaminated soils are considered to pose the greatest health risk to food safety and human health (Chen et al., 2015). Additionally, many contaminated agricultural lands are still being used for producing foodstuff to meet the increased demand for food supply (Yu et al., 2006). Apart from naturally deriving from soil parent materials, heavy metals in farmland soils are mainly associated with a variety of anthropogenic activities (including mining, smelting, sewage irrigation, sludge reuse, and agrochemical application) (Alloway, 2012; Luo et al., 2009; Wei and Yang, 2010).

Wheat is an important staple agricultural crop worldwide. Like most plants, wheat is at the beginning of the food chain and absorbs essential and non-essential elements from soils, which has raised substantial concerns about the possibility of toxic elements being transported to the higher food chain strata, especially human beings (Peralta-Videa et al., 2009). Previous studies have investigated the accumulation and transfer of heavy metals in the soil–wheat system (Adams et al., 2004; Baize et al., 2009; Huang et al., 2008; Oliver et al., 1994; Shi et al., 2015; Wang et al., 2012; Wu et al., 2002). Usually, the concentration of heavy metals in wheat grains is affected by soil conditions (Adams et al., 2004; Baize et al., 2009), cultivars (Shi et al., 2015), and geographic locations (Wu et al., 2002). Among the soil characteristics, soil heavy metal contents, pH, organic matter, and texture are commonly reported to influence the uptake of heavy metals by wheat. As regarding to different metals, Cd, a non-essential element, is readily taken up by crops and can be transferred to edible parts where it can be accumulated to a relatively high levels endangering human health without causing severe phyto-toxicity symptoms (Li et al., 2011; Palmgren et al., 2008). However, most of these previous studies were conducted by pot or field plot experiments, and were focused on the areas with special interest and associated risk assessment. Relatively little information is known about the spatial variability and spatial relationship of heavy metals between soil and crops at regional scales. Such information has important implications for developing and applying agronomic practices to lower heavy metals in wheat and other harvested crops.

Due to the impact of long-term, low-level mixed metals exposure, the relationships of heavy metals between soil and plant are more complex under regional field conditions. The combination of geostatistics and multivariate analysis is an effective technique to explore such relationships (Facchinelli et al., 2001; Ran et al., 2014; Zhang, 2006). Geostatistical methods have been widely used to study the spatial variability of soil and plant properties. Geo-chemical mapping enables the geostatistics information to be visualized and provides a reliable approach for identifying problem areas (Li et al., 2004). Ordinary kriging (OK) can provide the best linear unbiased estimate for spatial variables relative to other interpolation methods (Hang et al., 2009; Zhao et al., 2010). Cross-correlogram allows one to evaluate the strength and the direction of the relationship between spatial variables (Kravchenko et al., 2003). Principal components analysis (PCA) is widely used in environmental and ecological studies. PCA can handle a large set of biotic and abiotic data by reducing multi-dimensional data sets to lower dimensions and further enable one to extract interpretable information to explain physico-chemical parameters of a system (Çamdevýren et al., 2005).

In this study, we measured heavy metals (Cd, Cu, Ni, Pb, and Zn) in surface soils and wheat grains as well as associated soil properties under actual field conditions in Changshu County, one of the most rapidly developing areas in the Yangtze Delta, China. The specific objectives were to: (1) assess the contamination of heavy metals in the soil and wheat grain and related health risk; (2) investigate the spatial relationships between heavy metals in the soil and wheat grain, and the effects of soil properties; and (3) predict the transfer of Cd from soil to wheat grain with a set of models.

2. Materials and methods

2.1. Study area and sampling

This study was carried out in Changshu (31°31′–31°50′ N, 120°33′–121°03′ E) (Fig. 1), a typical county which is located in the central part of the Yangtze Delta region, China and has experienced a rapid urbanization and industrialization process since the early 1980s. It covers an area of 1276 km² and enjoys a warm and humid subtropical climate. Anthrosols (Inceptisols) and Cambosols (Entisols) are the major soil orders therein (Fig. 1), based on the Chinese Soil Taxonomy (CRG, 2001). Traditionally, this area is under a wheat-rice rotation.

Just before the wheat harvest season (May 2014), 99 pairs of surface soils (0–15 cm) and grains of winter wheat (*Triticum aestivum* L.) were collected (Fig. 1). Considering the soil type and terrain, the sampling sites were located in arable land by a GPS with an interval of approximately 4 km. Each sample was comprised of four to six sub-samples located within approximately 300 m². Samples were kept in plastic bags and brought back to laboratory.

Soil samples were air-dried and sieved to pass through a 2 mm mesh for the determination of particle size distribution, pH, and DTPA-extractable metals. Then, a portion of each sample (approximately 50 g) was ground to pass through a 0.25 mm mesh for the determination of cation exchange capacity (CEC) and a 0.149 mm mesh for the determination of total heavy metals, organic matter (OM), nitrogen (N), potassium (K), phosphorus (P), and calcium (Ca). All soil samples were stored in polyethylene bottles for later chemical analysis. Wheat grains were thoroughly washed with tap water, rinsed with deionized water, and oven-dried at 70 °C to constant weight. The hulls were removed by hand. Then, the wheat grain samples were ground to pass through a 0.149 mm mesh and stored in sealed polyethylene bags for heavy metal analysis.

2.2. Chemical analysis

Total concentrations of nine elements in the soils (Cd, Cu, Fe, K, Mn, Ni, P, Pb, and Zn) were obtained after digestion by a mixture of concentrated HNO₃, HClO₄, and HF (1:1:2, v/v/v) and analyzed by an inductively coupled plasma atomic emission spectrometry (ICP-AES) or inductively coupled plasma mass spectrometry (ICP-MS), depending on the element (Lu, 2000; Wang et al., 2014). The available heavy metals (Cd, Cu, Fe, Mn, Ni, Pb, and Zn) in the soil samples were extracted using the diethylenetriamine penta-acetic acid (DTPA) method and were quantified by an ICP-AES or ICP-MS (Lindsay and Norvell, 1978). The Clay content (<0.002 mm) of soil samples was determined using the hydrometer method (Kettler et al., 2001). The total concentrations of Ca in soil samples were determined by powder X-ray fluorescence (XRF) (Kilbride et al., 2006). Other soil properties (pH, OM, CEC, and N) were analyzed based on the methods of Lu (2000). Briefly, soil pH (soil:water = 1:2.5) was determined using an ion selective electrode. OM was measured by wet oxidation using K₂Cr₂O₇. CEC was determined with the ammonium acetate method. Total N was determined by the Kjeldahl method after digestion with H₂SO₄.

Powered wheat grain samples were digested by a mixture of HNO₃ and HClO₄ (3:1, v/v) (Hang et al., 2009). Then, concentrations of Cd, Cu, Fe, K, Mn, Ni, P, Pb, and Zn were determined by an ICP-AES or ICP-MS.

All glassware was soaked in nitric acid solution for more than 24 h and rinsed with deionized water. Blank samples, duplicate samples, and standard reference materials (SRMs) (GSS4, GSS5, and GBW07414a for soil, GBW10011 for wheat) were used during the analytical process for quality control. For ICP-MS analyses, Rh (10 µg L⁻¹) was used as internal standard to correct for instrumental drift and matrix effects. The recoveries of SRMs ranged from 86.5% to 111.6%, and the relative standard deviation was less than 10%.

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