



## Potentially harmful elements in rice paddy fields in mercury hot spots in Guizhou, China

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### ABSTRACT

Levels of the chalcophile metals Hg, Pb, Cd, Cu and Zn and the metalloid As in soils from rice paddy fields were assessed in two regions impacted by different industrial activities in Guizhou province, China. The two study areas (Wanshan and Qingzhen) have previously received attention in the international literature due to heavy Hg pollution, though levels of other potentially harmful elements have not previously been reported. The regions were selected as representing two important, but categorically different metal(loid) contaminated areas in China: Wanshan has been a major region for Hg production while Qingzhen is an important region for a large range of various heavy industries based on coal as an energy source.

Within a limited distance of the Hg mine tailings in Wanshan the rice paddy fields are heavily contaminated by Hg (median concentration  $25 \mu\text{g g}^{-1}$ , maximum  $119 \mu\text{g g}^{-1}$ ) and moderately contaminated by Zn and Cd (median concentrations of  $86$  and  $0.9 \mu\text{g g}^{-1}$ , respectively). Zinc and Cd levels correlate well with Hg contamination, which indicates a similar source and mechanism of transport and accumulation. Other studies have concluded that the main sources of Hg contamination in Wanshan are the numerous Hg mine tailings. This is likely as these metals are all geochemically associated with the mineral cinnabar (HgS). The other chalcophile elements (Pb, Cu and As) are nevertheless found at background levels only. In Qingzhen the soils were found to contain elevated levels of As and Hg (median concentrations of  $38$  and  $0.3 \mu\text{g g}^{-1}$ , respectively). These are elements that are known to be more associated with coal and released during coal combustion.

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### 1. Introduction

Potentially harmful elements (PHE) are ubiquitous and are natural constituents of rock and bedrock and are generally rather immobile in soils. For this reason, soils have natural background levels of these elements as a result of rock weathering. In addition to this, anthropogenic releases contribute to an increase in PHE concentrations in the environment (Alloway, 1995). Typically, the contaminants are released to air and water, and accumulate in the soil as a consequence of the strong affinity of PHE for humic matter and clay colloids (Alloway and Ayres, 1997). Due to the anthropogenic emissions the human exposure to elevated levels of PHE continues, despite well known adverse health effects, and in some emerging economies the level of exposure is increasing (Järup, 2003).

China's rapidly developing industry and energy sector has led to an increase in emissions of pollutants, including releases of a wide

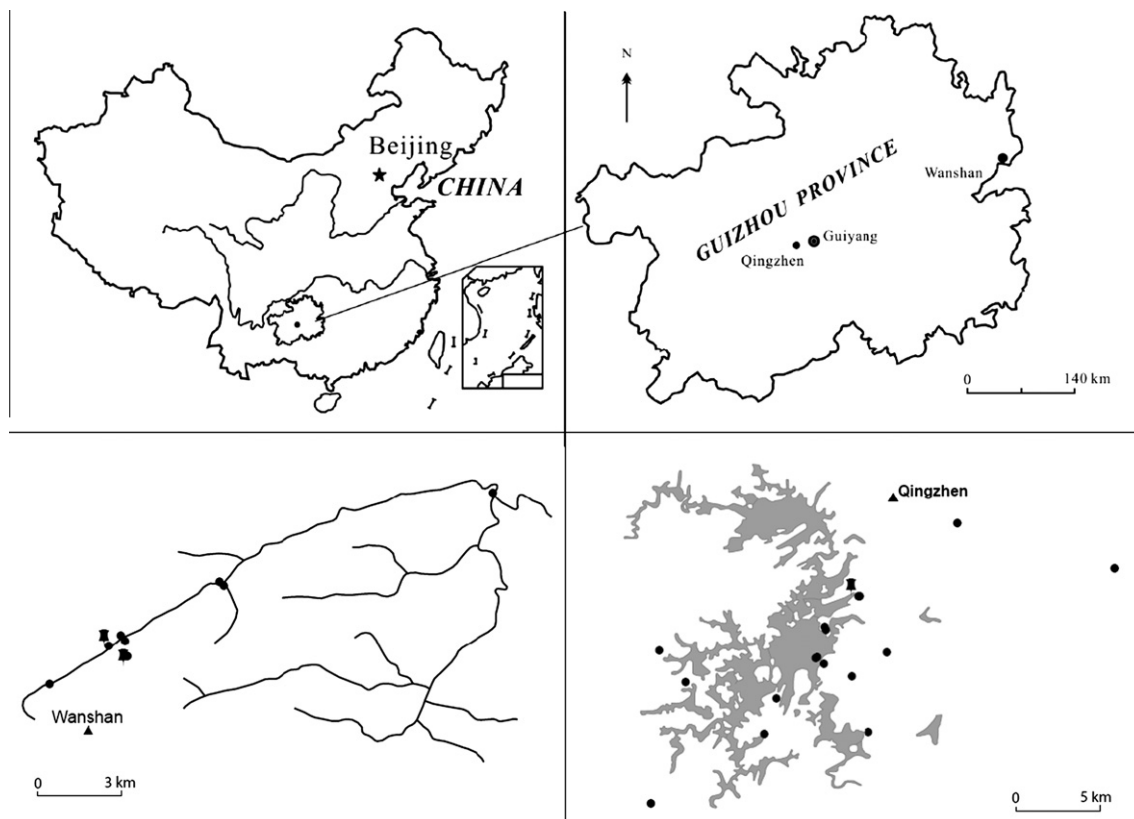
range of reactive forms of PHE to the environment. Widespread coal combustion and industries, such as mining and smelting, are important emission sources of PHE to air and water (Alloway, 1995). Metal mining and smelting activities release not only the metal of interest but also PHE which are associated in the ores (Alloway, 1995).

China's cinnabar (HgS) deposits rank third in the world (Jiang et al., 2006). Guizhou province, located on the circum pacific mercuriferous belt, holds approximately 70% of the total Hg reserves in China. Mercury has been mined in this province for more than 600 a (Jiang et al., 2006) causing elevated levels of Hg in the environment. Furthermore, the mercuriferous belt causes generally high Hg levels in the bedrock and in other natural resources, such as coal (Feng et al., 2002).

Mercury mining, which has generally ceased during the last decade, and industrial production based on coal fired energy have been important industrial sectors in Guizhou province. Both the Hg mining and refining activities and the intense use of coal in power plants have led to large anthropogenic releases of Hg into the environment (Feng et al., 2003a). In the present study, two sampling areas were chosen, representing each of the two types of industrial

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**Fig. 1.** Maps showing the location of Guizhou, the sampling areas and the sampling sites in each region. The Wanshan and the Aozhai Rivers are shown on the left with Hg mine tailings indicated by pin points. Qingzhen is shown on the right with the location of the power plant indicated by a pin point.

impacted areas: Wanshan as example of a Hg mining area and Qingzhen representing a region impacted by coal fired power production and a conglomeration of heavy industry (Fig. 1).

High concentrations of Hg in water, soil and vegetation in and around the Wanshan Hg mining and retorting area have been reported in several studies (Feng et al., 2003a; Horvat et al., 2003; Qiu et al., 2005; Zhang et al., 2004). The main anthropogenic sources of Hg to the environment are seepage from mine tailings mixed with calcine slag heaps as well as air emissions from former artisanal and large scale retorting activities and subsequent deposition on the soil. The focus of all previous studies in the area has been on Hg contamination while other contaminant metals have not been considered.

In the Qingzhen industrial area, PHE contamination is thought to originate from numerous sources, especially a large scale coal fired power plant. The area has, as Wanshan, previously been thoroughly assessed for its levels of Hg in the environment. Special focus has been on the Hg levels downstream of discharges from an organic chemical plant using similar production techniques as the plant causing the Minamata disaster (Horvat et al., 2003). Also in this area there is a lack of assessment of the levels of other PHE.

The objective of the study is to map the concentrations of the selected metals, Hg, Pb, Zn, Cd and Cu, and As, and to consider the relative importance of anthropogenic versus natural sources in the two different pollution regimes.

## 2. Materials and methods

### 2.1. Study areas

The Wanshan region, where Hg mining has been the major industrial activity for centuries, is located in the northeastern

remote part, while Qingzhen industrial area is located in the middle of Guizhou province, in southwestern China (Fig. 1).

The Wanshan and Qingzhen districts have clayey soils which consist of 60–70% and 70–80% clay, respectively (NEPA, 1994). The dominant clay mineral is kaolinite (NEPA, 1994; Yi et al., 1986). The study areas are dominated by a humid climate with mild temperatures and abundant precipitation of 1200–1400 mm annually (Feng and Qiu, 2008; Qiu et al., 2005). The Wanshan district has a karstic landscape with deep valleys and elevations from 270 m a.s.l. to 1149 m a.s.l. (Qiu et al., 2005). The topography of the Qingzhen area is flatter than in Wanshan, but the karstic landscape is still apparent. Elevation of the plain is 1200–1300 m, with valleys and peaks ranging from 765 to 1763 m a.s.l.

In Wanshan, the soil samples were collected in rice paddy fields in the valley along the Aozhai River, one of several major streams in the area, and along two of its tributary rivers (Fig. 1). There are two Hg mine- and retort tailings deposits within the study area; one situated close to the headwaters of the Aozhai River, while the other is situated in one of the tributaries (Meizixi).

In Qingzhen, a large coal fired power plant was used as a centre for the sampling design, and the samples were collected at distances of up to approximately 20 km from the power plant. Bottom ash from the plant is deposited in a big reservoir that was included in the sampling regime (samples B4 and B5). In addition soil samples from a rice paddy field covering an old bottom ash pond (samples B6 and B8) were collected. There are many heavy industries and thereby pollution sources in the area, including the Guizhou Organic Chemical Plant (GOCP), which used Hg-based technology to produce acetaldehyde until 1997 (Feng and Qiu, 2008). Wind is important for transport of PHE emitted to the air. The predominant wind direction in Qingzhen is from the NE (Feng et al., 2003b; LSIGP, 2006).

## 2.2. Sampling and analysis

A total of 37 soil samples were collected in September 2007 (Table 1). Samples were taken from the top 5 cm after removing a thin algal growth layer where present. Four samples were taken from different parts of each rice paddy field and combined into one composite sample. Disposable gloves were used during the sampling. The samples were stored in polyethylene bags with zip lock closure until further processing.

The soil samples were laid out to dry (within two weeks of sampling) for about 2 months at room temperature covered by Al foil to avoid contamination from adjacent air. Furthermore, as temperatures never exceeded 30 °C during the drying period, there should not have been any loss of analytes.

After drying, the samples were sieved through a 2 mm stainless steel sieve. Soil aggregates were crushed in an agate mortar prior to sieving. The sieved soil samples were stored in Teflon bags (Teflon) until analysis.

The samples were digested in concentrated HNO<sub>3</sub> in an autoclave following Norwegian Standard 4770. An aliquot (0.500 g ±

0.005 g) of each soil sample was transferred to a pre-cleaned nalgene bottle, 10 mL of concentrated HNO<sub>3</sub> (65% Romil-SpA Super Purity Acid) added, and the bottle capped before being brought to the boil in the autoclave at 2 atm and approximately 120 °C for 30 min. After cooling, the samples were transferred to pre-cleaned polypropylene bottles and diluted with purified water (>18.0 MΩ cm; at 25 °C) to a resulting volume of 50 mL.

Metal concentrations were determined using a Thermo Finnigan Element 2 (High Resolution) Inductively Coupled Plasma Mass Spectrometer (ICP-MS). Soil Hg concentrations were determined by a Milestone DMA-80 Direct Mercury Analyzer (DMA-80).

Quality assurance and control of the analyses was done through the use of sample replicates, reference material, control solutions and blanks. Results from control runs of reference material (STSD-4, Canadian Certified Reference Material Project) gave recoveries of 93.7%, 106% and 103% for three separate days, an average of 101%, for the DMA-80 method (Hg). For the ICP MS control runs of reference material (MESS-3, National Research Council Canada), the recoveries were 102%, 105%, 93.4%, 78.6% and 76.1% for the elements As, Cd, Pb, Zn and Cu, respectively. The low recoveries of some of the metals might be explained by the decomposition method (NS 4770), which may not extract the most strongly bound metal fractions.

**Table 1**

Short description of the sampling sites<sup>a</sup>.

Sites	Description
E1	Close to the Hongfeng Reservoir. Stagnant water in paddy fields
E2	Close to the Hongfeng Reservoir, opposite side from E1. Some houses nearby
E3	A couple of nearby "farms". Grape fields around the site. Stagnant water in the paddy field. Irrigated by rainwater
E4	Dry paddy field. Tomatoes grown near the site. Irrigated by rainwater?
E5	Site surrounded by karstic hills. Farming area. Wet soil, but not stagnant water
E6	Small valley where the paddy field is surrounded by forests. Close to a place with coal activity
B1	Paddy field approximately 300 m from the power plant. Stagnant water in the field
B2	Vegetable field close to B1
B3	Rice paddy field with stagnant water. Quite close to the power plant
B4	Soil from ash slurry field, by the outlet (bottom ash from the power plant)
B5	Soil from ash slurry field, opposite side of outlet
B6	Soil from paddy field below ash slurry field. Top and middle soil layer has been collected from elsewhere to grow rice. Bottom layer consists of bottom ash from power plant
B8	Another paddy field below ash slurry field. Bedrock visible around the sites (probably limestone)
B11	Stagnant water in rice paddy field. Karstic hills surrounding the site
B14	Paddy field close to a small village. The field has recently been drained
B15	Paddy field beside a stream which passes the GOCP. The paddy field is being drained
B22	The paddy field had riper rice than other sites
F2	By the Aozhai River just before it joins the Xiaxi River. Paddy field irrigated by river water. Downstream a small village
F4	Paddy field irrigated by tributary river. The site is close to a small village
F5	Before merging with the F4 tributary river. Paddy field irrigated by the Aozhai River. Wet soil
F6	Meizixi River (tributary river) irrigates the paddy field. Very wet soil, some black colour in the soil may point to reducing conditions
F7	Paddy field irrigated by the Aozhai River before the river joins the F6 tributary river. Not very wet soil
F8	Meizixi tributary river runs past Hg mine tailings. The paddy field is irrigated by the river. The field is not very wet
F9	Furthest up in the Aozhai valley; apparently above any Hg activity. Irrigation source to paddy field not known. The field is not very wet, and contains more stones
F10	Samples taken below Hg mine tailings. Channels irrigating the paddy field with river water. Mining activity on the other side from the mine tailings, will probably not affect the river water at this site (except from possible deposition)

<sup>a</sup> E and B samples come from the Qingzhen sampling area, while F samples are collected in the Aozhai Valley in Wanshan.

## 3. Results

### 3.1. Potentially harmful element levels in the mercury mining and smelting area, Wanshan

Mercury levels were found to be particularly high, though they show large spatial variations, with concentrations ranging between 1–119 µg g<sup>-1</sup> (Fig. 2). The concentrations are low upstream of the mine tailings and in the tributary without tailings. The highest concentrations in the paddy fields are found just downstream of the tailings and decrease rapidly with increasing distance downstream (Fig. 3). At the sampling site 0.7 km down the valley from the main tailings the Hg concentration is about a quarter of the concentration at the site next to the tailings.

Cadmium and Zn levels display the same spatial trend as is observed for the Hg concentrations. The rice paddy fields having the highest concentrations of Cd (2.2 and 1.3 µg g<sup>-1</sup>) and Zn (193 and 171 µg g<sup>-1</sup>) are situated immediately below Hg mine tailings (Fig. 2), with low concentrations upstream and decreasing concentrations downstream with distance from the tailings.

Arsenic concentrations are low and show no clear spatial trend. Rice paddy soil upstream of the Hg mining activities has similar, or somewhat higher, As concentrations to that observed in paddy fields further downstream (all in the range 3–22 µg g<sup>-1</sup>) (Fig. 2). Levels of Pb and Cu in the soils are low, ranging from 23–52 µg g<sup>-1</sup> and 21–44 µg g<sup>-1</sup>, respectively (Fig. 2).

### 3.2. Potentially harmful element levels in the Qingzhen industrial area

Mercury concentrations in rice paddy soils in Qingzhen region show a much lower range than in Wanshan (Fig. 4), with no clear spatial pattern (range 0.1–2 µg g<sup>-1</sup>). The highest concentration was found in a paddy field irrigated with water draining the GOCP (1.7 µg g<sup>-1</sup>) (Fig. 4). The bottom ash reservoir contained, as might be expected, low levels of Hg (0.1 and 0.3 µg g<sup>-1</sup>), due to the loss of Hg during coal combustion.

Cadmium and Zn were found in concentrations reaching 1.1 and 144 µg g<sup>-1</sup>, respectively. The lowest Cd level was found along with the lowest Hg concentration in the ash slurry pond from the power plant, while the highest amount of Zn was found with the highest Hg levels in the rice paddy field downstream of the GOCP.

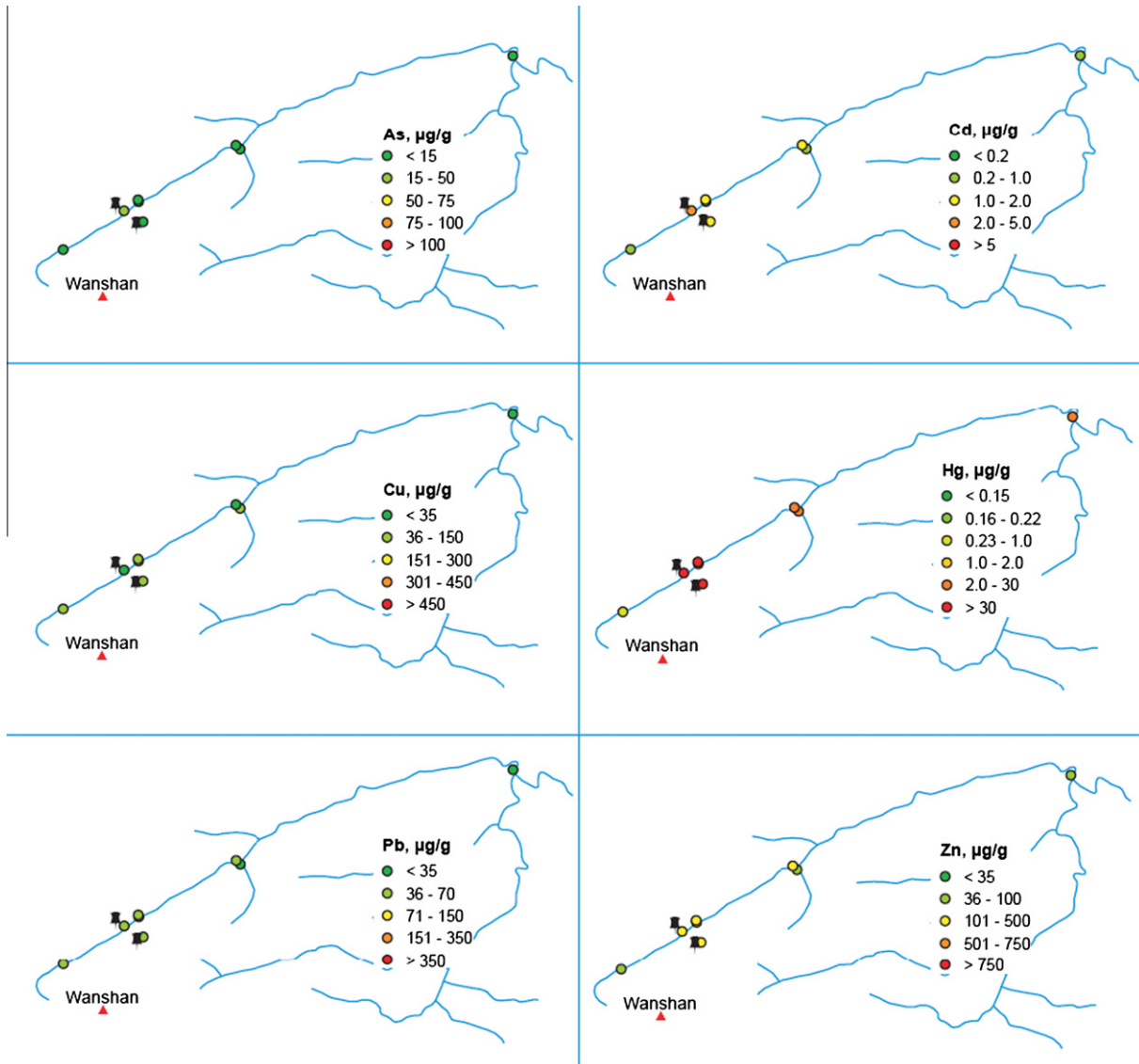


Fig. 2. Location maps of Wanshan (Aozhai River) with sampling points and the respective PHE concentrations in soil.

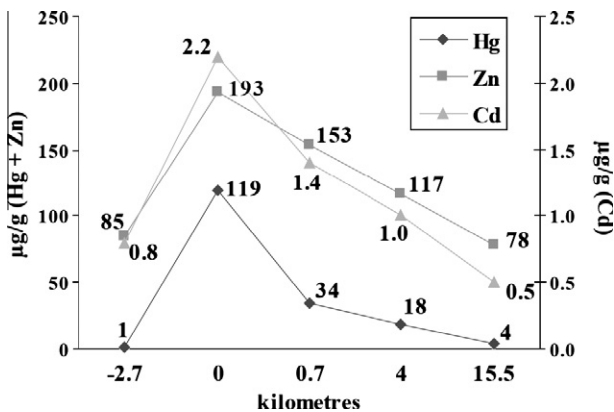


Fig. 3. The Hg, Zn and Cd concentrations in rice paddy fields upstream and downstream of the biggest Hg mine tailings ('0 km' is the location of the tailings). Distances downstream are given on the x-axis.

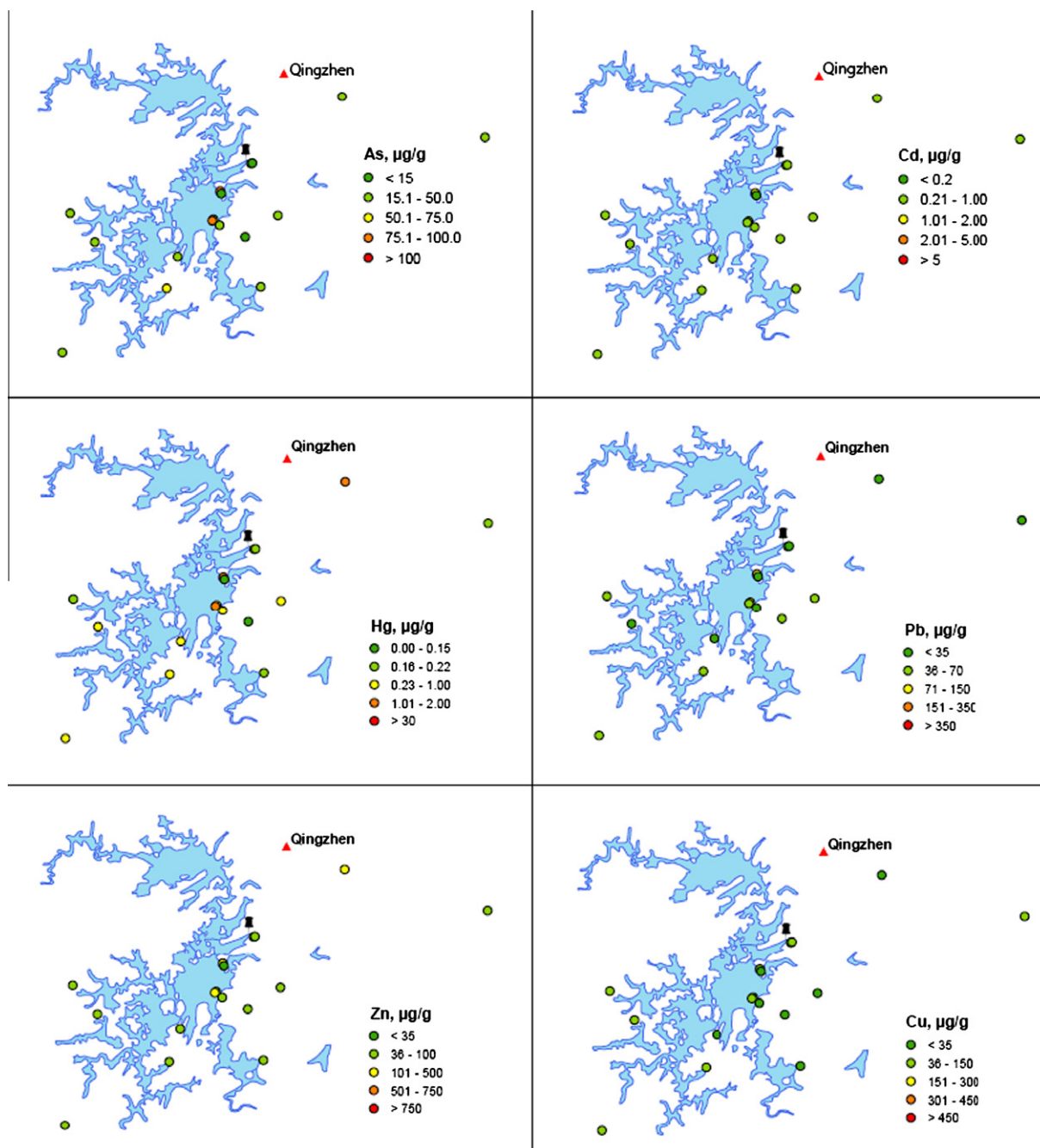
The Pb levels vary from 26–146 µg g<sup>-1</sup>, while Cu is found in concentrations ranging from only 13 to 58 µg g<sup>-1</sup>. Average concen-

tration of As in soils is 41 µg g<sup>-1</sup>, with the highest concentration (90 µg g<sup>-1</sup>) found in close vicinity to the power plant (Fig. 4).

#### 4. Discussion

Mercury levels in environmental media in the Wanshan area are known to be exceptionally high (Feng et al., 2003a; Horvat et al., 2003; Qiu et al., 2005; Zhang et al., 2004). The natural background levels of Hg in soils in the region are, according to NEPA (1994), in the range of 0.02–0.22 µg g<sup>-1</sup>. The concentrations found in this study all exceed this range, even in the sampling sites with no direct influence of water leaching from the mine tailings. With the exception of the sample from upstream of the main tailings, the contamination levels also exceed level C (1.5 µg g<sup>-1</sup>) of The Chinese National Standard for Soil Environmental Quality (Chen et al., 1999), which is slightly lower than category “moderately polluted” in the Norwegian classification of metals in soil (2–4 µg g<sup>-1</sup>) (SFT, 2009). This contamination level is considered non-suitable for e.g. agricultural or residential use.

In Qingzhen, 11 of the 29 samples have Hg concentrations which exceed level B in The Chinese National Standard for Soil



**Fig. 4.** Maps of Qingzhen with sampling points and the respective PHE concentrations in soil. (The samples from sampling points located within the reservoir are actually taken from the ash reservoir).

Environmental Quality,  $1 \mu\text{g g}^{-1}$  (Chen et al., 1999), and 3 samples exceed level C,  $1.5 \mu\text{g g}^{-1}$ . Very high concentrations ( $14.3\text{--}354 \mu\text{g g}^{-1}$ ) of Hg in soil downstream of the GOCP have previously been reported in a study by Horvat et al. (2003). The highest Hg concentration in this study, although much lower ( $2 \mu\text{g g}^{-1}$ ), was also found along the same stream draining from this plant.

The Hg levels in the soils from Qingzhen are in general much lower than in the Wanshan area (Figs. 2 and 4). As the Wanshan area has been subject to vast Hg mining and smelting activities for hundreds of years, this difference was expected. Nevertheless, most industry in Qingzhen emits Hg, mainly to the atmosphere. This along with a flatter topography allows the Hg to be transported and dispersed throughout the region from its emission sources.

Background levels of Cd in soil in the sampling area has been reported by NEPA (1994) to be in the range of  $0.1\text{--}2 \mu\text{g g}^{-1}$  and Zn in the range of  $35\text{--}140 \mu\text{g g}^{-1}$ . In Qingzhen, the levels of Cd in the samples are well within this range (average  $0.4 \mu\text{g g}^{-1}$ ), mostly at the lower end. However, in Wanshan the concentrations are higher (average  $1.1 \mu\text{g g}^{-1}$ ), with one sample exceeding the range and several samples at the higher end of this range. The average Zn concentration in Wanshan soil ( $128 \mu\text{g g}^{-1}$ ) is also higher than the average concentration in Qingzhen soil ( $81 \mu\text{g g}^{-1}$ ). Mining and smelting activities of sulfide ores are thus of greater concern for the local environment than coal combustion with regard to both Cd and Zn.

The lack of spatial trends and low amounts of As within the natural background levels ( $6.2\text{--}50 \mu\text{g g}^{-1}$ ) (NEPA, 1994) in rice paddy

fields in Wanshan indicates that there are no significant anthropogenic sources of this element to the local environment.

The anthropogenic As in Qingzhen was thought to be mainly from emissions to the atmosphere from the major coal fired power plant, although no conclusion regarding connection between the transport pattern and the main wind direction at the plant can be made, as there are too few samples. Moreover, coal burning in the numerous industrial units throughout the region appears to

**Table 2**  
Correlation matrices for heavy metals in Qingzhen and Wanshan.

	As	Cd	Cu	Hg	Pb	Zn
<i>Qingzhen</i>						
As	1					
Cd	0.64	1				
Cu	0.37	0.25	1			
Hg	0.70	0.82	0.21	1		
Pb	0.84	0.76	0.34	0.79	1	
Zn	0.56	0.59	0.45	0.73	0.59	1
<i>Wanshan</i>						
As	1					
Cd	0.85	1				
Cu	-0.09	0.18	1			
Hg	0.62	0.91	0.23	1		
Pb	0.77	0.77	0.54	0.58	1	
Zn	0.65	0.90	0.45	0.84	0.74	1

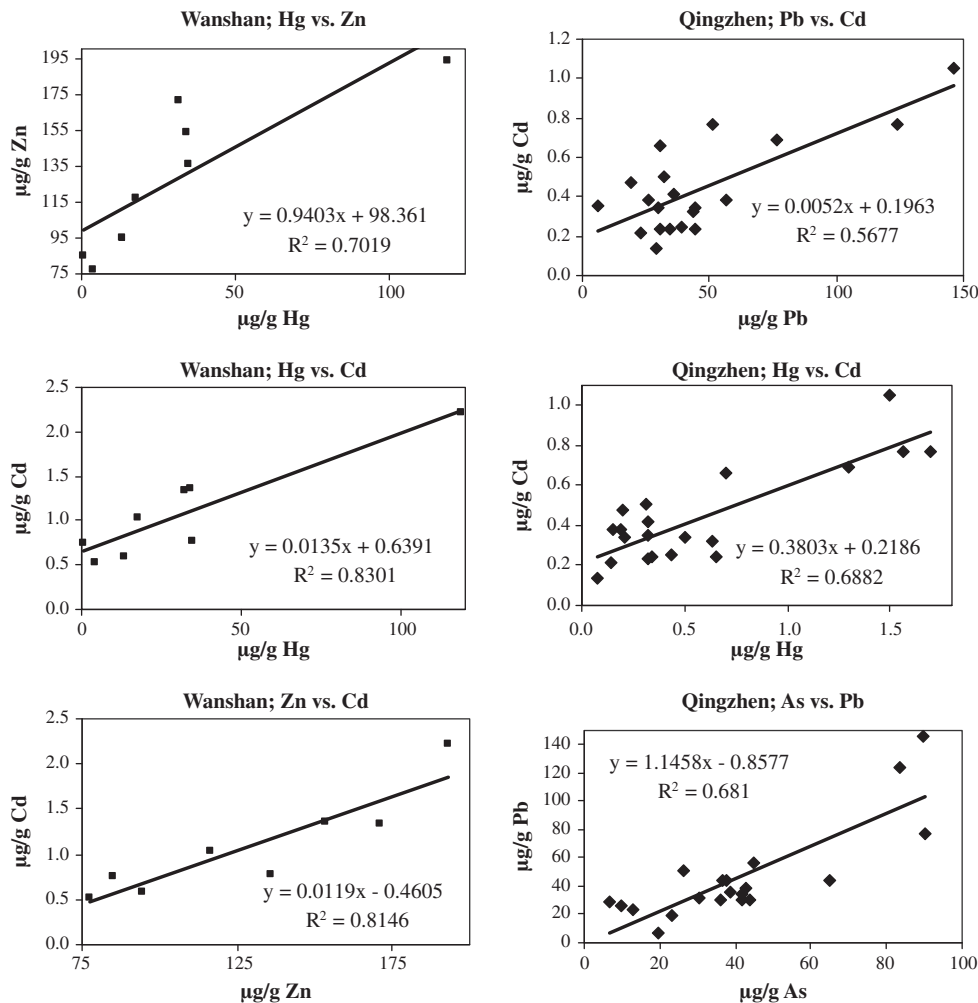
contribute significantly to the overall As contamination. Soil As concentrations are as high as  $90 \mu\text{g g}^{-1}$ , which exceeds natural background levels of  $6.2\text{--}50 \mu\text{g g}^{-1}$  (NEPA, 1994).

Lead levels in both Wanshan and Qingzhen soils are all within the natural background concentration range ( $10\text{--}300 \mu\text{g g}^{-1}$ ) for the region reported by NEPA (1994). Lead was used as an additive to gasoline in China until June 2000 (SEPA, 2000). The industrialized area of Qingzhen has an average concentration reaching  $47 \mu\text{g g}^{-1}$ , while Wanshan has a slightly lower average concentration of  $39 \mu\text{g g}^{-1}$ . The somewhat higher concentration in Qingzhen may be explained by a higher population and thus more emission of Pb from transportation prior to when the Pb additive was phased out.

Copper concentrations in Wanshan are all lower than grade B ( $100 \mu\text{g g}^{-1}$ ; soil pH 6.5 to  $>7$ ), and some are also lower than grade A ( $35 \mu\text{g g}^{-1}$ ), in The Chinese National Standard for Soil Environmental Quality (Chen et al., 1999). Low levels of Cu are also found in Qingzhen; average is  $35 \mu\text{g g}^{-1}$ , the same as grade A in The Chinese National Standard for Soil Environmental Quality (Chen et al., 1999).

Correlation matrices were obtained to discover empirical similarities among the metals and As (Table 2) along with scatter charts (some are given in Fig. 5). Correlation between metals may indicate similar origins or comparable chemical properties.

There is a very strong correlation between Cd and Zn concentrations in Wanshan ( $0.90$ ,  $R^2 = 0.82$ ,  $P < 0.01$ ). High correlations ( $0.91$ ,



**Fig. 5.** Scatter charts showing concentration relationships between different PHEs.

$R^2 = 0.83$ ,  $P < 0.01$ ; and  $0.84$ ,  $R^2 = 0.71$ ,  $P < 0.005$ ) between Cd and Hg, and Zn and Hg, are also found (Table 2, Fig. 5). As these are all chalcophile elements they have an affinity for S and are likely impurities in the mined Hg sulfide ores (Feng et al., 2004). This moreover implies that Hg mining and smelting activities are likely the main sources of all 3 metals.

Stream water is used for irrigation of the rice paddy fields downstream of the Hg tailings, and hence the paddy fields become contaminated by the polluted water. The sharp decrease in the concentrations of Hg, Cd and Zn in soils with increasing distance from Hg mine tailings is probably due to particle scavenging and/or settling in the river water. This theory is supported by the study of Yan et al. (2010), which found that 60–90% of Hg in river water in the Wanshan area is bound to particles which settle rapidly with increasing distance from the source.

The correlation between Zn and Cd might also originate from Zn ores where Cd is incorporated in the lattice (Nordberg and Nordberg, 2002).

In Qingzhen, the low Cd levels are strongly correlated with the level of Hg contamination ( $0.83$ ,  $R^2 = 0.69$ ,  $P < 0.001$ ) (Table 2, Fig. 5). Furthermore, Zn levels also correlate with Hg levels ( $0.72$ ,  $R^2 = 0.52$ ,  $P < 0.001$ ) (Table 2), similar to what is found in Wanshan, although somewhat weaker. The common correlation/similarity ascribes coal combustion as a shared source for these contaminants (US-EPA, 2007), along with the GOCP.

In Qingzhen, Pb correlates with As ( $0.83$ ,  $R^2 = 0.68$ ,  $P < 0.001$ ) and Cd ( $0.75$ ,  $R^2 = 0.57$ ,  $P < 0.001$ ), while the correlation between As and Cd is rather weak ( $0.64$ ,  $R^2 = 0.41$ ,  $P < 0.005$ ). All the elements may, therefore, originate from the widespread coal combustion in the industrialized area.

## 5. Conclusions

The Wanshan area is renowned for its high levels of Hg contamination. Mercury concentrations in the soils decrease rapidly with increasing distance from the sources (mine tailings). Cadmium and Zn levels are highly correlated with moderate to high levels in the soils directly downstream of the Hg mine tailings. The main contamination source for Cd and Zn in Wanshan is therefore also probably the mine tailings as such chalcophile elements are associated with the cinnabar ore. The levels of Cu, Pb and As in Wanshan were found to be similar to natural background values.

The Qingzhen industrial area has generally moderate to high levels of PHE contamination as a result of both coal combustion and numerous other industrial activities. The elements mainly associated with coal combustion, As and Hg, are found in concentrations higher than the background levels for the area.

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