Chemical Speciation & Bioavailability

ISSN: 0954-2299 (Print) 2047-6523 (Online) Journal homepage: https://www.tandfonline.com/loi/tcsb20

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To cite this article: Chengli Zhang, Shaoyun Ren, Hongdan Cheng, Weiping Zhang, Jianhua Ma, Chaosheng Zhang & Zhiyong Guo (2018) Thallium pollution and potential ecological risk in the vicinity of coal mines in Henan Province, China, Chemical Speciation & Bioavailability, 30:1, 107-111, DOI: 10.1080/09542299.2018.1513820

To link to this article: https://doi.org/10.1080/09542299.2018.1513820

Published online: 24 Aug 2018.

CHEMICAL SPECIATION & BIOAVAILABILITY
2018, VOL. 30, NO. 1, 107–111
https://doi.org/10.1080/09542299.2018.1513820

Thallium pollution and potential ecological risk in the vicinity of coal mines in Henan Province, China

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ABSTRACT
This study was aimed to analyze the thallium pollution and assess the potential ecological risks in the vicinity of coal mines in Henan province, China. We studied 90 surface farmland soil samples from 9 representative coal mines. The Tl concentrations were determined and the potential ecological risks were evaluated. Investigations revealed the farmland soils were modestly contaminated and the trace elements in coal mining areas transferred to the surface soils. Soil Tl contents and potential ecological risks in coal mining areas were significantly increased compared with the original soils which came from the villagers' mud houses built 40 years ago. The soil Tl concentrations ranged from 0.25 to 0.77(mean = 0.46) mg·kg^{-1}, which were higher than the original level (0.42 mg·kg^{-1}). The potential ecological risk index of Tl ranged from 24.00 to 73.2 (mean 44.08), representing a moderate pollution level as a whole of the soils in Henan. In general, high Tl concentrations and high potential ecological risk were found around SHQ and DTG. The soil Tl concentrations exceed the original level and pose noticeable ecological risks.

ARTICLE HISTORY
Received 1 July 2018
Accepted 6 August 2018

KEYWORDS
Coal mines; Tl; potential ecological risk

1. Introduction

Thallium (Tl) is one of the 13 priority pollutant metals that are more dangerous than cadmium (Cd) and mercury (Hg) [1]. This heavy, volatile and highly incompatible trace metal is uncommon in natural systems but important in anthropogenic systems due to its toxicity and probability of causing severe ecological risk at low concentration in environment [2]. Tl has been detected in concentrations sufficiently high to be of economic interest as a byproduct from a number of ore deposits in China [3]. The first reported adverse health impact of Tl pollution in China occurred in a rural area at the Hg-Tl deposit at Lanmuchang in Southwest China in 2004, where Tl enrichment in the aqueous system imposed potential environment risk [4]. Similarly coal resource mining and smelting industry would discharge Tl and its compounds into environment, increasing Tl levels in water, soils and crops, which can directly threaten human health through contacting and food chains. However, there is lack of information in literatures regarding Tl exposure from coal mining areas. Thus, it is urgent to characterize Tl and evaluate its potential ecological risk.

Henan, a large coal producing province, produces nearly 1/10 of the total output in China [5]. National Statistical Yearbook 2015 shows Henan has coal reserves of 8.694×10^8 t. The total areas of coal strata in Henan is 62,815 km^2, or 37.6% of the total provincial areas [6]. In this study, we collected 90 topsoil (0–20 cm) samples from different sites of different coal mining areas in Henan. The objectives were to (1) evaluate the degree of Tl contamination from farmland soils; (2) assess the potential ecological risks posed by these contaminated soils; (3) provide a reference for the local coal mines Tl pollution evaluation.

2. Materials and methods

2.1. *Study areas*

The studied mine sites are located in Xinmi, Dengfeng and Yuzhou, in the northwest of Henan (34°15′13″-36° 31′58.35″ N, 113°4′42.23″ −113°29′51.36″ E), three of the main coal mining regions in China. These three regions enjoy a typical monsoon climate of medium latitudes with an average annual temperature of 14°C and rainfall of 500–900 mm. The topography there is dominated by plains and mountains and a small proportion of low mountains in the southwest, with elevations ranging from 23.2 to 2413.8 m. These regions have developed agricultural activities, and the main crops are wheat and maize. Our investigation shows a number of mining, manufacturing, metallurgy, fertilizer, chemical, and other industrial plants are located in these areas. These industries might be mainly responsible for a load of pollutants that directly and significantly impact the soil environmental quality in the vicinity of mines.

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2.2. *Mining soil sampling and sample preparation*

Totally 90 surface soil samples (0–20 cm) were collected around 9 coal mining areas from May 2016 to September 2016. Take the coal mine as the pollution source, the sampling point inside 2 Km and along the four directions in each mining areas. According to the local situation, we have collected 10 samples in HD, ZZYX, XDP, GG, SHQ, DTG and JS coal mines, respectively. And we collected 11 samples in HST, 9 samples in BPX. Sampling sites were selected on farmland and georeferenced with portable GPS. At each sampling site, surface soil (0–20 cm) was sampled with a stainless steel spade and stored in a plastic bag. Each sample was a combination of 5 subsamples collected from the same site. Impurities such as pieces of brick and tile, iron scraps, wood scraps, plastics and lime grains were removed. Then the samples were air-dried, ground, and sieved through a 0.15-mm nylon sieve for heavy metal analysis.

2.3. *Laboratory analysis*

Soil pH was determined with a 1:2.5 (w/v) ratio of soil to water using a digital pH meter (pHS-3C, Shanghai INESA Scientific Instrument Co., Ltd., China) [7]. Soil organic matter content was determined by the method of potassium dichromate heating oxidation-volumetric [8]. The soil moisture was estimated by weighing method [9]. Soil particle size was determined by the laser diffraction method [10].

Soil samples were digested in a graphite digestion instrument with a mixture solution of concentrated HCl–HNO_3–HF–$HClO_4$ [11]. The digested soil samples were each washed into a 100-mL flask, diluted with double-deionized water and filtered through 0.45-μm membranes. Soil Tl concentrations were measured using an inductively coupled plasma mass spectrometer (ICP-MS; X-Series II Model, Thermo Fisher Scientific, USA). The quality assurance and control procedures were conducted by the standard reference ESS-2 from China with the recoveries of the seven heavy metals between 95% and 105%. Duplicated samples for each metal were analyzed simultaneously with the standard deviations within 5%.

2.4. *Potential ecological risk index*

The potential ecological risk index presented by Hakanson was used to assess the ecological risk of heavy metals [12]. This index is based on the natural and environmental characteristics of heavy metals put forward from the perspective of sedimentation, and evaluates heavy metals in soil or sediment. In addition to heavy metal contents, this method also associates with ecological effects, environmental effects and toxicology, and adopts a comparable and equivalent attribute classification [13]. It assigns the metal/metalloid pollution and classifies the pollution degree of a heavy metal into five (0–5 grade) classes, ranging from background concentration to very heavy contamination. The single-factor pollution index (C_f^i) and single-factor potential ecological risk index (E_r^i) are calculated as follow:

$$C_f^i = C_D^i / C_R^i \tag{1}$$

$$E_r^i = T_r^i \times C_f^i \tag{2}$$

where C_D^i is the measured concentration of a pollutant in a sample; C_R^i is the standard value of the pollutant, namely the original soil quality;T_r^i is toxic response parameter.$C_f^i < 1, 1 \le C_f^i < 3, 3 \le C_f^i < 6$, and $C_f^i \ge 6$ represent slight, moderate, heavy and serious pollution respectively [14]. Five categories of E_r^i are low ($E_r^i < 40$), moderate ($40 \le E_r^i < 80$), considerable ($80 \le E_r^i < 160$), high ($160 \le E_r^i < 320$) and very high ($E_r^i \ge 320$) [15]. In this study, we take Tl content of the original soils as C_R^i value, and the C_R^i value is 0.42 mg·kg^{-1}. Since there is no toxic response parameter of Tl, and Tl is more toxic than Hg, so we take the toxic response parameter of Hg (40) as the T_r^i value.

3. Discussion and results

3.1. *Major physicochemical characteristics of soils*

Analysis on the pH of soil in samples was shown in Figure 1. The studied area has the trend to neutral or alkaline soil. As we can see from picture, in the samples we studied, most soil samples were greater than 7 and 86% (percent was sampling points ration) of the pH values was between 7.21 and 8.63. In the 90 soil samples we studied, there were 9 (10%) soil samples lower than 6.5, and 14 (16%) soil samples between 6.5 and 7.5, and 67 (74%) soil samples were greater than 7.5. The soil with pH from 5.35 to 8.63 and the average value is 7.71. It is worth mentioning that, among the 9 mine areas, HD, HST, ZXYX, BPX and JS were belong to alkaline soil, only GG was acidic soil.

Soil organic matter in the samples we studied was shown in Figure 2. Soil organic matter could influence soil physicochemical properties and reflect soil nutrition [16]. The range of soil organic matter is from 0.36 to 13.71% (percent was sampling points ration) with the average of 4.28%. The content of soil organic matter in DTG coal mine was the highest and with the average value of 7.82%.

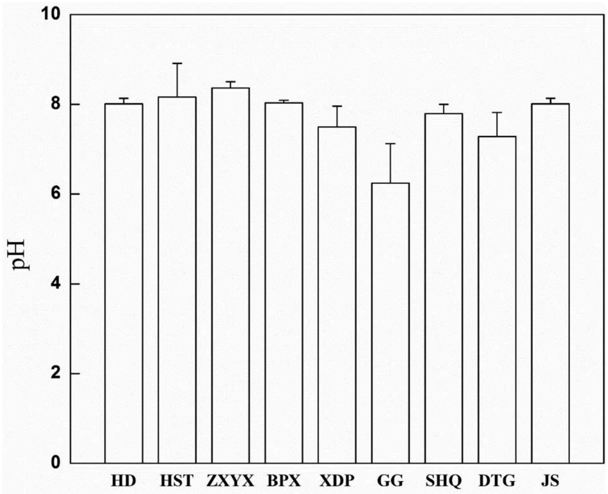

Figure 1. The distribution of soil pH.

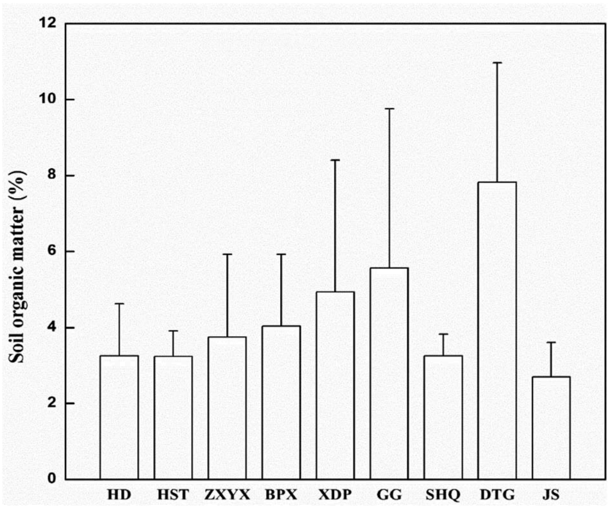

Figure 2. The distribution of soil organic matter.

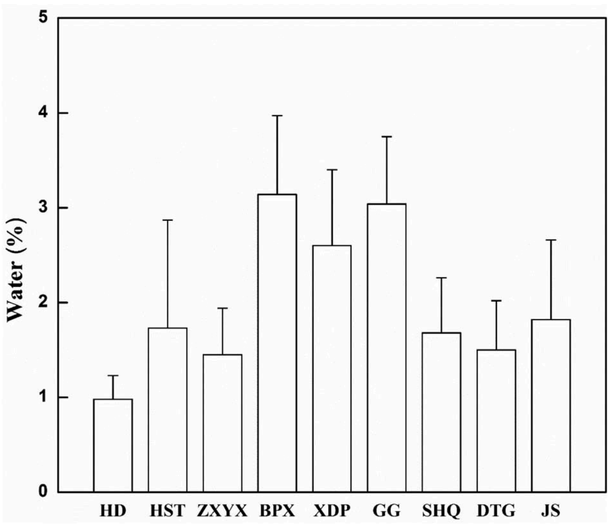

Figure 3. The distribution of soil water content.

Soil water content is an important parameter of soil physical and chemical characteristic [17]. As shown in Figure 3, in the samples we studied, the range of soil water content is from 0.32 to 4.76% (percent was sampling points ration) with the average of 1.99%.

3.2. *Analysis of Tl concentrations in soil samples*

The Tl concentrations in all samples are presented in Table 1. Among all samples, the measured Tl concentrations ($mg \cdot kg^{-1}$) vary between 0.38 and 0.49 (mean ± SD = 0.45 ± 0.04) in HD Mine, 0.33 and 0.52 (0.45 ± 0.05) in HST Mine, 0.39 and 0.52 (0.46 ± 0.04) in ZXYX Mine, 0.25 and 0.77 (0.46 ± 0.14)in BPX Mine, 0.38 and 0.58 (0.47 ± 0.06) in XDP Mine, 0.26 and 0.51 (0.46 ± 0.07) in GG Mine, 0.44 and 0.58 (0.48 ± 0.04) in SHQ Mine, 0.44 and 0.53 (0.48 ± 0.03) in DTG Mine, 0.40 and 0.52 (0.46 ± 0.04) in JS Mine. In the samples we studied, the Tl concentrations of these sites have shown two characteristics (Table 1). (1) The average Tl contents in surface soil are mostly higher than the original value (0.42 $mg \cdot kg^{-1}$), there were 73 (81.1%) (percent was sampling points ration), 3 (3.3%) and 14 (15.6%) samples were above, equal to and below the original value, respectively. (2) The Tl contents in soil samples vary largely between 0.25 and 0.77$mg \cdot kg^{-1}$. The results indicate the Tl contamination in Henan.

In the samples we studied, DTG has the highest soil organic matter and concentration of Tl. Heavy metals were largely enriched in particulate organic matter, which could impact the further mineralization of soil organic matter [18].

3.3. *Potential ecological risk assessment*

Potential ecological risk index is generally considered as a metric that quantitatively reflects the overall potential ecological risk due to co-contamination [19].The E_r^i values of Tl represent the low slight to moderate potential ecological risk （Table 2）. $E_r^i$is unevenly distributed: the maximum, mean and minimum E_r^i are 73.20, 44.08 and 24.00, respectively. The maximum E_r^i of soil Tl belongs to the moderate potential ecological risk. The E_r^i mean value maximizes in DTG (45.80) and minimizes in HST (42.36), posing a moderate potential ecological risk in all samples. Both SHQ and DTG show moderate potential risks ($40 \leq E_r^i < 80$). The other samples show low ($E_r^i < 40$) or moderate ($40 \leq E_r^i < 80$) potential ecological risks.

In the samples we studied, we found the values of the potential ecological risk indexes in BPX and GG varied greatly among sampling sites. The values of BPX are low in the west and south, high in the east and north, and have a large slope. The values of GG are low in the middle, high in the north and south. This is mainly caused by the topographic, resulting in a change in the spread of the pollution sources.

Table 1. Statistics of soil Tl contents in different coal mines/mg·kg^{-1}.

	HD	HST	ZXYX	BPX	XDP	GG	SHQ	DTG	JS
N	10	11	10	9	10	10	10	10	10
Max	0.49	0.52	0.52	0.77	0.58	0.51	0.58	0.53	0.52
Min	0.38	0.33	0.39	0.25	0.38	0.26	0.44	0.44	0.40
Mean	0.45	0.45	0.46	0.46	0.47	0.46	0.48	0.48	0.46
Median	0.45	0.45	0.47	0.47	0.46	0.48	0.47	0.48	0.46
SD	0.04	0.05	0.04	0.14	0.06	0.07	0.04	0.03	0.04
Cv (%)	8.39	10.24	8.90	0.29	13.67	14.60	7.78	5.38	7.54

Table 2. Single potential ecological risk of sample frequency distribution (E_r^i).

				Sample frequency distribution				
				E_r^i<40	40≤E_r^i<80	80≤E_r^i<160	160≤E_r^i<320	E_r^i≥320
Level	Minimum	Maximum	Mean	Low	Moderate	Considerable	High	Very high
HD	36.00	46.80	42.56	3	7	0	0	0
HST	31.60	49.60	42.36	2	9	0	0	0
ZXYX	37.20	49.60	44.20	1	9	0	0	0
BPX	24.00	73.20	44.08	3	6	0	0	0
XDP	36.00	55.20	44.60	3	7	0	0	0
GG	24.80	48.40	43.36	1	9	0	0	0
SHQ	42.00	55.20	45.64	0	10	0	0	0
DTG	42.00	50.40	45.80	0	10	0	0	0
JS	38.00	49.60	44.24	1	9	0	0	0

4. Conclusions

The mean Tl concentrations exceeded the original value among 90 soil samples collected from coal mining areas, Henan province. In general, analysis showed the average Tl contents in surface soils exceeded the original value. The thallium contents in soil samples varied largely between 0.25 and 0.77 mg·kg^{-1}. Compared with the original thallium content (0.42 mg·kg^{-1}), Tl concentrations were moderate in most samples. SHQ and DTG were under moderate potential risks. The contributions of other samples were low or moderate potential ecological risks. Therefore, identifying the soil Tl contents, particularly in developed industrial areas, is imperative for policy-making on controlling the contamination level and improving soil quality.

Acknowledgments

We sincerely appreciate the laboratory and diving assistance by the Science & Technology Innovation Team in Universities of Henan Province (19IRTSTHN029).

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work was supported by the National Science Foundation for Young Scholars of China under Grant [41601522], China Postdoctoral Science Foundation under Grant [2017M612387], and Henan Postdoctoral Science Foundation under Grant [165913].

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