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Sources, Enrichment, and Geochemical Fractions of Soil Trace Metals in Ulaanbaatar, Mongolia

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Abstract

Mongolia is a rapidly developing country that has experienced growing industrialization and urbanization in recent decades. This study was conducted to evaluate the enrichment and labile fractions of metals in urban soils of Mongolia and to identify major sources of soil metal pollution. The concentrations and geochemical fractions of Al, Fe, Mn, Cr, Cu, Cd, Co, Zn, V, Mo, As, Sb, and Pb in soils of the city Ulaanbaatar were investigated. The results demonstrate that only Fe, Mn, Co, Mo, and V occur at natural levels with enrichment factors close to unity. The majority of investigated toxic metals, including Cu, Zn, Cr, Sb, As, Cd, and Pb, are serious pollutants in urban soils, with enrichment factors of up to 2.8, 5.1, 2.1, 16, 13, 15, and 11, respectively. Studies of the chemical fractions of metals demonstrate that Zn is mainly found in its labile form and is considered a high risk to humans and biota. Industrial release, household ash, coal combustion, and tire abrasion were identified as key sources of toxic metals entering into the soil of Ulaanbaatar City, which should be controlled effectively to prevent the population as well as pollution distribution over a wider area by long-range atmospheric transport.

Soil metal pollution is one of the most important environmental issues (Abbas et al. 2017; Lu et al. 2015). When metals in soils are released to water and plants and consequently enter the food chain, the problem is a real threat (Chowdhury et al. 2016; Lu et al. 2015). In many developing countries experiencing rapid industrialization and urbanization, a large amount of toxic metals are released into the environment without proper monitoring and regulation and consequently accumulate in nearby soils by dry and wet deposition (Abbas et al. 2017; Chowdhury et al. 2016; Islam et al. 2015; Naidansuren et al. 2017). Determination of the total concentration of a metal does not provide sufficient information about the characteristics that influence its toxicity, such as its ability of transferring in the environment and/or potential environmental risks (Cuvier et al. 2016; Ke et al. 2017). The bioavailability of metals depends highly on their chemical fractions in soil (Ke et al. 2017). Therefore, wider investigation of the total contents and geochemical fractions of metals is the best method for the assessment of pollution and risks from metals in soil (Cuvier et al. 2016; Ke et al. 2017).

Mongolia is rapidly developing within the past decades (Batsaikhan et al. 2018). It has a population of about only 3.3 million people but is the 18th-largest country in the world covering 1.5 million square kilometers area, which makes it one of the lowest populated countries in the world (NSOM 2018). Because Mongolia is also one of the most important sources of Asian dust, environmental quality and pollution have great importance to worldwide (Guttikunda et al. 2013; Kim et al. 2016; Nishikawa et al. 2011). In recent years, East Asia is affected by the meteorological phenomenon as called Asian dust-so called yellow dust. The air quality over the ocean obstructed by these dusts (fine, dry soil particles) originated from the deserts of China, Mongolia, and Kazakhstan (Uno et al. 2009). Rapid industrialization along with new technological development of source areas has led to the emission of various contaminants assimilated in dust particles, resulting in

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substantial health problems (Chen et al. 2018). In addition, deforestation and global warming have enhanced desertification, which results in longer and more frequent occurrences of Asian dust (Zhou et al. 2019). After the socioeconomic transition of the country in the 1990s, rapid urbanization and industrialization occurred in Mongolia (Sato 2012; Soyol-Erdene et al. 2019) and caused the growth of industry, population, and number of vehicles (Sato 2012). Consequently, significant degradation and pollution in the environment (Batsaikhan et al. 2018; Chung and Chon 2014; Naidansuren et al. 2017; World Bank 2008) have occurred. How it seriously affects the quality of the environment and shows a risk to human health and metal pollution both in urban and rural areas has become a critical environmental issue in Mongolia (Battogtokh et al. 2014; Chung and Chon 2014; Guttikunda et al. 2013; Pfeiffer et al. 2015).

We report the systematic investigation on the sources, enrichment, and geochemical fractions of metals (Al, Fe, Mn, Cr, Cu, Cd, Co, Zn, V, Mo, As, Sb, and Pb) in urban soil with an example of Ulaanbaatar City, Mongolia. Our investigation focused on (1) the distribution and geochemical fractions of metals in soil from distinctive functional areas; (2) the pollution levels and ecological risks of these soils using the crustal enrichment factor (EFc, for total content) and risk assessment code (RAC, for chemical fractions); and (3) the major sources of soil metal pollution by principal component analysis (PCA). The outcomes of this study can be a reference for other developing countries with similar situations due to the populated metropolitan setting in the study region and its socioeconomic and environmental conditions. Additionally, the results of this study will be beneficial to explore the current impact and as well as predicting future contributions from soil contamination on global atmospheric quality through Asian dust.

Materials and Methods

Study Area and Sampling

Ulaanbaatar (UB) is the capital of Mongolia and is the largest city and the most important commercial and industrial center (Fig. 1) (Soyol-Erdene et al. 2019). The city has a population of around 1.3 million, which is almost half of the country's total population of 3.3 million (NSOM 2018). The rapid increase in population and the high urbanization rate have made it the most densely populated area in the country (Batsaikhan et al. 2018). The study area, UB city, typically has a mid-latitude steppe climate with an average temperature of −0.1 °C, ranging from −23.0 °C in January to +20.0 °C in July and an average annual rainfall of 257.7 mm (Sato 2012). Neogene and Quaternary red to yellow clays, sands, and gravels are widely distributed near the city (Dalai and Ishiga 2013). The soil has high water permeability because of the soil type (loamy sandy and sandy loamy), with low humus content (0.5–1.5%) and 0.7–5.0% $CaCO_3$ with a pH of 7.5–9.0 (Kasimov et al. 2011). In the UB city, soils are strongly exposed to anthropogenic impact; from the central part of the city toward the ger areas (Kasimov et al. 2011). In addition to the high density of residents and pollution sources in UB, very few studies have been performed on soil pollution by metals or organic pollutants (Batjargal et al. 2010; Chung and Chon 2014; Kasimov

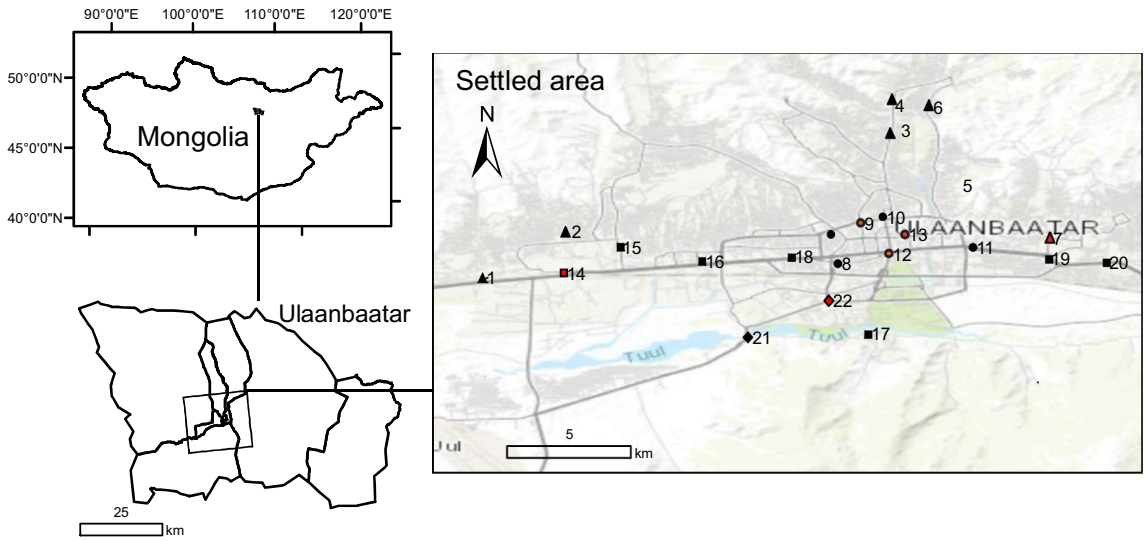

Fig. 1 Study area location map. Sample locations noted as triangles (ger area), circles (downtown), squares (suburban), and diamonds (industrial). Sequentially extracted samples are in red

et al. 2011). Previous studies found a low degree of metal pollution in UB soils. According to one of previous works (Batjargal et al. 2010), there was no significant increase in the concentrations of metals (As, Cd, Cr, Cu, Ni, and Zn) in surface soil compared with those in subsoil, and the leaching potential of metals was found to be quite low through sequential extraction. Another work also evaluated soil metal pollution and reported that soils of the city are assessed as weakly polluted with no significant changes from the state of the soil cover in the 1990s, previously (Kasimov et al. 2011). The authors used pollution indexes for evaluation based on concentrations of nine elements (Pb, Zn, Mo, Cr, Cu, Cd, Ni, Co, Sr, As, and V) by comparing with those in a rural site (Kasimov et al. 2011). More recently, moderate pollution of mercury (Hg) in soils of Ulaanbaatar was found and considered to be mainly related to coal combustion (Chung and Chon 2014), which has possibly resulted from population migration during the past two decades to the low-income ger area of the city where people burn raw coal to produce heat.

We collected surface soil samples in September 2015 from different functional zones (Fig. 1) of the city, i.e., downtown, industrial, suburban settlement, and ger (traditional dwellings) areas. Samples were taken from the uppermost layer of the soil profile (0–10 cm) using a plastic tool and a polyethylene bag. The composite sample collected by five subsamples from the same site. A total of 22 topsoil samples were collected and were then air-dried. All samples passed through a 2-mm sieve and stored in polyethylene bags until further analyses.

Soil Analyses

Organic carbon contents and pH of all soil samples were analyzed using standard procedures. Soil pH was determined in a 1:5 (w/v) suspension of soil and water by a glass electrode. The organic carbon content was analyzed through the oxidation method using by potassium dichromate as oxidant. Metal analyses were performed by inductively coupled plasma mass spectrometry (ICP-MS, NeXION 300Q, Perkin Elmer) and inductively coupled plasma optic emission spectrometry (ICP-OES, Optima 7300DV, Perkin Elmer) at the SGS International Laboratory in Ulaanbaatar, Mongolia. Acid digestion of soil samples was conducted with a combination of concentrated acids, HCl-HF–HNO_3–$HClO_4$ (36%, 48%, 65%, and 60%, respectively), in Teflon tubes on a hot block, which were heated at 220 ± 10 °C for 3–4 h. After drying, the residue was then dissolved with a combination of HCl and HNO_3 for 5 min. Aliquots were then diluted with deionized water for further analysis. Calibration standard solutions were prepared by sequential dilution from 1000 µg mL^{-1} multielement standard solutions (Inorganic ventures, USA). The concentrations of the standard solutions used for the calibration curves were 0, 0.25, 0.5, 1, 2.5, and 5 µg mL^{-1} for Mn, Cr, Cu, Cd, Co, Zn, V, Mo, As, Sb, and Pb quantification and 0, 2, 5, 10, 25, and 50 µg mL^{-1} for Al and Fe quantification. For these concentration ranges, the correlation coefficients (r^2) of the calibration curves were higher than 0.999. To avoid instrumental drift, a Lu internal standard (5 µg mL^{-1}) (Inorganic ventures, USA) was applied. Analyses of the blank solution (1% HNO_3) and standard solution (2 µg mL^{-1}) were repeated for each 15 samples. The internal precision during the analyses was less than 5% for all analyzed metals. The accuracy of analyses was checked by simultaneous analyses of a certified reference material (NIST 2702). The analytical results for the reference material agreed well with certified values within the analytical uncertainty (< 10%) (Table 1).

A subset of six selected soil samples was extracted according to a BCR sequential extraction method (Rauret et al. 2000). Through this BCR extraction method, soil metals are separated into four different fractions based on their chemical forms in soil: acid leachable (labile) (F1, mobile and exchangeable), reducible (F2, Fe–Mn hydroxide), oxidizable (F3, organic-bound), and residual (F4) (Rauret et al. 2000). The acid leachable fraction indicates metal's ability to transfer and bioavailability (Filgueiras et al. 2002). Reducible and oxidizable fractions are more sensitive to environmental changes than residual fraction, and the residual fraction can be considered stable and no significant risk to the ecosystem (Filgueiras et al. 2002). The metal residual fraction originates from natural sources, e.g., rock materials, whereas other fractions usually originate from anthropogenic activity (Filgueiras et al. 2002; Hu and Cheng 2016). All extracts and residual fractions were analyzed for metal (Cr, Cu, Pb, Zn, Co, As, Sb, V) concentrations in the SGS international laboratory according to the procedures as

Table 1 Comparison of analytical results and certified values for NIST2702 reference material

Elements	Certified (mg kg^{-1})	Measured (mg kg^{-1})
Al (%)	8.41 ± 0.22	9.45
As	45.3 ± 1.8	45.3
Cd	0.817 ± 0.011	0.911
Co	27.76 ± 0.58	27.5
Cr	352 ± 22	304
Cu	117.7 ± 5.6	96.8
Fe (%)	7.91 ± 0.24	7.38
Mn	1757 ± 58	1597
Mo	10.8 ± 1.6	10.1
Pb	132.8 ± 1.1	122.8
Sb	5.6 ± 0.24	5.1
V	357.6 ± 9.2	357.9
Zn	485.3 ± 4.2	432.1

described before. The quality of the analytical data for the BCR sequential extraction procedure was assessed by carrying out analyses of the certified reference material (CRM) BCR-701 (LGC Promochem, Barcelona, Spain). Cadmium (Cd) and Mo were excluded from the chemical fraction analyses because of discrepancies between the results and the certified values. The results for the standard material agreed for all three-step extracts and residual fractions within 10% for Zn, Pb, and Cu (Rauret et al. 2001). The concentrations of Cr were in good agreement with the certified values for all three-step extracts but did not match for the residual fraction (Table 2). Several researchers have determined chemical fractions by the BCR extraction method for BCR-701 CRM (Jochum et al. 2005). Reported values varied widely between 49.2 mg kg^{-1} and 110 mg kg^{-1} (Jochum et al. 2005). Our result for the residual fraction of Cr (118.7 ± 12.4 mg kg^{-1}) is close to the higher value of 110.8 mg kg^{-1} in the literature (Ohta 2015). Co, As, and V have no certified values and agreed well with existing literature references (Table 2) (Jochum et al. 2005). There are no previously published values for Sb fractions from BCR-701 CRM.

Table 2 Comparison of sequential extraction results and certified values for BCR 701 reference material (mg kg^{-1})

	Certified	Measured ($n=2$)
Cr[a]		
Exchangeable	2.26 ± 0.16	1.78 ± 0.01
Oxidizable	45.7 ± 2.0	54.5 ± 0.6
Reducible	143 ± 7	135.2 ± 4.8
Residual	63 ± 8	118.7 ± 12.4
Cu[a]		
Exchangeable	49.3 ± 1.7	54.4 ± 4.2
Oxidizable	124 ± 3.0	140.6 ± 1.2
Reducible	55 ± 4	56.5 ± 3.0
Residual	39 ± 12	27.4 ± 3.0
Pb[a]		
Exchangeable	2.72 ± 0.3	2.3 ± 0.3
Oxidizable	126 ± 3.0	128.2 ± 2.6
Reducible	9.3 ± 2.0	10.6 ± 0.6
Residual	11 ± 6	21.7 ± 0.4
Zn[a]		
Exchangeable	205 ± 6	196.0 ± 9.6
Oxidizable	114 ± 5	125.8 ± 3.1
Reducible	46 ± 4.0	55.5 ± 1.6
Residual	95 ± 13	107.4 ± 7.9
Co[b]		
Exchangeable	2.15 ± 0.64	2.10 ± 0.12
Oxidizable		4.14 ± 0.04
Reducible		1.73 ± 0.13
Residual		10.1 ± 0.4
As[b]		
Exchangeable	2.1 ± 0.1	2.4 ± 0.1
Oxidizable	21.2 ± 1.1	20.9 ± 0.5
Reducible		15.0 ± 1.5
Residual		13.0 ± 1.3
Sb[c]		
Exchangeable		0.080 ± 0.005
Oxidizable		0.149 ± 0.004
Reducible		0.143 ± 0.011
Residual		1.938 ± 0.089
V[b]		
Exchangeable	0.399 ± 0.017	0.56 ± 0.01
Oxidizable		12. 9 ± 0.1
Reducible		4.7 ± 0.1
Residual		74.2 ± 0.4

[a]Certified values (Rauret et al. 2001)

[b]No certified value, compiled from Georem (Jochum et al. 2005)

[c]No certified or compiled values

Assessment of Pollution Level and Ecological Risk

We used a crustal enrichment factor (EF) to assess the extent of soil pollution. The enrichment factor is one of the most widely used indicators to assess the presence and intensity of anthropogenic contaminant deposition on surface soil (Chen et al. 2007; Chung and Chon 2014; Cuvier et al. 2016; Lin et al. 2000). EF is calculated by the normalization of metal concentration in the topsoil with respect to the concentration of a reference element and is calculated as follows:

$$EF = (Metal/Al)_{soil}/(Metal/Al)_{UCC}$$

where $(Metal/Al)_{soil}$ and $(Metal/Al)_{UCC}$ are ratios of the concentration of the metal to the concentration of Al in the soil and to the average concentration in the upper continental crust (Wedepohl 1995), respectively. Based on EF values, the extent of soil pollution can be classified as follows: no enrichment (EF < 1), minor enrichment (EF = 1- 3), moderate enrichment (EF = 3–5), moderately severe enrichment (EF = 5–10), severe enrichment (EF = 10–25), very severe enrichment (EF = 25–50), and extremely severe enrichment (EF > 50) (Chen et al. 2007).

The risk assessment code (RAC) has been widely used for evaluating the transferring ability and bioavailability of metals in soils (Liu et al. 2011; Marrugo-Negrete et al. 2017; Rodríguez et al. 2009). The risk assessment code (RAC) is illustrated by the ratio of the content of the acid leachable fraction (exchangeable and carbonate fractions) to the sum of all fractions. Based on RAC values, soil pollution by metals can be described as class I, RAC < 1, no risk; class II, 1 < RAC < 10, light risk; class III, 11 < RAC < 30, moderate risk; class IV, 31 < RAC < 50, high risk; and class V, RAC > 50–75, very high risk (Liu

et al. 2011; Marrugo-Negrete et al. 2017; Rodríguez et al. 2009).

Statistical Analyses

Descriptive and principal components analyses (PCA), the most common multivariate statistical methods for data treatment, were conducted by using SPSS 20 software (SPSS Inc., Chicago, IL) to check variability and significant relationships among the metals in the soil samples. Principal component analysis is often used to divide metals into several categories associated with natural or anthropogenic sources (Marrugo-Negrete et al. 2017; Pan et al. 2017). To facilitate the interpretation of results, a varimax rotation was applied in PCA. The statistical analyses were performed with a 95% confidence interval (significance $p < 0.05$).

Results and Discussion

Physicochemical Properties of Soil Samples

Selected physicochemical parameters (pH and soil organic matter) determined for the urban soils of Ulaanbaatar are shown in Table 3. The organic matter (OM) content ranged from 0.03 to 7.5%, with average being approximately 2.3%. The pH values were in the range of 7.01–8.86, indicating the neutral and weak-alkaline conditions of the samples.

Concentrations of Metals in Soil

The descriptive statistics for heavy metals are shown in Table 3. The concentrations of metals at each site studied were compared with the average concentrations of the upper continental crust (Wedepohl 1995) in Fig. 2. Grain size is one important factor to control metal concentrations; Al normalized concentrations also were illustrated in order to check effect of grain size on spatial distribution of metals. Overall trends for spatial distributions of Al normalized and non-normalized concentrations were similar for all studied metals (Fig. 2).

Aluminum concentrations (Fig. 2) in the topsoil were in the range of 5.10–7.81%, with an average value of 6.54 ± 0.59%. Compared with the variability in the concentration of other metals, less variability with a relative standard deviation of only 9% was obtained for Al concentration. High concentrations of Al were observed mostly in heavy-traffic roadside soils, with a maximum of 7.81% at St. 11 followed by 7.43% at St. 4. Lower concentrations of Al were found in soils in both the suburban and downtown areas (Fig. 2).

The distributions of Fe, Mn, Cr, Cu, Cd, Co, V, Zn, As, Mo, Sb, and Pb in the urban topsoil showed large spatial variations (Fig. 2). The concentrations of metals studied, except those of Fe, Mn, Cr, Co, and Mo, were higher than the upper continental crust average values at most sites (Table 3; Fig. 2), indicating significant metal pollution in the topsoil of Ulaanbaatar. The average concentrations were 30.0 ± 12.6 mg kg^{-1} for Cr, 20.8 ± 10.0 mg kg^{-1} for Cu, 2.26 ± 0.51 mg kg^{-1} for Fe, 556 ± 139 mg kg^{-1} for Mn, 57.8 ± 13.3 mg kg^{-1} for V, 116 ± 46 mg kg^{-1} for Zn,

Table 3 Descriptive statistics of soil metal concentrations (mg kg^{-1}) and soil properties (pH, OM)

	Mean	Median	Std. dev.	Min	Max	Percentiles					UCC[a]
						5	25	50	75	95	
Al, %	6.54	6.47	0.59	5.10	7.81	5.21	6.20	6.47	7.01	7.75	7.744
Cr	30.0	26.4	12.6	14.8	67.2	15.1	20.8	26.4	39.2	64.2	35
Cu	20.8	19.2	10.0	9.17	57.1	9.44	14.3	19.2	24.0	53.5	14.3
Fe, %	2.26	2.12	0.51	1.53	3.26	1.55	1.90	2.12	2.80	3.24	3.089
Mn	556	505	139	380	898	385	462	505	671	886	527
V	57.8	53.8	13.3	41.6	85.3	42.0	47.1	53.8	68.6	85.2	53
Zn	116	102	46	56.4	220	56.8	85.3	102	150	216	52
Co	9.16	8.23	2.83	5.90	15.3	5.94	6.89	8.23	10.9	15.2	11.6
As	9.41	9.14	4.04	2.08	22.7	2.47	6.98	9.14	11.5	21.2	2
Mo	1.51	1.33	0.63	0.62	3.03	0.65	1.15	1.33	1.80	2.99	1.4
Cd	0.25	0.20	0.24	0.09	1.27	0.10	0.12	0.20	0.26	1.14	0.102
Sb	2.18	1.89	0.84	1.25	4.08	1.26	1.55	1.89	2.78	4.03	0.31
Pb	45.1	36.9	32.0	14.1	164	14.9	25.0	36.9	51.8	152	17
pH	7.44	7.38	0.41	7.00	8.86	7.00	7.18	7.38	7.65	8.75	7.744
OM %	2.32	2.30	1.88	0.03	7.50	0.04	0.57	2.30	3.47	7.10	35

[a]Upper continental crust average (Wedepohl 1995)

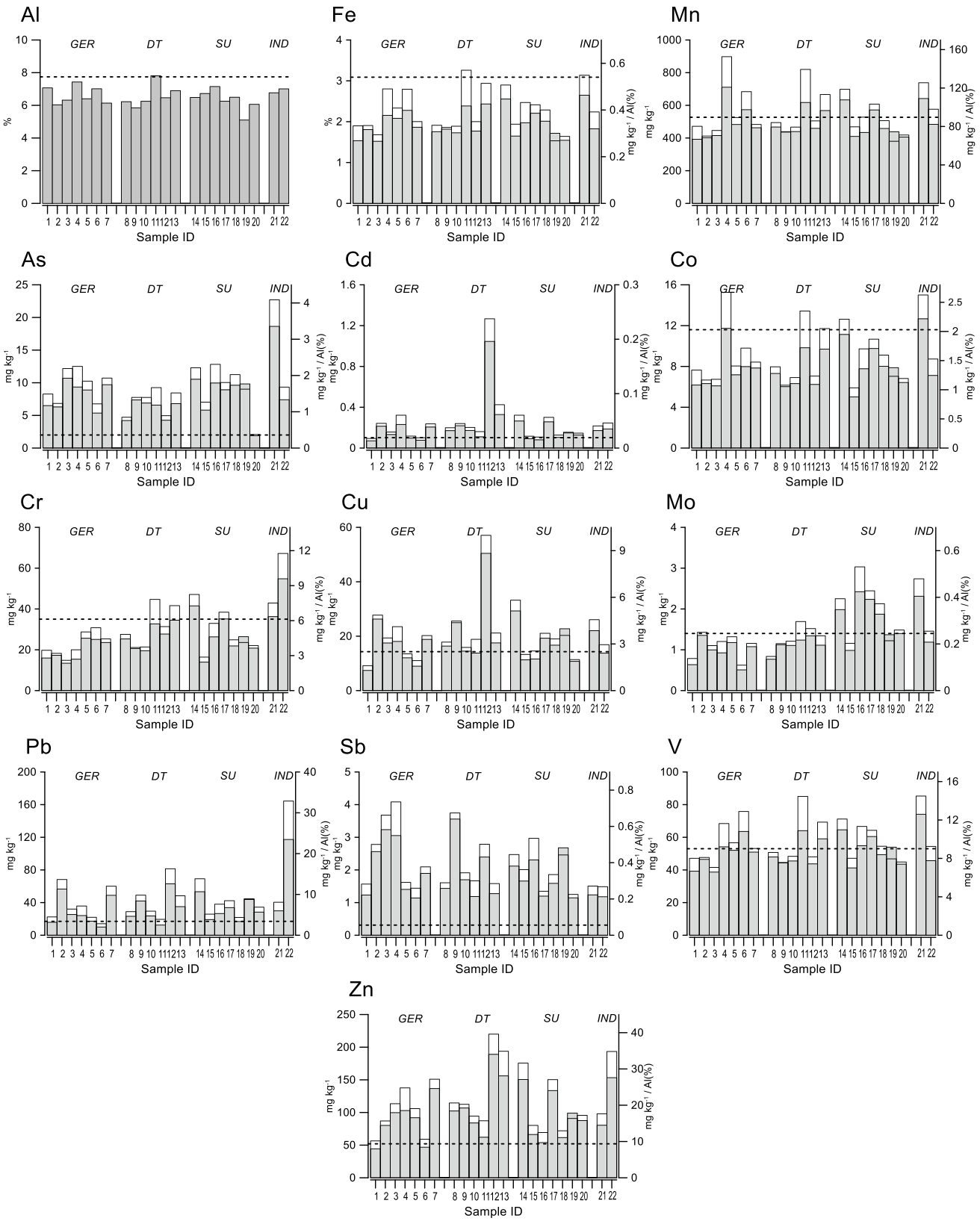

Fig. 2 Metal concentrations at sampling sites. Gray areas represent Al normalized concentrations. Horizontal dashed lines indicate average concentrations in the upper continental crust (Wedepohl 1995). *GER* ger area; *DT* downtown; *SU* suburban settlement; *IND* industrial area

9.16 ± 2.83 mg kg^{-1} for Co, 9.41 ± 4.04 mg kg^{-1} for As, 1.51 ± 0.63 mg kg^{-1} for Mo, 0.25 ± 0.24 mg kg^{-1} for Cd, 2.18 ± 0.84 mg kg^{-1} for Sb, and 45.1 ± 32.0 mg kg^{-1} for Pb (Table 3). The concentrations of As, Pb, and Cr in the soil were higher in the industrial area than in the other areas, reaching maximum values of 22.7 mg kg^{-1}, 164 mg kg^{-1}, and 67.2 mg kg^{-1}, respectively. Chromium (Cr) and Mo showed moderate enrichment at the downtown, suburban, and industrial sites but lower concentrations in the ger area (Fig. 2). Relatively higher values for Cd, Cu, and Zn concentrations were observed at the downtown site, St. 12, than at the other sites (1.27 mg kg^{-1} for Cd, 57.1 mg kg^{-1} for Cu, and 220 mg kg^{-1} for Zn). Zinc (Zn) concentrations showed large variations among the sites; however, no noticeable difference in Zn concentration was observed among the different functional zones of the city, suggesting that Zn is distributed throughout the city (Fig. 2). Unlike other metals, Sb was more enriched in the soils of the ger area, with a maximum of 4.08 mg kg^{-1} at St. 4. Iron (Fe) and Co concentrations were lower than or similar to the UCC averages at the majority of the sites in this study.

Pollution Level Assessment

To evaluate the extent of anthropogenic metal pollution in the soil, the crustal enrichment factor (EF) was applied (illustrated in Fig. 3). EF values less than or close to unity are considered in the range of natural variability, whereas values above unity indicate enrichment corresponding to anthropogenic inputs (Chen et al. 2007; Marrugo-Negrete et al. 2017). The average upper continental crust (UCC) concentration (Wedepohl 1995) was used as a reference material in EF calculations, because there is no information available on the typical background values for Mongolian soils or for the metal concentrations in the soils of Ulaanbaatar (Batjargal et al. 2010; Kasimov et al. 2011). Significant enrichments in metals were found in soils (Fig. 3). Chromium (Cr), Fe, Mn, V, Co, and Mo were not significantly enriched ($EF < 3$, no enrichment to minor enrichment). The EFs of Cu, Zn, As, Cd, Sb, and Pb underline the moderate to severe degree of contamination in the topsoil. An anthropogenic contribution ($10 < EF$) was found for Sb, As, Cd, and Pb, depending on the nature of the contaminated samples. Overall, the extent of contamination for the metals decreased in the order Sb > As > Pb > Zn > Cd > Cu > Mo > V > Cr > Mn > Co > Fe (Fig. 3). The background concentration was higher than the UCC concentration, as previously observed for As in suburban samples (Kasimov et al. 2011) from Ulaanbaatar. Thus, the EF UCC values are probably overestimated for As. Considering local background value (8.6 mg kg^{-1}) from the literature (Kasimov et al. 2011), a decreased degree of contamination ($EF \leq 3$) is expected for As.

Table 4 presents the crustal EF mean values and ranges of the metals studied in the four different functional areas. Relatively high EF values (up to 13 for As, 11 for Pb, 5.6 for Sb, 4.1 for Zn, 2.1 for Cr) were found in soils near the industrial area, which receives large amounts of metallic pollutants from nearby industrial and power plants (AQA-UB 2013; Naidansuren et al. 2017). Antimony (Sb) had the highest EF values in the ger area of all metals in this study, with an average of 9.3 ± 3.9 (range from 5.1 to 15). Zinc (1.7–5.1), Cu (1.3–4.8), and Cd (1.6–15) showed considerably higher EF values in the downtown area along the main roads than in the other areas. Vanadium (V), Co, Mn, and Fe exhibited the lowest EF values among the metals studied ($EF \approx 1$). By combining the EF values of the metals studied, the degree of pollution was found to decrease in the following order: industrial > downtown > suburban settlement > ger area.

Chemical Fractions of Metals and RAC

There are significant differences in bioavailability and toxicity properties in the different geochemical fractions of metals in soil (Filgueiras et al. 2002; Hu and Cheng 2016).

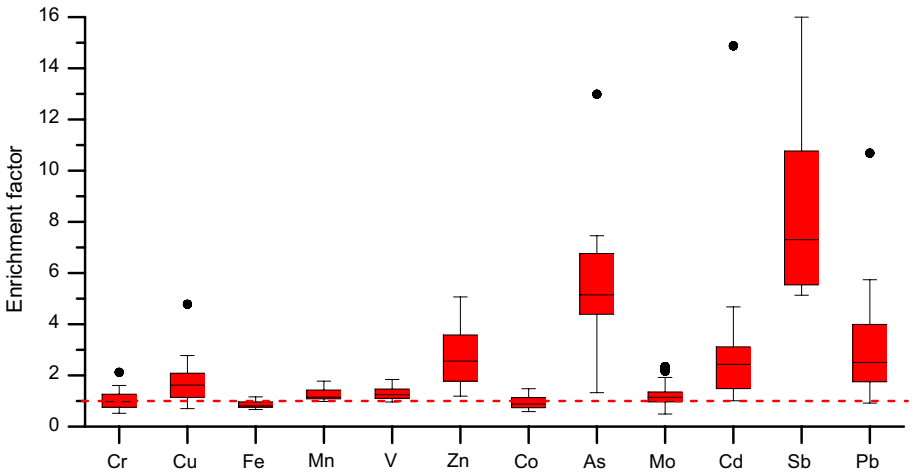

Fig. 3 Box plot of enrichment factors of metals in soil of Ulaanbaatar (Tukey style). Boxes represent 25th, 50th, and 75th percentiles. Whiskers extend to the 5th and 95th percentiles. Outliers are in black dots. The red dashed line indicates EF = 1

Table 4 Enrichment factors (EFs) of soil metals

	Cr	Cu	Fe	Mn	V	Zn	Co	As	Mo	Cd	Sb	Pb
Ger area ($n=7$)												
Mean	0.8	1.5	0.8	1.2	1.2	2.3	0.9	*5.7*	0.9	2.1	*9.3*	2.6
Std (1σ)	0.2	0.6	0.1	0.3	0.2	0.9	0.2	1.4	0.3	1.0	3.9	1.6
Min	0.5	0.7	0.7	1.0	1.0	1.2	0.7	3.7	0.5	1.0	*5.1*	0.9
Max	1.0	2.5	1.0	1.8	1.6	3.7	1.4	*7.5*	1.3	3.3	**15**	*5.2*
Downtown ($n=6$)												
Mean	1.0	2.2	0.9	1.2	1.3	3.1	0.9	4.2	1.1	4.9	*8.7*	3.0
Std (1σ)	0.2	1.3	0.1	0.2	0.2	1.3	0.2	1.0	0.2	*5.0*	4.1	1.6
Min	0.8	1.3	0.8	1.1	1.1	1.7	0.7	2.9	0.7	1.6	*5.3*	1.1
Max	1.3	4.8	1.1	1.5	1.6	*5.1*	1.1	*5.1*	1.3	**15**	**16**	*5.7*
Suburban ($n=7$)												
Mean	1.0	1.6	0.9	1.2	1.3	2.5	0.9	5.6	1.7	2.2	*8.2*	2.9
Std (1σ)	0.4	0.7	0.2	0.2	0.2	1.0	0.2	2.2	0.5	1.1	2.6	1.2
Min	0.5	1.0	0.7	1.0	1.0	1.4	0.6	1.3	1.0	1.1	*5.2*	1.5
Max	1.6	2.8	1.1	1.6	1.6	4.0	1.3	*7.3*	2.3	3.8	**12**	4.9
Industrial ($n=2$)												
Mean	1.8	1.7	1.0	1.4	1.5	3.1	1.2	*9.1*	1.7	2.5	*5.4*	*6.7*
Std (1σ)	0.5	0.6	0.3	0.3	0.5	1.4	0.5	*5.5*	0.8	0.2	0.2	5.6
Min	1.4	1.3	0.8	1.2	1.1	2.2	0.8	*5.2*	1.1	2.4	*5.3*	2.7
Max	2.1	2.1	1.2	1.6	1.8	4.1	1.5	**13**	2.2	2.7	*5.6*	**11**

Italics: EF = 5–10; Bold: EF = 10–25

Considering only the content of metals does not provide accurate information on their bioavailability and risk to the environment (Cuvier et al. 2016; Ke et al. 2017). Therefore, a sequential extraction procedure was proposed to evaluate the labile fractions and geochemical behavior of metals within the soil (Rauret et al. 2000). One of the most widely used sequential extraction methods is the BCR method, which was developed by the European Community Bureau of Reference (BCR) (Hu and Cheng 2016; Ke et al. 2017).

The distributions of the geochemical speciation of each metal studied in the six selected soil samples from Ulaanbaatar are presented in Fig. 4. Copper (Cu) data missed for site 7. All of the metals except for Zn, Pb, and Cu had an appreciable portion, more than 50% on average, of the residual fraction in all of the soil samples, showing that the metals exhibited the strongest association with the crystalline structures of the minerals and were stable under natural conditions with low mobility. Among all studied metals in this study, the acid soluble fraction of Zn was the highest, with a mean proportion of 28.2% (10.7–39.6%). Compared with the proportions of the other metals, these values suggested that Zn exhibited in labile fraction and higher potential bioavailability. The acid soluble fraction percentages of Mn (19.3%), Co (11.3%), As (4.4%), and Cu (1.3%) were higher than those of the rest of the metals. A notable percentage of all the metals were in the mobile fraction, indicating that they show some ecological risk in sediment. Antimony (Sb), As, Co, V, Fe, and Mn were dominated by the residual fraction, with mean proportions of 88.3%, 57.8%, 53.5%, 81.7%, 82.0%, and 55.0%, respectively (Fig. 4). These fractions pose a decreased ecological risk because of the decreased possibility of transferring in the environment. The percentage of the total variable fraction of metals (all except for the residual fraction) was calculated to prevent a sole metal fraction in urban soil from influencing the analysis due to changes in environmental or geochemical conditions (Landner and Reuther 2005). The nonresidual fractions of the metals, Zn, Pb, Cu, Mn, Co, and As accounted for 72.7%, 77.7%, 64.0%, 45.0%, 46.5%, and 42.2% of the total fraction, respectively. All of these elements were readily mobilized in the soil of Ulaanbaatar. According to the RAC classification, Zn exhibited the highest in labile fraction and potential bioavailability risk, especially at sites 7, 12, and 13, at which more than 30% of Zn was in the acid soluble fraction. Manganese (Mn) showed a moderate risk in all of the samples. Arsenic (As) posed a light risk in all six samples, with an acid leachable fraction of 2.7–5.9%. A light risk was also assessed for Ni, Cu, Pb, and Sb. Overall, the environmental risk of the bioavailable fraction of the ten metals, based on their RAC values, decreased in the following order: Zn > Mn > Co > As > Cu > Sb > Pb > Cr > Fe > V, indicating that Zn was the key pollutant with ecological risk in the Ulaanbaatar area.

Fig. 4 Distribution of trace metals in soil samples identified by sequential extraction

Source Evaluation of Metals in Soil

The major anthropogenic sources of toxic metals in many urban areas are traffic, vehicle emissions, brake abrasion and tire wear (Huang et al. 2015; Luo et al. 2015; Nazarpour et al. 2019). Furthermore, industrial effluent, power plants, small local mining activities, fossil fuel combustion, industrial zones, household release, weathering of asphalt, and roadside litter can contribute to toxic metal release into the urban environment (Batjargal et al. 2010; Chung and Chon 2014; Luo et al. 2015; Nazarpour et al. 2019).

A principal component analysis was conducted to identify the possible sources of metals, and the results are presented in Table 5 and Fig. 5. To exclude grain size contributions to soil metal contents, Al-normalized concentrations (Soyol-Erdene et al. 2019) were used for PCA. The Kaiser–Meyer–Olkin (KMO) measure of sampling adequacy (0.626) and Bartlett's test of sphericity (262.82, $p=0.000$) showed that the twelve metal concentrations were suitable for PCA (Cerny and Kaiser 1977). In the results, five factors were extracted that could account for greater than 90% of the total variation in metal concentrations. The rotated component matrix indicated that Fe, Mn, V, and Co were strongly associated with the first factor (C1) with high loadings (0.92, 0.96, 90, and 0.93, respectively), explaining 30% of the total variance. Factor 2 (C2), dominated by Cu, Zn,

Table 5 Rotated component matrix of factor loadings

	Factor				
	1	2	3	4	5
Cr/Al	0.46		0.76		-0.37
Cu/Al		0.94			
Fe/Al	0.92				
Mn/Al	0.96				
V/Al	0.90			0.33	
Zn/Al		0.68	0.63		
Co/Al	0.93			0.27	
As/Al	0.41			0.74	0.31
Mo/Al				0.87	
Cd/Al		0.90			
Sb/Al	-0.34	0.40			0.70
Pb/Al		0.27	0.91		
pH					0.88
OM	0.40	0.69		0.37	
Initial Eigenvalues	5.113	3.362	1.849	1.135	1.106
Variance, %	30	21	14	13	12
Cumulative, %	30	51	65	78	90

Extraction method: principal component analysis

Rotation method: varimax with Kaiser normalization

< 0.25 considered not important

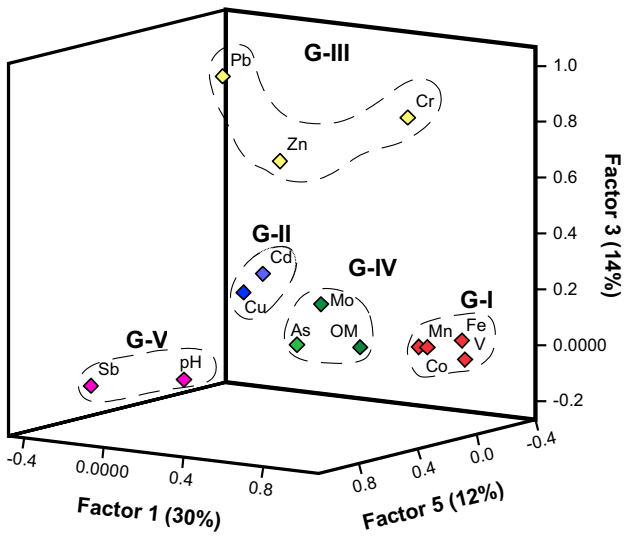

Fig. 5 Principal component analysis (PCA) diagram with the metal concentrations and selected physical parameters of soil samples

Cd, and OM, accounted for 21% of the total variance with loadings of 0.94, 0.68, and 0.90, respectively. The third factor (C3) mainly included the data for Cr, Pb, and Zn with high loadings (0.76, 0.91, 0.63) and explained 14% of the total variance. Arsenic (As) and Mo were extracted in the fourth factor (C4) with high loadings of 0.74 and 0.87, respectively, explaining 13% of the total variance. Antimony (Sb) and soil pH were isolated with loadings of 0.70 and 0.88 in the last factor (C5), accounting for 12% of the total variance. Meanwhile, it also was found that Cr was partially represented in C1 (with a relatively high loading of 0.46), suggesting a complex source for Cr. Similarly, partial representations were found for Zn in C3 (loading 0.63), for V in C4 (0.33), for As in C1 and C5 (0.41 and 0.31, respectively), and for Pb and Sb in C2 (0.27 and 0.40, respectively), indicating that these soil metals have complex sources.

One group of elements comprised Cr, Fe, Mn, V, Co, and As. Soil organic matter also was loaded in this group. A negative loading for Sb also extracted in this group. The concentrations of Cr, Fe, Mn, V, and Co were slightly lower than or close to background level (Fig. 2; Table 3) with lower enrichment factors (Table 4; Fig. 3). These findings suggested that this group of elements might originate from natural sources, such as mineral weathering (Chen et al. 2007).

The second group included Cu, Zn, Cd, Sb, and Pb, combined with organic matter. A high degree of pollution for those metals was shown by their maximum enrichment factors of up to 4.8, 5.1, 15, 16, and 11, respectively. Zinc (Zn) pollution is distributed throughout the city (Fig. 2), with no clear differences among the various functional areas. Traffic emissions due to tire abrasion are the most important contributor to Zn emissions in urban environments, because Zn is the most abundant metal released from tire wear (Smolders and Degryse 2002; Wang et al. 2012). Cu, Cd, Sb, and Pb, the other metals of this second group, also are associated with traffic emissions, including oil combustion, road abrasion, and tire and break wear (Batjargal et al. 2010; Chen et al. 2010; Lough et al. 2005; Mielke et al. 2010). Due to the well-known high emissions of lead (Pb), many countries banned the usage of leaded gasoline in the 2000s (Mielke et al. 2010). However, in Mongolia, leaded gasoline was still used until the middle of the 2010s and can greatly affect urban environmental pollution (Batjargal et al. 2010; Guttikunda et al. 2013). Antimony (Sb) and Cu are the main metal pollutants emitted from break wear abrasion (Lough et al. 2005). Anthropogenic emissions of Cd are mainly released by zinc production; therefore, a significant amount of Cd is contained in tires (Cheng et al. 2014) and emitted by tire abrasion to the environment (Hjortenkrans et al. 2007). The combustion of fossil fuels and heavy oils also contributes a significant proportion of Cd emissions (Cheng et al. 2014). The high loading of soil organic matter in C2 can indicate organic pollution resulting from incomplete combustion of motor vehicle oils. Therefore, based on the results of the PCA, soil Zn, Cu, Cd, and Sb originate from traffic emissions. As a result of rural-to-urban migration, the population of UB has almost doubled during the last two decades (NSOM 2018). This sharp increase was not only

in the city's population but also in the number of vehicles, which increased by a factor of six during the same period and reached ~ 540,000 (NSOM 2018). In many cities in developing countries, such as Ulaanbaatar, sharp increases in automobile numbers and in the consequent emission of traffic-related environmental pollutants are mainly attributed to inadequate public transportation systems. Especially in Ulaanbaatar, which has a very long, cold winter with a mean air temperature of − 19 °C (Ganbat et al. 2013), the public transportation system (i.e., subway and tram) is not well-developed; most people use private cars every day, resulting in heavy traffic and related pollutant emissions (Davy et al. 2011; Guttikunda et al. 2013). Additionally, due to rapid urbanization, a large number of used tires are imported from developed countries (Customs 2016). According to a report from the Mongolian customs administration, in 2015, approximately 469 thousand used tires (mainly from Japan), and only 103 thousand new tires were imported to Mongolia (Customs 2016). Used tires have a much lower life span than new tires and are consumed at high rates. Moreover, according to the World Bank, low-income people in the ger area burn waste materials, such as used tires in traditional low-efficiency stoves (World Bank 2008) to produce heat during the cold winter season; this burning can contribute to the release of toxic metals from tires to the environment (Downard et al. 2015). This suggests that nationwide uncontrolled use of used (old) tires can possibly cause a large amount of metal pollution in urban areas of developing countries.

The third group consisted of Cr, Zn, and Pb. These metals also had high pollution degrees and high enrichment factors, especially in the industrial areas: 2.1 (Cr), 4.1 (Zn), and 11 (Pb) (Table 4). Hence, this group of elements may originate from industrial activities in the city. High degree of pollution from Cr and other metals in areas surrounding tanneries were investigated (Naidansuren et al. 2017). There are 140 tannery facilities in Mongolia, and most are located in the industrial district of Ulaanbaatar (Naidansuren et al. 2017). Tanneries use chromium salt and other environmentally hazardous chemicals, often over several decades, and release toxic metals in the surrounding soil and water (Naidansuren et al. 2017). Other possible industrial sources of these metals are coal burning power plants and their ash ponds (Soyol-Erdene et al. 2019). Three main power plants (PP) in Mongolia are located in the industrial area of UB (AQA-UB 2013; Kasimov et al. 2011). Metal-enriched ash produced from power plants is stored in open ponds near the plants and supply metals to the surrounding environments through the air and rain washout (AQA-UB 2013; Soyol-Erdene et al. 2019). Power plant ash is considerably enriched by As, containing up to 203 mg kg^{-1} (Batkhuyag et al. 2016; Kosheleva et al. 2010); however, it was not extracted in factor 3 by the PCA (Table 5), suggesting a limited contribution of PP ash to this group of elements.

The fourth group consisted of As, Mo, V, Co, and soil organic matter. Arsenic (As) is the main pollutant derived from coal combustion (Hong et al. 2012; Nriagu 1996); since organic matter is also in this group, the most reasonable candidate for this factor (C4) is coal combustion in UB. Furthermore, it has been found that a large amount of vanadium (V) releases to the environment by coal combustion because the most important source of anthropogenic vanadium in the world is related to fossil fuel combustion (Schlesinger et al. 2017). The other metals in this group, Co and Mo, are also mainly released by coal burning since it is also the most dominant source for global Co and Mo emissions (Van de Velde et al. 1999). Due to the very cold and long winter season, hundreds of thousands of low-income families living in gers use coal and wood for heating. As a consequences of the incomplete combustion in traditional inefficient stoves, coal combustion in gers is the main source of air pollution, and it makes UB one of the most air-polluted cities in the world (Davy et al. 2011; Guttikunda et al. 2013; Lim et al. 2018). There are approximately 200,000 gers in Ulaanbaatar (Badarch et al. 2018), and each burns an average of 5 tons of coal and 3 m^3 of wood per year (Guttikunda et al. 2013). Atmospheric particulate matter pollution, which is mainly derived from incomplete coal combustion in gers, frequently exceeds ten times the national permissible level (50 μg/m^3) in winter (Lim et al. 2018). Therefore, coal combustion emissions may be one of the major sources of metal pollution in UB soil.

The fifth group consisted of Sb and As. Soil pH is also loaded in this group. The degree of Sb pollution was very high based on the enrichment factor (Fig. 4). The spatial distribution characteristics of Sb also were clearly different from those of other metals, with higher concentrations and enrichment factors appearing in the ger area (St. 2, St. 3, St. 4) (Table 4; Fig. 2). Furthermore, positive loading of pH in factor indicates that sources of these metals also increase soil pH. Coal ash from households and power plants usually produces an alkaline pH (Gomes et al. 2016). Extremely high pH was found in household ash in Ulaanbaatar (Batkhuyag et al. 2016), which may have resulted from wood ash burned with coal (Demeyer et al. 2001; Guttikunda et al. 2013). Moreover, coal combustion is known to be one of the main anthropogenic sources of Sb and As in the environment (Hong et al. 2012; Kosheleva et al. 2010; Tian et al. 2011). There are two main pathways for releasing Sb and As to the environment during coal combustion. They can be emitted to the atmosphere as volatile fractions and/or as solid combustion byproducts, i.e., ashes (Kosheleva et al. 2010; Llorens et al. 2001). Considerable enrichment of these metals was observed in Mongolian coal and coal ash (Batkhuyag

et al. 2016; Kosheleva et al. 2010). Therefore, in summary, the fifth factor (C5) is possibly associated with household ash wastes in the ger area. Household ash produced from gers is transported to landfill sites (Batkhuyag et al. 2016) but is distributed to surrounding areas by the wind before the ash is moved to landfills due to the low frequency of waste collection in the ger area (Amarsaikhan et al. 2014; Kamata et al. 2010).

Our findings indicate that inefficient control of industrial releases and household ash, incomplete coal combustion, and the nationwide use of aged tires can be major sources of toxic metals entering into soil of UB city. Thus, the toxic metal emission sources should be controlled effectively to prevent the population from health risk as well as the spreading of pollution into the environment through atmospheric long-range transport.

Conclusions

We investigated the enrichment, geochemical fractions, and potential sources of metals, including Al, Fe, Mn, Cr, Cu, Cd, Co, Zn, V, Mo, As, Sb, and Pb, in the urban soil of Mongolia. Significant anthropogenic enrichments were identified for Sb, Zn, Pb, Cu, As, and Cd in urban soil. Zinc (Zn) had the highest pollution risk, with more than 30% in the exchangeable fraction. Multivariate statistics coupled with analyses of spatial distributions and the pollution assessment indexes EF and RAC successfully separated the metal sources. Among the 12 contaminating elements, the accumulation of soil Sb and As were mainly attributed to coal combustion, whereas Zn, Cu, Cd, and Pb were mainly derived from traffic-related factors, i.e., tire abrasion and possibly oil combustion. In addition to the impacts of traffic-related factors and dusts, industrial factors were the main sources of Pb, Zn, and Cr. Thus, it is necessary to monitor the sources to minimize the exposure risk of general population to the toxic metals and also prevent the long-rage atmospheric transport over a wider area.

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Compliance with Ethical Standards

Conflict of interest The authors declare that there is no conflict of interest regarding the publication of this article.

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