



Pollution, ecological-health risks, and sources of heavy metals in soil of the northeastern Qinghai-Tibet Plateau

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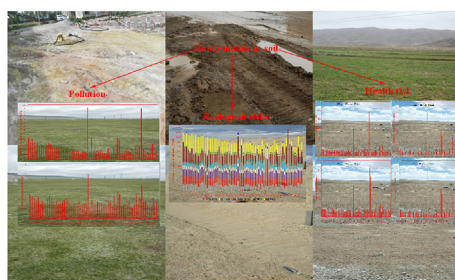
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HIGHLIGHTS

- Serious soil heavy metal pollution occurred in the study area.
- The study area showed high ecological risks posed by soil heavy metals.
- Cancer risks of heavy metals were high, especially for children.
- Non-cancer risks of heavy metals for children were high.
- Industrial activities might be the main source for heavy metals in soil.

GRAPHICAL ABSTRACT



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ABSTRACT

The Qinghai-Tibet Plateau, especially the northeastern region, is not a pure land any more due to recently increasing anthropogenic activities. This study collected soil samples from 70 sites of the northeastern Qinghai-Tibet Plateau to evaluate pollution, ecological-health risks, and possible pollution sources of heavy metals. The concentrations of heavy metals in soil were relatively high. Values of geo-accumulation index exhibited that Hg pollution was the most serious meanwhile Hg possessed the strongest enrichment feature based on enrichment factor values. The modified degrees of contamination showed that about 54.3% and 17.1% of sampling sites were at moderate and high contamination degree while pollution load indexes illustrated that 72.9% and 27.1% of sampling sites possessed moderate and high contamination level, respectively. Ecological risk indexes of heavy metals in soil ranged from 234.6 to 3759.0, suggesting that most of sites were under considerable/very high risks. Cancer risks for adults and children were determined as high and high-very high levels while non-cancer risks for children were high although those for adults were low. Industrial source contributed to the main fraction of ecological and health risks. Summarily speaking, heavy metals in soil of the study area has caused significantly serious pollution and exerted high potential ecological and health risks, especially for children who are more susceptible to hurt from pollutants. Therefore, more efficient and strict pollution control and management in study area should be put out as soon as possible.

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1. Introduction

Soil, an important sink of nutrients and pollutants, plays critical function in social-ecological stability and safety. However, soil pollution has become an important obstacle for regional development and human health in recent decades (Jiang et al., 2017; Liang et al., 2017; Padoan et al., 2017; Pan et al., 2016; Peng et al., 2017; Sakai et al., 2017). Heavy metals, defined as metals or metalloids with a specific density larger than 5 g/cm³ (Järup, 2003; Oves et al., 2012), have become an important kind of pollutants in soil all over the world (Kowalska et al., 2016; Rachwał et al., 2017; Tepanosyan et al., 2017a; Tian et al., 2016; Wu et al., 2016a).

Originated from natural and anthropogenic sources, heavy metals possess acute and chronic toxicity, environmental persistence, and bioaccumulation to exert potential risks to the ecosystem and human health (Borges et al., 2015; Jiang et al., 2017; Salmanighabeshi et al., 2015; Zhu et al., 2017). Therefore, more research starts focusing on the ecological and health risks of heavy metals besides their distribution and pollution (Jiang et al., 2017; Peng et al., 2017; Tepanosyan et al., 2017a, 2017b; Xiao et al., 2017; Zhu et al., 2017). Moreover, multiple methods such as geo-accumulation index (I_{geo}), modified degree of contamination (mC_d), enrichment factor (EF), and pollution load index (PLI) have been employed to comprehensively evaluate the heavy metal pollution in soil (Ćujić et al., 2016; Tian et al., 2017; Zhu et al., 2017). Furthermore, source apportionment of heavy metals in soil also attracts increasing attention (Jiang et al., 2017; Peng et al., 2017).

The Qinghai-Tibet Plateau, generally regarded as the pure land, has exhibited unexpected heavy metal pollution (Bing et al., 2014; Huang et al., 2008; Sheng et al., 2012; Wu et al., 2016a; Yang et al., 2007, 2011; Zhang et al., 2013). Heavy metals were frequently detected in biota of the Qinghai-Tibet Plateau with the highest Pb concentrations in fish/plant samples of 0.079/62.1 mg/kg and the maximal Hg concentration in fish samples of 2384 µg/kg, respectively (Bing et al., 2014; Yang et al., 2007, 2011). Heavy metals were also widely detected in different environmental matrices with the highest Pb/Cr concentrations in soil and water samples of 1075.69/3429.00 mg/kg and 781/2.74 µg/L, respectively (Huang et al., 2008; Sheng et al., 2012; Zhang et al., 2013). Therefore, heavy metal pollution in the Qinghai-Tibet Plateau should not be neglected. However, it is regretful that the previous studies have not illustrated the thorough information on the pollution, ecological and health risks, and source identification of heavy metals in soil of the Qinghai-Tibet Plateau, especially in the northeastern part that is the area with the most extensive anthropogenic disturbance of the whole plateau. Therefore, this study adopted different methods to assess the pollution, ecological-health risks, and source of heavy metals in soil of the northeastern Qinghai-Tibet Plateau. The objectives of this study are to provide complete and comprehensive information on heavy metals in soils of the northeastern Qinghai-Tibet Plateau and lay a basis for the soil pollution prevention and control of the high-elevation areas.

2. Materials and methods

2.1. Study area, sampling strategy, and detection methods

The study area locates in the northeastern Qinghai-Tibet Plateau. Field sampling was performed during May 31th to June 13th, 2016. Total 70 topsoil (0–20 cm) samples were collected (Fig. S1), covering the main industrial, mining, and agricultural zones and main traffic lines of the study area. The samples were *in situ* homogenized and stored in the sample bags until back to the laboratory. The soil samples were air dried at the room temperature, and then passed through 0.074 mm sieve for chemical

analysis.

Soil pH was determined with the supernatants of water-soil ratio of 2.5:1 using a pH meter (Shanghai INESA Scientific Instrument Co., China). Soil total organic carbon (TOC) was measured by a multi N/C 3100 analyzer (Analytik Jena AG, Germany). Microwave-digested soil samples were analyzed by an Agilent7900 inductively coupled plasma mass spectrometry (ICP-MS, Agilent Inc, USA). Concentrations of 12 typical heavy metals including vanadium (V), chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), molybdenum (Mo), cadmium (Cd), tin (Sn), antimony (Sb), mercury (Hg), and lead (Pb) were determined.

2.2. Evaluation on soil heavy metal pollution

Four methods including I_{geo} , EF, mC_d , and PLI were adopted to evaluate the soil heavy metal pollution. Proposed by Müller (1969) and defined as the following, I_{geo} generally exhibits the pollution intensity of individual heavy metal.

$$I_{geo} = \log_2 \frac{C_x^i}{1.5 \times C_b^i}$$

where C_x^i and C_b^i refer to the concentration of the *i*th heavy metal in the soil sample and its background concentration in soil that referred to MEPC (1990), respectively.

EF is mainly used to quantify anthropogenic influences on heavy metal pollution (Chester and Stoner, 1973; Clark et al., 2014; Zhu et al., 2017), defined as the following:

$$EF = \frac{\left(\frac{C_x}{R_{soil}}\right)}{\left(\frac{C_b}{R_b}\right)}$$

where R_{soil} and R_b represent the reference element concentration in soil sample and background soil, respectively. Elements Ti, Al, Fe, Mn, Sc or Ca can generally serve as acceptable EF reference element (Salmanighabeshi et al., 2015). This study used Ti as reference element considering that its contents in soil were relatively high and determined accurately by ICP-MS.

PLI and mC_d are comprehensive indexes to characterize the pollution degree by all target heavy metals. mC_d is defined by the following equation (Abraham and Parker, 2008; Wu et al., 2016a).

$$mC_d = \frac{\sum_{i=1}^n \frac{C_x^i}{C_b^i}}{n}$$

where *n* refers to the number of pollutants.

PLI is determined as follows (Bhuiyan et al., 2010; Ćujić et al., 2016; Tian et al., 2017):

$$PLI = \left(\frac{C_x^1}{C_b^1} \times \frac{C_x^2}{C_b^2} \times \dots \times \frac{C_x^n}{C_b^n} \right)^{\frac{1}{n}}$$

2.3. Ecological risks of soil heavy metals

Potential ecological risk index (PERI) is a factor to comprehensively assess the potential ecological risks posed by heavy metals in soil/sediment (Hakanson, 1980; Ke et al., 2017; Madiseh et al., 2009). PERI is calculated using the following equation:

$$PERI = \sum_{i=1}^n T_x^i \times \frac{C_x^i}{C_b^i}$$

where T_x^i means the biological toxicity factor of an individual heavy metal, which is determined as Cr = 2, Ni = Cu = Pb = 5, Zn = 1, and Hg = 40 (Ke et al., 2017; Hakanson, 1980; Tian et al., 2017). T_x^i values for V, Mo, Sn, and Sb were determined according to Hakanson (1980).

2.4. Health risks of soil heavy metals

Health risks of heavy metals in soil of the northeastern Qinghai-Tibet Plateau were evaluated using cancer risks and non-cancer risks through three pathways including ingestion, dermal contact, and inhalation of soil particles (Ferreira-Baptista and De Miguel, 2005; Jiang et al., 2017; Peng et al., 2017; USEPA, 2016; Wu et al., 2016b). Non-cancer risks were evaluated using hazard quotients.

The cancer risks and hazard quotients of the pollutants through ingestion, dermal contact, and inhalation of soil particles are calculated using the following equations (Jiang et al., 2017; Peng et al., 2017; Rovira et al., 2010; Sultana et al., 2014; USEPA, 2016):

$$CR_{ingest} = \frac{C_{soil} \times IngR \times EF_e \times ED_e}{BW \times AT} \times CF \times SF$$

$$HQ_{ingest} = \frac{C_{soil} \times IngR \times EF_e \times ED_e}{BW \times AT \times RfD_{ing}} \times CF$$

$$CR_{dermal} = \frac{C_{soil} \times SA \times AF_{soil} \times ABS \times EF_e \times ED_e}{BW \times AT} \times CF \times SF \times GIABS$$

$$HQ_{dermal} = \frac{C_{soil} \times SA \times AF_{soil} \times ABS \times EF_e \times ED_e}{BW \times AT \times RfD_{der}} \times CF \times GIABS$$

$$CR_{inhal} = \frac{C_{soil} \times InhR \times EF_e \times ED_e}{PET \times BW \times AT} \times SF$$

$$HQ_{inhal} = \frac{C_{soil} \times InhR \times EF_e \times ED_e}{PET \times BW \times AT \times RfD_{inh}}$$

$$CR = CR_{ingest} + CR_{dermal} + CR_{inhal}$$

$$HQ = HQ_{ingest} + HQ_{dermal} + HQ_{inhal}$$

where $CR_{ingest}/CR_{dermal}/CR_{inhal}$ refers to cancer risk via accidental ingestion of soil/dermal contact of soil/inhalation of soil; $HQ_{ingest}/HQ_{dermal}/HQ_{inhal}$ refers to hazard quotient via accidental ingestion of soil/dermal contact of soil/inhalation of soil; C_{soil} refers to the concentration of pollutant in soil (mg/kg); $IngR$ is the ingestion rate of soil (values referred to USEPA, 2016); EF_e is soil exposure

frequency (350 d/yr based on USEPA, 2016); ED_e is exposure duration (6 years for children and 26 years for adults based on USEPA, 2016); CF is the average conversion factor (1×10^{-6} kg/mg based on USEPA, 2016); BW refers to body weight (kg, children: $BW = 15$, adults: $BW = 60$); AT is average time (values for children and adults referred to USEPA, 2016); SA refers to surface area of the skin (values for children and adults referred to USEPA, 2016); AF_{soil} refers to the skin adherence factor for soil (0.2 mg/cm^2); ABS is the dermal absorption factor (chemical specific, obtained from USEPA, 2016); $GIABS$ is the fraction of pollutant absorbed in gastrointestinal tract (set at 1 based on USEPA, 2016); PEF is the particle emission factor ($1.36 \times 10^9 \text{ m}^3/\text{kg}$); SF refers to cancer slope factor ($\text{kg} \cdot \text{d}/\text{mg}$); RfD_{ing} , RfD_{der} , and RfD_{inh} is the corresponding reference dose through ingestion, dermal contact, and inhalation ($\text{mg}/\text{kg}/\text{d}$, obtained from Ferreira-Baptista and De Miguel, 2005; Jiang et al., 2017; Peng et al., 2017; USEPA, 2016); CR and HQ are total cancer risk and total hazard quotient posed by heavy metals in soil; $InhR = 20 \text{ m}^3/\text{d}$ for adults and $7.6 \text{ m}^3/\text{d}$ for children (Ferreira-Baptista and De Miguel, 2005; USEPA, 2016).

2.5. Positive matrix factorization model

Recommended by U.S. Environmental Protection Agency (USEPA, 2014), positive matrix factorization (PMF) has shown good ability of general source apportionment (Jiang et al., 2017; Peng et al., 2017). This study also employed PMF to analyze the possible sources influencing heavy metals in soil. The theory of PMF and input requirements including the determination of uncertainty data refer to guideline of USEPA (2014).

3. Results and discussion

3.1. Soil heavy metal pollution

The major type of soil samples in the study area was sandy clay loam based on soil texture analysis (data not shown). Soils of the study area were alkaline, with pH ranging from 7.39 to 10.02 (Table 1). Soil TOC exhibited significant spatial variety, ranging from 0.42 to 76.93 g/kg with average value of 10.07 g/kg.

Heavy metals in soil of the study area showed spatial and element-specific variety (Table 1). The average values of Hg and Cd were 0.28 and 0.68 mg/kg while their maximal concentrations reached 0.80 and 14.84 mg/kg, respectively. The maximal concentrations of Hg and Cd in soil exceeded over 40 and 108 times of their background values. Pb possessed the highest concentrations among all heavy metals with the value of 2076.28 mg/kg. Zn, Cr, and V illustrated relatively high average concentrations with values of 145.64, 93.29, and 83.10 mg/kg, respectively. Soil quality of about 8.57% (target metal: Cd)–87.14% (target metal: Ni) of sampling sites was evaluated as Level I based on Environmental Quality Standard For Soils of China (GB15618-1995) that only listed the classification criterion for Cr, Ni, Cu, Zn, Cd, As (not measured by this study), Hg, and Pb. Accordingly, soil quality of about 10.00% (target metal: Ni)–72.86% (target metal: Cd) of sampling sites was evaluated as Level II

Table 1

Statistical summary of pH, TOC, and heavy metal concentrations of soil samples (n = 70, g/kg for TOC and mg/kg for all heavy metals).

	V	Cr	Co	Ni	Cu	Zn	Mo	Cd	Sn	Sb	Hg	Pb	TOC	pH
Mean	83.10	93.29	11.59	54.73	40.74	145.64	1.97	0.68	7.26	2.35	0.28	72.49	10.07	8.33
SE	2.00	15.17	0.91	18.54	3.84	18.46	0.57	0.22	0.14	0.20	0.02	30.53	1.85	0.06
Minimum	46.54	42.42	5.48	14.53	17.07	70.13	0.48	0.11	5.00	0.97	0.07	8.63	0.42	7.39
Median	80.23	69.52	10.18	26.76	30.35	104.98	1.14	0.29	7.17	1.93	0.19	31.07	4.59	8.27
Maximum	136.33	913.07	56.00	1015.63	253.29	1002.39	40.68	14.84	12.35	10.25	0.80	2076.68	76.93	10.02

Note: TOC means total organic carbon; SE means standard error.

Table 2Statistical summary of I_{geo} and EF of soil heavy metals and percentage of classification ($n = 70$).

I_{geo}	V	Cr	Co	Ni	Cu	Zn	Mo	Cd	Sn	Sb	Hg	Pb
Mean	−0.40	−0.48	−0.53	−0.54	0.07	−0.01	−0.01	0.83	1.26	−0.08	2.87	0.07
SE	0.03	0.08	0.07	0.11	0.09	0.09	0.10	0.14	0.03	0.07	0.12	0.14
minimum	−1.21	−1.31	−1.47	−1.61	−0.96	−0.78	−1.49	−0.90	0.74	−1.19	1.19	−1.86
median	−0.42	−0.60	−0.57	−0.73	−0.13	−0.20	−0.25	0.48	1.26	−0.19	2.68	−0.01
maximum	0.34	3.12	1.89	4.52	2.93	3.06	4.91	6.17	2.04	2.22	4.74	6.05
I_{geo} class I	94.3%	94.3%	91.4%	92.9%	60.0%	70.0%	61.4%	11.4%	/	70.0%	/	52.9%
I_{geo} class II	5.7%	2.9%	5.7%	4.3%	22.9%	22.9%	31.4%	62.9%	8.6%	21.4%	/	38.6%
I_{geo} class III	/	/	2.9%	/	15.7%	2.9%	5.7%	10.0%	90.0%	5.7%	20.0%	4.3%
I_{geo} class IV	/	1.4%	/	/	1.4%	2.9%	/	11.4%	1.4%	2.9%	42.9%	1.4%
I_{geo} class V	/	1.4%	/	/	/	1.4%	/	1.4%	/	/	15.7%	/
I_{geo} class VI	/	/	/	2.9%	/	/	1.4%	1.4%	/	/	21.4%	1.4%
I_{geo} class VII	/	/	/	/	/	/	/	1.4%	/	/	/	1.4%
EF	V	Cr	Co	Ni	Cu	Zn	Mo	Cd	Sn	Sb	Hg	Pb
Mean	0.96	1.25	1.00	1.97	1.79	1.63	2.22	3.92	3.07	1.37	11.16	2.52
SE	0.02	0.29	0.12	0.82	0.30	0.29	0.83	1.28	0.09	0.13	0.89	0.90
minimum	0.63	0.62	0.43	0.36	0.64	0.71	0.41	0.80	1.64	0.45	2.90	0.42
median	0.93	0.81	0.81	0.77	1.07	1.05	1.03	1.78	2.93	1.04	8.20	1.18
maximum	1.45	15.71	6.69	41.94	20.04	19.07	58.61	88.84	5.58	7.17	32.20	59.23
EF class I	100.0%	97.1%	97.1%	97.1%	81.4%	90.0%	84.3%	60.0%	1.4%	88.6%	/	90.0%
EF class II	/	/	/	/	12.9%	4.3%	11.4%	27.1%	95.7%	8.6%	15.7%	5.7%
EF class III	/	2.9%	2.9%	/	4.3%	5.7%	2.9%	10.0%	2.9%	2.9%	70.0%	1.4%
EF class IV	/	/	/	/	1.4%	/	/	1.4%	/	/	14.3%	1.4%
EF class V	/	/	/	2.9%	/	/	1.4%	1.4%	/	/	/	1.4%

Note: I_{geo} and EF refer to index of geo-accumulation and enrichment factor; SE means standard error; I_{geo} classes I, II, III, IV, V, VI, and VII refer to uncontaminated, uncontaminated to moderately contaminated, moderately contaminated, moderately contaminated to heavily contaminated, heavily contaminated, heavily contaminated to extremely contaminated, and extremely contaminated, respectively; EF classes I, II, III, IV, and V refer to deficiency to minimal enrichment, moderate enrichment, significant enrichment, very high enrichment, and extremely high enrichment.

while that of about 0.00% (target metal: Hg)–18.57% (target metal: Cd) of sampling sites was evaluated as Level III or worse.

Heavy metal pollution was evaluated by four methods (Table 2 and Fig. 1). Average I_{geo} values ranged from −0.54 (Ni) to 2.87

(Hg) while the minimal and maximal I_{geo} values were −1.86 (Pb) and 6.17 (Cd), respectively (Table 1). Based on classification criterion (Ćujić et al., 2016; Tian et al., 2016; Wen et al., 2017; Zhang et al., 2017), heavy metals including V, Cr, Co, and Ni showed the

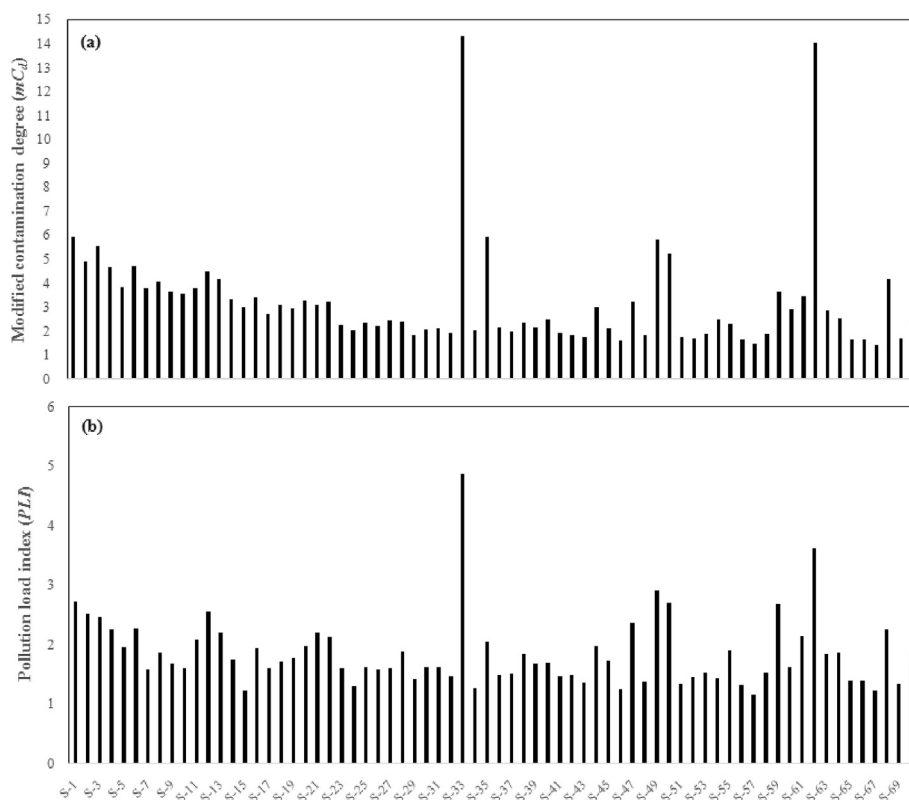


Fig. 1. Modified contamination degree (mCd) and pollution load index (PLI) of soil heavy metals in the sampling sites.

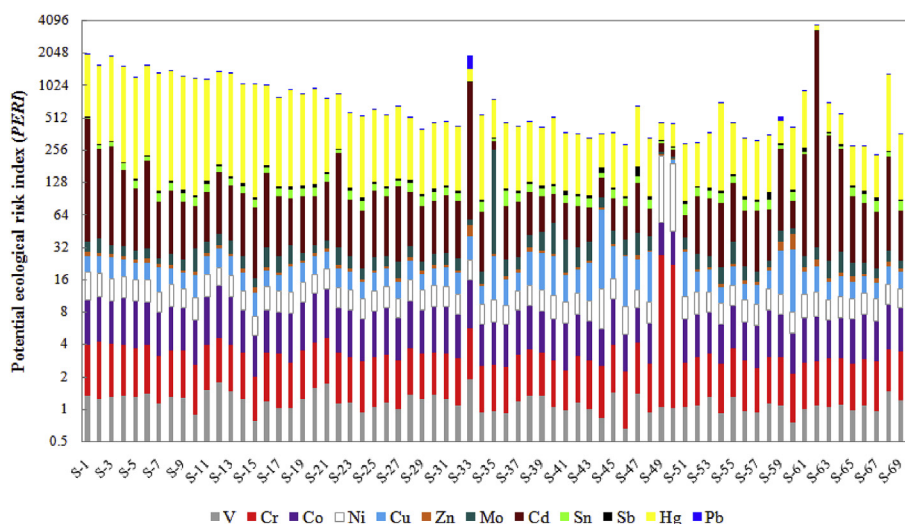


Fig. 2. Potential ecological risks of soil heavy metals in the sampling sites.

similar contamination degree that over 90% of samples were classified into uncontaminated status while Cu, Zn, Mo, and Sb exhibited the similar patterns with classification of uncontamination/uncontamination to moderate contamination percentages of 60.0%/22.9%, 70.0%/22.9%, 61.4%/31.4%, and 70.0%/21.4%, respectively. Interestingly, Cd, Sn, Hg, and Pb showed different contamination patterns. Sn mainly posed moderate contamination

while Cd exerted uncontamination to moderate contamination, moderate contamination, and moderate contamination to heavy contamination in soils with percentages of 62.9%, 10.0%, and 11.4%, respectively. Pb in soils mainly showed uncontamination (52.9%) and uncontamination to moderate contamination (38.6%). Hg in soils illustrated the most serious contamination among all heavy metals with percentages of 20.0%, 42.9%, 15.7%, and 21.4% for

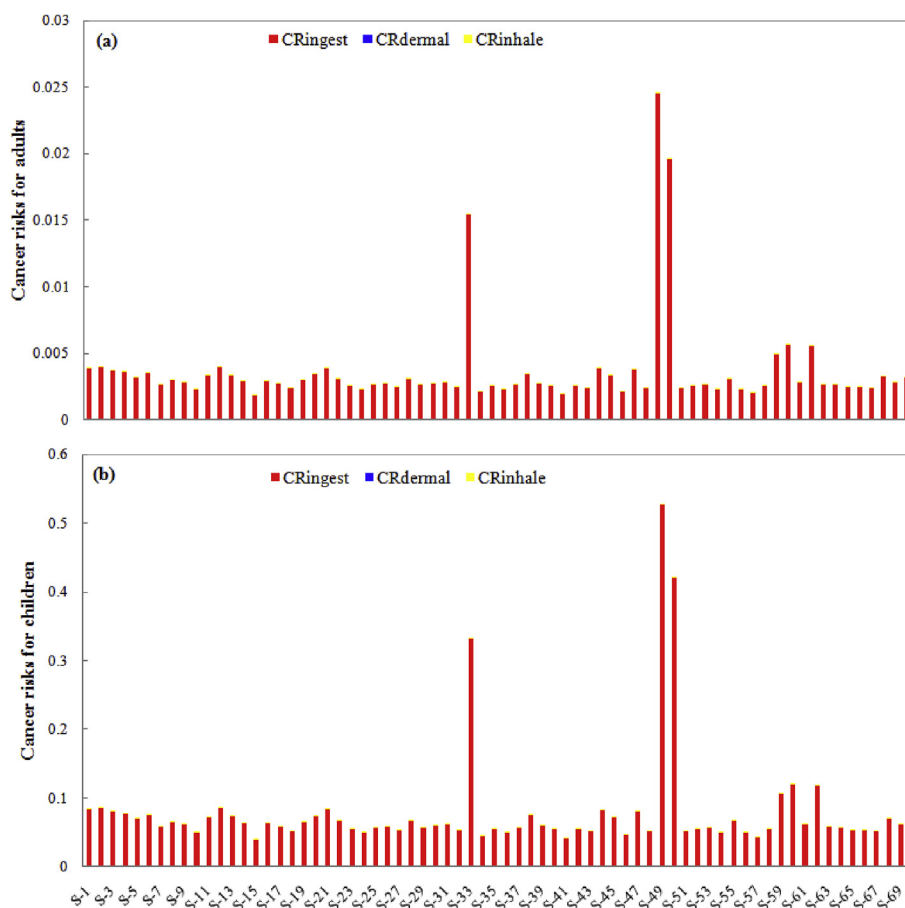


Fig. 3. Cancer risks of soil heavy metals in the sampling sites.

moderate contamination, moderate contamination to heavy contamination, heavy contamination, and heavy contamination to extreme contamination.

Soil EF values ranged from 0.36 (Ni) to 88.84 (Cd) while average values were in the range of 0.96 (V) to 11.16 (Hg), illustrating significant spatial variety (Table 1). V, Cr, Co, Ni, Cu, Zn, Mo, Sb, and Pb in soils showed the slight enrichment while Sn in soils illustrated moderate enrichment based on criterion (Ćujić et al., 2016; Wen et al., 2017). Cd in soils exhibited different enrichment patterns with 60.0% of minimal enrichment, 27.1% of moderate enrichment, and 10.0% of significant enrichment while Hg in soils illustrated 15.7% of moderate enrichment, 70.0% of significant enrichment, and 14.3% of very high enrichment, showing significant enrichment feature. Interestingly, Cd in soil sample collected from S-62 showed the highest I_{Geo} and EF values while several metals such as Sn and Hg did not exhibit similar pattern. For example, Sn in soil sample from S-36 and Hg in sample from S-3 possessed the highest I_{Geo} values of 2.04 and 4.74 while Sn in sample from S-60 and Hg in sample from S-15 showed the maximal EF values of 5.58 and 32.20, respectively. These results suggested that soil pollution by individual heavy metal of the study area should be caused by multiple factors besides metal enrichment degree.

Contamination degrees of heavy metals in soil were unexpectedly high (Fig. 1a). Only 2 and 16 sampling sites were in the status of nil to very low contamination degree (when $mC_d < 1.5$ according to

Abraham and Parker, 2008) and low contamination degree (when $1.5 \leq mC_d < 2$), respectively. The numbers of sampling sites were respectively 12 and 2 for high degree of contamination (when $4 \leq mC_d < 8$) and very high contamination degree ($8 \leq mC_d < 16$). The remaining 38 sites showed moderate degree of contamination (when $2 \leq mC_d < 4$). Hg, Cd, and Sn contributed to main mC_d in many sites while Pb, Mo, and Zn were the dominant contributors of mC_d in several sites.

Based on PLI evaluation criterion (Ćujić et al., 2016; Tian et al., 2017), 51 sampling sites showed moderate contamination degree (when $1 < PLI \leq 2$) while 19 sites were in the high contamination status (when $2 < PLI \leq 5$) (Fig. 1b). Compared with mC_d , PLI evaluation results narrowed the pollution range by weakening the extreme results. Generally, the study area is experiencing provoking heavy metal pollution according to both mC_d and PLI evaluation results, more serious than the previous studies (Wu et al., 2016a).

3.2. Ecological risks of heavy metals in soil

Ecological risk indexes of heavy metals in soil ranged from 234.6 to 3759.0 (Fig. 2). It is surprising that all sampling sites possessed relatively high ecological risks based on the classification criterion (Madiseh et al., 2009; Ke et al., 2017). About 6 out of 70 sampling sites exhibited moderate ecological risks (when $150 \leq PERI < 300$) while half of the remaining sites showed considerable ecological

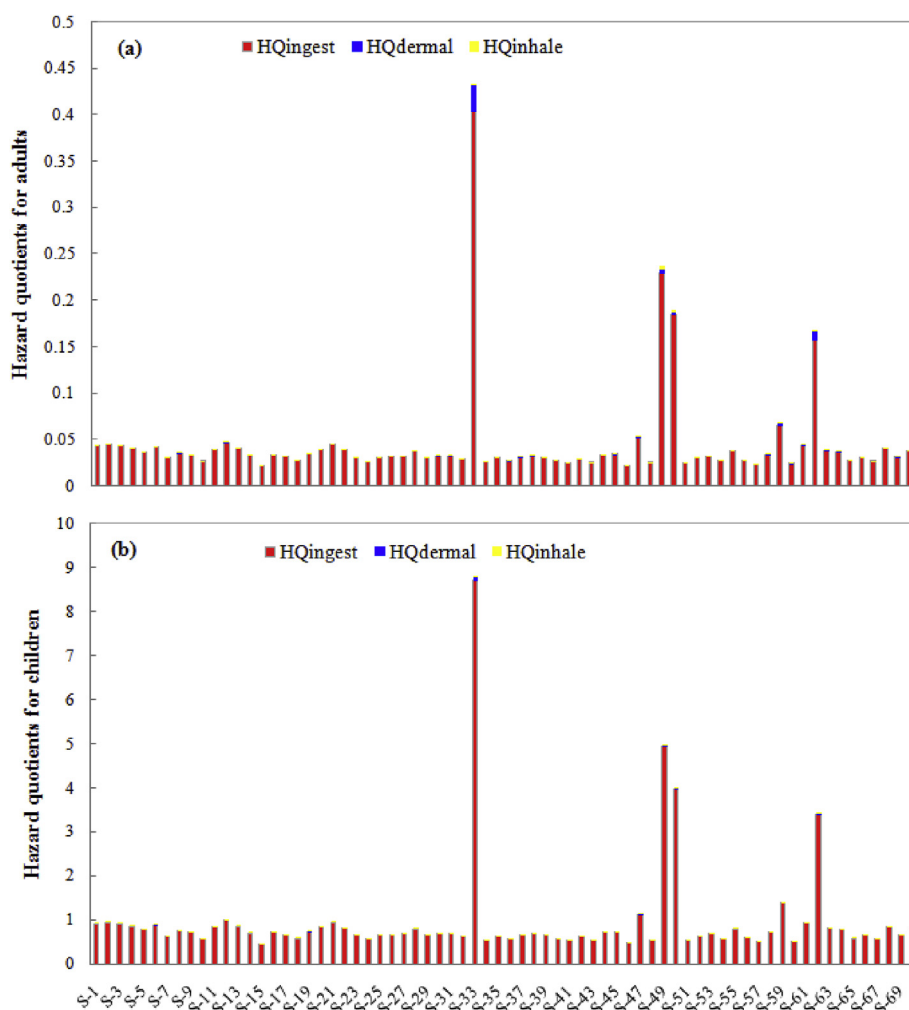


Fig. 4. Non-cancer risks of soil heavy metals in the sampling sites.

risks (when $300 \leq PERI < 600$) and the other 32 sites possessed very high ecological risks (when $PERI \geq 600$). Serving as the dominant ecological-risk factor, Hg contributed over 50% ecological risks to 64 sampling sites with contribution percentage ranging from 50.7% to 91.6%. Ecological risks deriving from Hg ranged from 136.57 to 1602.95. Cd was the second dominant ecological-risk factor with the maximal contribution percentage of 86.4%. The highest $PERI$ contribution percentages of V, Co, Sb, Sn, and Cr ranged from 0.41% (V) to 7.76% (Sn). Ecological risks deriving from Sn ranged from 12.50 to 30.88. For several sites, contributions of Ni, Cu, Mo, and Pb were also high with the maximal contribution percentages of 36.7%, 15.7%, 29.6%, and 25.8%, respectively. Ecological risks exerted by heavy metals in soil reached considerably high level in the study area to deserve urgent and effective pollution control.

3.3. Health risks posed by heavy metals in soil

Cancer risks (CR) of heavy metals for adults and children were in the ranges of 1.91×10^{-3} – 2.46×10^{-2} and 4.09×10^{-2} – 5.29×10^{-1} , respectively (Fig. 3). Cancer risks for children were generally over 20 times higher than those for adults, suggesting that children are much more susceptible to adverse effects of soil heavy metals. The cancer risks through pathways of ingestion, dermal contact, and inhalation followed the order of $CR_{inhalation} < CR_{dermal} < CR_{ingestion}$. Risks of heavy metals through ingestion accounted for over 99.1% of cancer risks while risks through dermal contact were usually over 20 times higher than those through inhalation. Cr was the dominant contributor for the cancer risks through ingestion, with the highest contribution percentages of 92.7% for both adults and

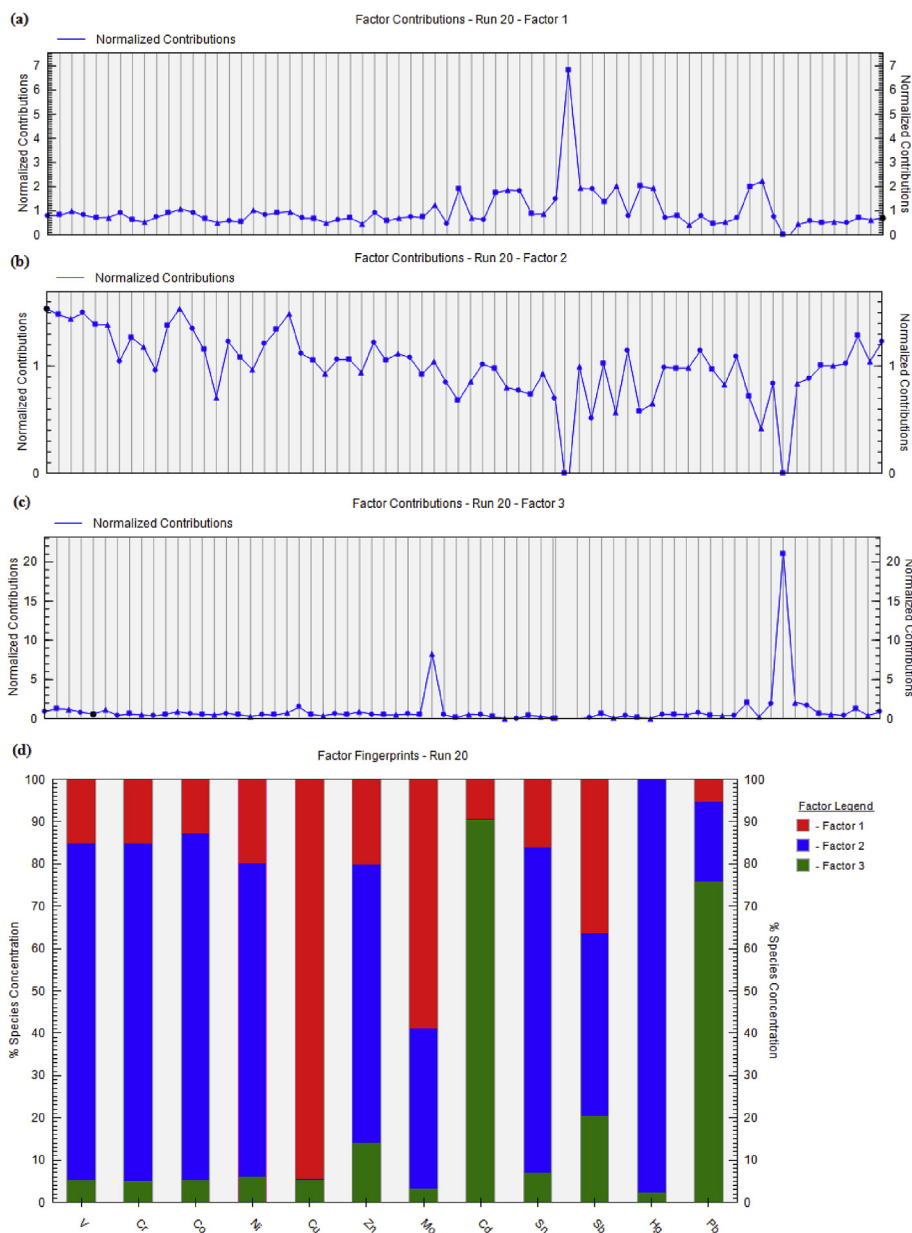


Fig. 5. Factor normalized contributions and factor fingerprint.

children (Fig. S2). Zn, V, Pb, Cu, and Co also contributed to cancer risks through ingestion to relatively significant extent. Zn served as the main contributor for the cancer risks through dermal contact, and then followed by Pb and Cu. Cr and Zn mainly accounted for the cancer risks through inhalation, similar with those through ingestion. Based on the screening criterion (Ge et al., 2013), cancer risks for adults of all sites were evaluated as high (when $1 \times 10^{-3} < CR \leq 1 \times 10^{-1}$). Correspondingly, there were 6 and 64 sites to illustrate very high and high cancer risks for children, respectively.

The total hazard quotients of soil heavy metals for adults and children were in the ranges of 0.023–0.434 and 0.466–8.815 with the average value of 0.048 and 0.987, respectively (Fig. 4). Hazard quotients for adults were less than threshold of 1.0, suggesting the non-cancer risks for adults were low. However, hazard quotients for children in 68 sites exceeded 0.500 and those in 7 sites exceeded 1.0 with maximal value of 8.815, suggesting the high non-cancer risks for children in the study area. Non-cancer risks through ingestion accounted for over 99.2% and 98.5% of the total risks for adults and children, respectively (Fig. S3). Non-cancer risks through dermal contact in most of sites were 2–6 times higher than those through inhalation with the maximal ratio over 38 and 40 for adults and children. Total hazard quotients for children were 20 times higher than those for adults, also suggesting that children are more susceptible to potential hurt from heavy metals in soil. Cr was the dominant contributor for the non-cancer risks through ingestion, and then followed by Pb, V, Ni, Sb, and Hg also contributed to non-cancer risks through ingestion. Pb, V, Cr, Sb, and Hg served as the main contributors for the non-cancer risks through dermal contact. Different from cancer risks, Cr and Co accounted for over 92.8% of non-cancer risks through inhalation. The difference in dominant contribution heavy metals for cancer and non-cancer risks through 3 pathways exhibited the different toxicity mechanism of heavy metals.

3.4. Possible sources of heavy metals in soil

After screening by PMF model, 3 factors with corresponding contributions were determined for the possible sources of heavy metals in soil (Fig. 5). Comparing the dominant heavy metals of each factor and the background information of the study area, the possible sources were determined as industry (factor 2), mining (factor 1), and transport and agriculture (factor 3). Potential ecological and health risks of 3 factors were evaluated, respectively (Figs. S4–S6). *PERI* values of factor 1, factor 2, and factor 3 were in the ranges of 16.7–344.9, 161.8–1607.6, 32.9–3077.2, respectively, with the order of factor 1 < factor 3 < factor 2 for most of the sites (Fig. S4). Cr was the dominant contributors for *PERI* of factor 1 and 3 while Hg was the dominant contributor for that of factor 2. Cancer risks of factor 1, factor 2, and factor 3 for adults/children were in the ranges of 3.65×10^{-4} – 3.88×10^{-3} / 7.83×10^{-3} – 8.35×10^{-2} , 1.34×10^{-3} – 1.93×10^{-2} / 2.88×10^{-2} – 4.16×10^{-1} , and 1.77×10^{-4} – 6.68×10^{-3} / 3.79×10^{-3} – 1.43×10^{-1} , respectively (Fig. S5). Cancer risks of three factors followed the order of factor 3 < factor 1 < factor 2. Hazard quotients of factor 1, factor 2, and factor 3 for adults/children were in the ranges of 3.53×10^{-3} – 3.88×10^{-2} / 7.33×10^{-2} – 8.13×10^{-1} , 1.42×10^{-2} – 1.82×10^{-1} / 2.95×10^{-1} – 3.82 , and 3.31×10^{-3} – 2.95×10^{-1} / 6.79×10^{-2} – 5.97 , respectively (Fig. S6). Similar with *PERI*, non-cancer risks of three factors followed the order of factor 1 < factor 3 < factor 2 for most of the sites, suggesting the difference in the toxicity mechanisms of heavy metals for cancer and non-cancer risks.

4. Conclusions

Heavy metal pollution of soils in the study area was serious. Pb possessed the highest concentrations while the maximal concentrations of Hg and Cd in soil were over 40 times higher than their background values. Hg caused the most serious pollution according to I_{geo} results and also showed the strong enrichment features. Based on mC_d evaluation results, about 54.3%, 17.1%, and 2.9% of sampling sites showed moderate, high, and very high contamination degree, respectively. Accordingly, pollution load index results illustrated 72.9% and 27.1% of sampling sites possessed moderate and high contamination level, respectively. Consequently, ecological risk indexes posed by heavy metals in soil ranged from 234.6 to 3759.0, suggesting that most of sites were under considerable/very high risks. Cancer risks/hazard quotients of heavy metals for adults and children were in the ranges of 1.91×10^{-3} – 2.46×10^{-2} / 0.023 – 0.434 and 4.09×10^{-2} – 5.29×10^{-1} / 0.466 – 8.815 , respectively. Cancer risks for adults and children were determined as high and high-very high levels while non-cancer risks for children were high although those for adults seemed low. Three possible sources were determined by PMF model and industrial source contributed to main part of ecological and health risks. In summary, the northeastern Qinghai-Tibet Plateau has endured the serious heavy metal pollution to exert high potential ecological and health risks, especially for children. Therefore, more efficient and strict pollution control and management should be put out as soon as possible.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.chemosphere.2018.02.122>.

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