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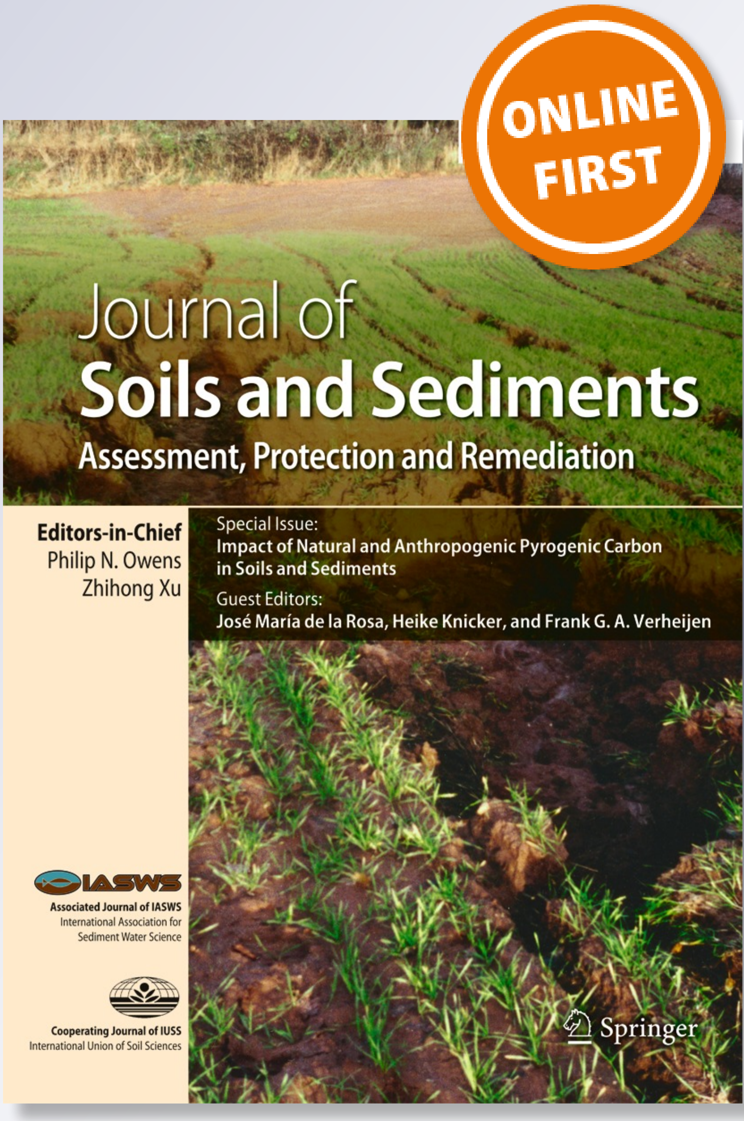

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SOIL POLLUTION AND REMEDIATION

Geochemical transformation of soil cover in copper–molybdenum mining areas (Erdenet, Mongolia)

Ivan V. Timofeev[1] · Natalia E. Kosheleva[1] · Nikolay S. Kasimov[1] · Petr D. Gunin[2] · Enkh-Amgalan Sandag[3]

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Abstract
Purpose The aim of the present study is to evaluate geochemical transformation of soil cover in the territory of Erdenet (Mongolia) and to assess the environmental risk associated with soil cover contamination. The objectives of the present study included: (1) the determination of heavy metals (HMs) and metalloids contents in surface horizons of background and urban soils and the assessment of geochemical transformation of the city's soil cover; (2) the identification of elements' associations and patterns of their spatial distribution in the soil cover of the city; (3) the assessment of environmental hazard, related to contamination of soils with complexes of HMs and metalloids.
Materials and methods Soil–geochemical survey was conducted by the authors in the summer periods of 2010 and 2011. In total, 225 samples, including 32 backgrounds, were collected. Bulk contents of HMs and metalloids in soil samples were analyzed by mass-spectral method with inductively coupled plasma at All-Russian Research Institute of Mineral Raw Materials (Moscow) using Elan-6100 and Optima-4300 devices (Perkin Elmer, USA).
Results and discussion Mo, Cu, and Se appeared to be the priority pollutants nearly in all land-use zones. The maximum accumulation of Mo, Cu, Se, As, Sb, and W is restricted to the industrial area where total pollution index of soils (Zc) equals 74.8. Three technogenic associations of elements, derived mainly from petrochemical features of Erdenet ore field and characterized by similar spatial distribution within the city, are identified. Environmental assessment of surface soil horizon geochemistry in Erdenet showed that 1/5 of its area has dangerous and extremely dangerous levels of soil pollution.
Conclusions Experience of the environmental–geochemical assessment of soil cover in the impact zone of mining enterprises could be useful for other fields of the non-ferrous metals with high lithological–geochemical heterogeneity of the territory. It suggests the need of accounting for the geological diversity and specific features of metallogeny of an area. Geochemical indices local enrichment factor/local depletion factor should be calculated against the individual background values for each soil-forming rock. Such approach allows more accurate assessment of the degree of technogenic geochemical transformation of soils and the environmental hazard of pollution.

Keywords Environmental hazards · Heavy metals · Mining landscapes · Technogenic anomalies · Type of land use · Urban soils

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1 Introduction

Increased consumption of mineral resources accompanies global population growth. In mining regions, technogenic pressure on the environment causes multiple problems associated with pollution of air, water, and soil (Perel'man and Kasimov 1999; Avessalomova et al. 2004; Hudson-Edwards

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et al. 2011; Csavina et al. 2012; Koz et al. 2012; Osipova et al. 2014; Dragović et al. 2014; Schaidera et al. 2014). Development of non-ferrous metal deposits has given rise to cities around mines and ore processing plants. The geochemical features of these urban environments are largely based on metallogenic specialization of underlying lithologies. However, anthropogenic activities disturb biological cycling, geochemical structures, and ecological functions in these landscapes (Bech et al. 1997). Natural ecosystems are replaced by technogenic systems in which a significant accumulation of heavy metal (HM) and metalloid elements may occur. Because the geochemical transformation of natural landscapes often threatens public health (Loredo et al. 2006; Pruvot et al. 2006; Li et al. 2014), environmental consequences of mining activities have been the focus of scientific research worldwide.

A great number of studies focuses on contamination of various natural media in the impact zones of ore-dressing and processing plants and their tailings (Elpat'evskyi 1993; Milu et al. 2002; Owor et al. 2007; Gomez-Alvares et al. 2009; Lizarraga-Mendiola et al. 2009; Rey et al. 2013); some of them deal with the problem of bioindication of air pollution (Serbula et al. 2012) and the analysis of contaminant fluxes within soil–plant–animal systems (Gedgafova and Uligova 2007). Other studies identify spatial and vertical distribution of various fractions of HMs and metalloids in soils (Garcia-Sanchez et al. 2010; Rastmanech et al. 2011; Candeis et al. 2011).

The aim of the present study is to evaluate the geochemical transformation of soil cover in Erdenet, Mongolia, and to assess the environmental risk associated with soil cover contamination. Erdenet is a mining city where one of the world's largest deposits of copper and molybdenum ore has been exploited since 1976. A joint Mongolian–Russian venture, the Erdenet Mining Corporation, extracts 27.8 million tons of ore per year. After 40 years of mining operations, a huge tailing area—covering more than 1,500 ha—has formed in the valley of the Zun-Gol River. Existing literature concentrates on the geological features of mining territories (e.g., Berzina and Sotnikov 2007), mining cycle (e.g., Naboichenko et al. 1982; Tsogtkhangai et al. 2011; Pestryak and Erdenetuya 2012) and the impacts of mining on urban ecosystems (Byambaa 2007). HM and metalloid pollution in urban soils has received insufficient attention.

The objectives of the present study included:

- The determination of HM and metalloid contents in surface horizons of background and urban soils and the assessment of geochemical transformation of the city's soil cover;
- The identification of elements' associations and patterns of their spatial distribution in the soil cover of the city;
- The assessment of environmental hazard, related to contamination of soils with complexes of HMs and metalloids.

2 Object of study

Natural setting. The 180-km^2 study area is on the interfluves of the Selenga and Orkhon rivers in the Erdenetii-Gol river valley. It is an uplifted and dissected highland with elevations of 1,000–2,150 m above sea level. Mountain massifs stretch from the northwest to the east. According to data collected at the Erdenet weather station (www.worldweather.wmo.int), the area's continental climate is known for extremely cold winters ($t_{January}$=−15÷−25 °C) and hot (t_{June}=+20÷27 °C) humid summers when 60–70 % of the total annual precipitation occurs. Winds blow predominantly from the west and southwest in winter and from the north in summer.

The area is part of the Early Caledonian structures of the Selenga-Yablonoi fold system (Waul et al. 1968; Kominek et al. 1969) in the Orkhon-Selenga depression, which is the largest volcanogenic structure in the riftogenic zone of the North Mongolian Permo-Triassic plutonic region (Yarmolyuk and Kovalenko 2003; Berzina and Sotnikov 2007). Tectonically, the study area is within a northwest faulting zone complicated by submeridional faults.

Geochemical features of the study area derive from its location in the northwest section of the Erdenet ore field, comprising Permian volcanic and volcanic–sedimentary formations of the Hanuy age intruded by Late Permian granitoids of the Selenga age. The deposit is of porphyry molybdenum–copper type with Re, Ag, and Se in productive concentrations. The ore also has significant concentrations of Pb, Zn, As, Sr, Bi, Co, Ni, Ge, Ta, Ga, In, and Cd. The host rocks are of three different associations: gabbroid, granitoid, and subalkaline granites with sienogranites (Berzina and Sotnikov 2007). The ore-bearing complex is in stocks (up to 2 km^2) and dike-like bodies (up to 250 m long); it is primarily made up of quartz diorite–porphyrite, granodiorite, and granite porphyry (Gavrilova et al. 2010).

The relief features of soil parent materials vary. On mountaintops and adjacent slopes, soil formation takes place on eluvium (rottenstone, rocky ground, and rock debris). On gentle slopes and at footslope positions, parent materials are characterized by talus deposits (loams, sandy loams, sands, and other terrigenous materials from 2 to 43 m thick). Soils on fans are developed on diluvium and proluvium, whereas in river valleys they are formed on alluvial and proluvial sediments.

The city of Erdenet and its periphery are in the zone of dark Katanozems. Most of the study area is an undulating landscape in which natural soils are thick, dark, and slightly saline Katanozems. In industrial and residential areas, the upper soil horizons are replaced by manmade layers—mixed "urbic" horizons with some organic matter and anthropeic material (solid domestic and industrial waste).

The study area is in a mountain steppe and forest zone similar to the forest steppe of South Transbaikalia, where forest and steppe communities alternate based on mountainous relief and slope characteristics. In the city, natural short grass vegetation has been preserved only on the slopes of the Erdenetii-Gol and Gaveli-Gol river valleys. Natural larch forests are closer to the mountaintops. Isolated larch and poplar plantations are typical in the urban area. Grass cover is minimal due to intensive livestock grazing (Vostokova and Gunin 2005).

Technogenic impact. Erdenet Mining Corporation (EMC) is one of the biggest ore extraction and processing operations in Asia. It is a relatively large complex that processes 26 million tons of ore per year. It also produces approximately 530, 000 tons of copper concentrate and 4,500 tons of molybdenum concentrates annually (www.erdenetmc.mn 2013). Ores are extracted through open-pit mining. The length of the pit is 2,800 m, and its width is 1,600 m; benches are at intervals of 15 m. Since 1976, 440 million m^3 of commercially useful rock and overburden have been excavated. The Erdenet mineral processing plant is the main production unit, with technological circuits that include crushing and grinding ore (producing a large quantity of dust), flotation with acid, filtration, drying, and shipping copper and molybdenum concentrates. Near the Erdenet processing plant, a smaller plant—called Erdmin and established in 1994—extracts copper from waste rock using sulfuric acid. The application of sulfuric acids may enhance the migration ability of HMs and metalloids in aqueous solutions.

The Erdenet power station is located between the mining and residential areas. Built in 1988, this plant uses sulfur coal from the Sharingol and Baganur strip mines. Like other sulfur coals, they are rich in chalkophile elements. Burning coal releases harmful substances (e.g., sulfur and carbon oxides, HM compounds, carbon black, particles, benzpyrene) into the atmosphere. Erdenet Hivs, the largest company in Mongolia, is also in the vicinity. It was founded in 1981 and produces wool carpets, wool, and camelhair woven products, cashmere, and felt cloth. The substances used to dye wool contain Cr, Ni, Zn, and Cd (Saet et al. 1990).

Land-use zoning. Five land-use zones have been identified in Erdenet city: residential (one with traditional *ger* houses and one with multistory buildings), industrial, transit, and pasture. All industrial enterprises are in the eastern part of the city, in line with the wind rose and 3–5 km away from the modern multistory residential area to the west. The *ger* districts are in the southwestern part of the city, on the upland slopes (Fig. 1). Pastureland occupies the largest area, surrounding the city and buffering residential districts from industrial operations.

3 Materials and methods

A soil geochemical survey was conducted in Erdenet during the summers of 2010 and 2011 within the framework of Russian–Mongolian collaboration on integrated biological research. Samples were taken from the surface horizon (0–10 cm) using a regular grid with 500–700 m spacing. The quantity and location of sampling sites coincided with the study area's lithological heterogeneity. In total, 225 samples were collected including 32 reference samples representing background soils. Reference sites were on hilltops and gentle hill slopes in the valley of the Erdenitii-Gol River and its tributaries (the Zuna-Gol and Gavelin-Gol rivers), 5–6 km from the city (Fig. 1).

HMs and metalloids in soil samples were analyzed via inductively coupled plasma mass spectrometry, at the All-Russian Research Institute of Mineral Raw Materials in Moscow, using Elan-6100 and Optima-4300 devices (Perkin Elmer, USA). Bulk concentrations of 54 elements were determined in this way, and 18 were identified as priority pollutants. They included As, Zn, Se, Cd, and Pb (assigned first-order hazards); Co, Ni, Cu, Mo, and Sb (second-order); V, Sr, Ba, and W (third-order); along with Sn, Cs, and Bi. To determine all the elements, soil samples were transferred in the state of the solution using a single acid opening in the mixture of four acids: HCl, $HClO_4$, HNO_3, with triple evaporation and complexation of HF with boric acid. The residue was then dissolved in HCl.

Particle size data were obtained by means of laser diffraction using laser particle sizer (Fritsch, Germany). The information about soil acidity was received by measuring actual pH by pH-meter (pH340i/set) in 1:5 soil/water suspension. The content of organic matter was determined by the $K_2Cr_2O_7$ wet-oxidation method according to standard procedure (Arinushkina 1992). Total contents of Fe, Al, and Mn oxides were determined by inductively coupled plasma atomic emission spectroscopy.

Based on initial measurements, the average background concentrations of HMs and metalloids (C_r) were estimated for soil groups formed on specific parent rocks in natural landscapes. They were then compared with average concentrations C for uncontaminated world soils (Kabata-Pendias 2011) by calculating the Regional Enrichment Factor, REF = C_r/C (if $C_r > C$), or Regional Depletion Factor, RDF = C/C_r (if $C_r < C$). The extent of soil pollution and geochemical transformation of soil cover was found using the Local Enrichment Factor and Integrated Pollution Index. The Local Enrichment Factor (LEF) and Local Depletion Factor (LDF) were calculated for each element and sampling site using the formulas

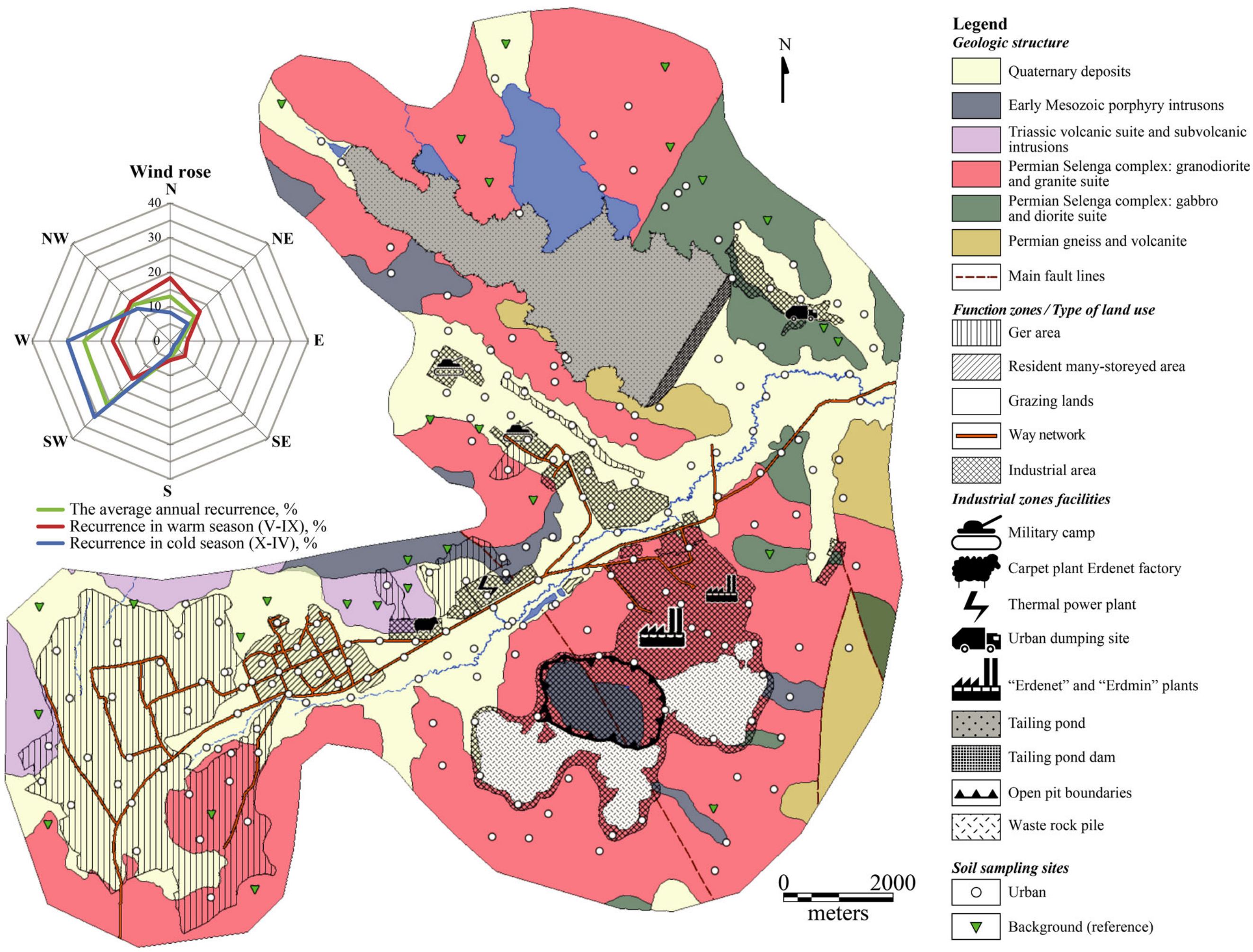


Fig. 1 Geological structure map of Erdenet city with function zones and points of topsoil samples

LEF = C_a/C_r (if $C_a > C_r$) and LDF = C_r/C_a (if $C_r > C_a$), where C_a is the measured concentration of each metal or metalloid in urban soils and C_r is the reference concentration of the element in background soils derived from similar parent material. Based on the calculated enrichment factors, spider diagrams were plotted for reference and urban soils. Integrated Pollution Index Z_c was used to evaluate the extent of geochemical transformation of urban soils due to the impact of EMC

$$Z_c = \sum\nolimits_{i=1}^{n} \mathrm{LEF} - (n-1),$$

where n represents the number of metals with $C_a/C_r > 1$. The values of Integrated Pollution Index Z_c were classified in Table 1.

Statistical analysis with correlation matrices and univariate statistics was performed using Statistica 8 and MS-Excel software. Maximum (max), minimum (min), average (m), standard deviation (σ), coefficient of variation ($C_v = \sigma/m$ 100 %), correlation coefficient (r), and other properties were estimated.

Spider diagrams and dendrograms—obtained through hierarchical cluster analysis—were used to determine element relations in background soils developed on different parent materials and in urban soils from distinct land-use zones. Complete-linkage clustering of chemical elements was applied to reveal HM associations; the Pearson correlation coefficient was used as measure of the affinity of elements.

Table 1 The levels of multi-element contamination of soils and the correspondent degrees of environmental risks (Environmental Geochemistry 1990; Kasimov et al. 2012)

Z_c and IIR values	Contamination level	Environmental risk
<16	Low	Acceptable
16–32	Moderate	Moderate
32–64	High	High
64–128	Very high	Very high
>128	Extremely high	Extremely high

Regression trees were constructed in SPLUS (MathSoft 2004) to reveal the major quantitative as well as qualitative landscape and geochemical factors that impact the enrichment of urban soils with chemical elements. This method helps predict element concentrations and evaluate the impact of different factor combinations (Kasimov et al. 2011).

Soil and geochemical data were visualized and geochemical maps created using local interpolation, or kriging, in MapInfo 11.5 and Surfer 11 software. A modified geological map (Watanabe and Stein 2000) and a city map served as basic means of geographical orientation. To avoid overestimating pollution, extraordinarily high HM concentrations—representing subpopulation extremes, or outliers—were excluded during interpolation. Sites with extreme concentrations are displayed on the maps as pointed anomalies.

Several parameters and indices for environmental risk assessment were used to evaluate the impacts of HM and metalloid contamination in Erdenet soil cover. Maximum permissible concentration (MPC) of HMs is specified in the Mongolia Soil Quality Standards (Х рсний чанар 2008). The ratio of HM and metalloid concentrations to MPC is represented as $K_o = C_a/\text{MPC}$. The Integrated Index of Risk (IIR) associated with soil pollution was calculated based on average concentrations for uncontaminated world soil C and data on pollutant toxicity:

$$\text{IIR} = \sum \left(C_a \Big/ C^* K_T \right) - (n-1),$$

where C_a/C is the enrichment of a sample with an element relative to its average level in soils worldwide, n represents the number of elements with $C_a/C>1$, and K_T is the toxicity coefficient—K_T equals 1.5 for a first-order hazard, 1.0 for a second-order hazard, and 0.5 for a third-order hazard (Vodyanitskyi 2012). The values for this index are classified in Table 1.

Selection of the two integrated indices—Z_c and IIR—was based on the mining area's location within metallogenic anomalies. Z_c represents geochemical transformation of soil cover due to pollution, while IIR indicates the environmental risk associated with soil contamination with HMs and metalloids. Because regional background concentrations of these elements tend to exceed sanitary norms, average concentrations in world soils were used as reference values for calculating IIR.

4 Results and discussion

4.1 Geochemical features of background soils

The geochemical characteristics of regional background soils were ascertained by examining five types formed on distinct parent materials (Table 2). The number of samples in each subpopulation corresponds with the areal proportion of parent rocks in the study area (S). The dominant lithology is made up of Permian granodiorite and granite suites of the Selenga complex (S=50 %). The background subpopulation includes 13 soil samples. It is characterized by high average concentrations of V, Cr, Co, Ni, Zn, and Sr as well as maximum concentrations of elements associated with ore deposit: Cu, Zn, Mo, and W.

Early Mesozoic porphyry intrusions were found in restricted areas (S=3–4 % of the study area), and the corresponding subpopulation consisted of only two reference samples. They were rich in As, Cd, Sn, and Pb. Minimum concentrations of these elements were found in surface horizons of soils from the Triassic volcanic suite and subvolcanic intrusions.

Analysis of the spider diagram showing five groups of soils formed on different parent material according to the ranged elements' enrichment and depletion factors (Fig. 2) indicates that background concentrations of Se, Mo, Sb, and Sr in the study area exceed averages for uncontaminated world soils. The regional enrichment factor varies from 5.2 to 7 for Se, from 1.7 to 5.1 for Mo, from 1.2 to 2.4 for Sb, and from 1.8 to 2.3 for Sr. Comparatively high background levels were found in soils derived from Early Mesozoic porphyry intrusions (W and As) as well as Triassic volcanic suite and subvolcanic intrusions (W and Ba).

The results of geochemical analysis showed a different level of variation for HM and metalloid concentrations in background soils not affected by mining activity. The first group of elements—V, Cr, Co, Ni, Zn, Cd, Sn, Cs, Ba, and Pb—had low variation coefficients (C_v=0–32 %). The second group—Sr, As, and Bi—contained elements with medium variation (42–67 %). The third group—Cu (287 %), Mo (218 %), Sb (127 %), W (107 %), and Se (96 %)—had highly varied concentrations for background soils within the study area.

4.2 Geochemical characteristics of urban soils and their anthropogenic transformation

Analysis of the spider diagram of soils from different land-use zones (Fig. 3) indicated that most pollution occurs in the industrial zone, where soils are contaminated with Mo (LEF=10.7), Cu (10.6), Se (2.3), As, Sb (1.5), and W (1.5). This zone contains deposits rich in ore-linked elements (Gavrilova et al. 2010). Atmospheric emissions from ore processing significantly increase metal concentrations in the upper horizons of neighboring soils.

Upper soil horizons were contaminated with Mo, Cu, and Se in all land-use areas except the *ger* residential districts. Soils in the multistory residential area contained Bi, Cd, Pb, and Zn. Levels of these elements were, respectively, 2.3, 2.2, 1.3, and 1.4 times higher than in reference samples. The first two elements, found in the output of coal combustion, are

Table 2 Statistical parameters and regional geochemical coefficients for heavy metals and metalloids in the surface 0–10 cm horizons of background (uncontaminated) soils located in the vicinity of the city of Erdenet

Parameters	V	Cr	Co	Ni	Cu	Zn	As	Se	Sr	Mo	Cd	Sn	Sb	Cs	Ba	W	Pb	Bi
Soils on Quaternary deposits (n=8)																		
Average, ppm	142.1	49.5	14.8	25.5	34.8	82.3	8.6	3.1	308.8	2.3	0.2	2.7	1.4	4.9	575.0	2.0	22.4	0.3
σ, ppm	30.5	11.4	4.1	7.2	6.3	21.5	1.4	1.3	82.0	1.8	0.1	0.5	1.5	0.6	94.3	0.5	5.1	0.1
REF (RDF)	1.1	(1.2)	1.3	(1.1)	(1.1)	1.2	1.3	7.0	1.8	2.1	(1.6)	1.1	2.1	(1.0)	1.3	1.2	(1.2)	(1.5)
Soils on Early Mesozoic porphyry intrusions (n=2)																		
Average, ppm	165.0	54.5	16.5	26.5	50.5	95.5	13.0	2.3	325.0	3.9	0.3	3.2	1.2	4.1	600.0	3.4	25.5	0.3
σ, ppm	7.1	0.7	0.7	0.7	10.6	20.5	1.4	0.8	7.1	0.8	0.0	0.7	0.0	0.4	113.1	1.0	4.9	0.0
REF (RDF)	1.3	(1.1)	1.5	(1.1)	1.3	1.4	1.9	5.2	1.9	3.5	(1.5)	1.3	1.8	(1.2)	1.3	2.0	(1.1)	(1.3)
Soils on Triassic volcanic suit and subvolcanic intrusions (n=4)																		
Average, ppm	165.0	47.0	16.0	23.5	50.8	80.5	8.5	2.6	340.0	2.9	0.2	3.1	0.8	3.3	792.5	5.4	23.8	0.3
σ, ppm	45.1	6.5	6.9	6.1	21.1	12.7	3.6	0.8	72.1	1.2	0.0	0.8	0.3	0.8	125.3	4.5	4.8	0.2
REF (RDF)	1.3	(1.3)	1.4	(1.2)	1.3	1.2	1.2	6.0	1.9	2.7	(1.7)	1.2	1.2	(1.5)	1.7	3.2	(1.1)	(1.2)
Soils on granodiorite and granite suites of Permian Selenga complex (n=13)																		
Average, ppm	172.3	56.8	17.2	30.3	42.8	98.0	8.3	2.7	410.0	1.9	0.3	3.0	1.1	4.6	550.8	2.5	20.9	0.3
σ, ppm	42.3	19.9	5.1	8.8	11.0	16.7	3.4	1.9	153.0	0.6	0.0	0.7	0.5	1.2	154.8	1.9	4.5	0.1
REF (RDF)	1.3	(1.0)	1.5	1.0	1.1	1.4	1.2	6.2	2.3	1.7	(1.6)	1.2	1.6	(1.1)	1.2	1.4	(1.3)	(1.7)
Soils on gabbro and diorite suites of Permian Selenga complex (n=5)																		
Average, ppm	170.00	47.40	15.00	27.80	54.25	89.60	9.88	3.00	410.00	5.56	0.23	2.62	0.96	3.40	586.00	1.84	19.20	0.23
σ, ppm	30.00	18.90	2.45	6.10	21.85	15.77	5.47	1.41	47.43	6.46	0.04	0.45	0.54	0.75	99.65	0.56	2.39	0.06
REF (RDF)	1.32	(1.26)	1.33	(1.04)	1.39	1.28	1.45	6.82	2.34	5.05	(1.80)	1.05	1.44	(1.49)	1.27	1.08	(1.41)	(1.84)
Background soils, total population (n=32)																		
Average, ppm	162.9	51.0	15.9	26.7	46.6	89.2	9.7	2.8	358.8	3.3	0.2	2.9	1.1	4.0	620.9	3.0	22.4	0.3
σ, ppm	37.1	15.8	4.5	7.5	133.9	21.7	5.0	2.7	149.6	7.2	0.0	0.6	1.4	1.1	143.1	3.2	6.4	0.2
REF (RDF)	1.26	(1.17)	1.41	(1.09)	1.20	1.27	1.41	6.25	2.05	3.00	(1.65)	1.16	1.64	(1.28)	1.35	1.78	(1.22)	(1.51)

n the number of samples, *REF* regional enrichment factors, taking into account average levels of the elements in uncontaminated world *C*, in brackets, *RDF* regional depletion factor

typical for deposits from the Permian age. The second two are from vehicle emissions.

The lowest HM and metalloid levels were in the *ger* area. This may be due to its greater distance from the ore field (5–7 km) and favorable position with respect to prevailing west and southwest winds. For other elements, geochemical differences between the urban and background soils were insignificant.

Comparing the geochemical characteristics of urban and reference soils revealed that the former were rich in ore-related elements (Mo and Cu) as well as HMs released through combustion of fuel, coal, and domestic waste (Bi, Cd, Pb, and Zn). However, the main source of high Se, Mo, Sr, W, Cu, and As concentrations in background soils from the study area was natural metallogenic specialization of parent rocks.

The cumulative technogenic impact on soil cover in Erdenet was assessed using the Integrated Pollution Index Z_c. It was estimated for each sampling site with attention to the geochemical heterogeneity of parent rocks. Its average value in urban soils was 17.9, which indicates moderate contamination. The industrial zone—encompassing the production facilities, pit, waste rock pile, and tailing area—showed especially high contamination (Z_c=74.8). The other land-use zones were either “uncontaminated” (Z_c<8) or low in cumulative pollution (Z_c<16).

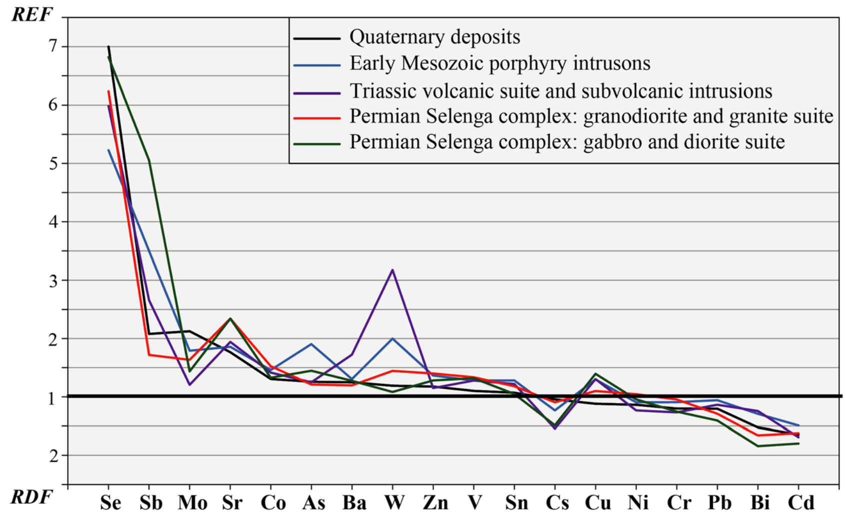


Fig. 2 Regional enrichment (REF) and depletion (RDF) factors for topsoil reference samples derived from different parent rocks

4.3 The associations of HMs and metalloids in urban soils

The elements with similar spatial distributions in soil cover, with common accumulation or depletion patterns, are incorporated into three stable paragenitic associations: Cu–Mo–As–Sb–Bi, V–Co–Cr–Ni, and Zn–Cd–Pb–Sn, obtained through hierarchical cluster analysis. The first association includes chalcophile elements found naturally in sulfide minerals (Greenwood and Ernsho 1997), which are abundant in the ore bodies and host rocks of the Erdenet complex.

The second association consists of four elements. The highest correlation was between Cr and Ni (r=0.79). Cr has the potential to form soluble organic complexes. Ni assumes its simple cation form under normal soil conditions, but it can also generate complex ions and associate with organic matter in topsoil horizons (Perel'man and Kasimov 1999). The other two elements—V and Co (r=0.65)—exhibit very different redox behavior. However, like Cr and Ni, they are siderophile elements and showed similar distribution among parent rocks in the study area. This is because the dominant rocks in the first stage of Selenga complex formation were amphibole gabbro and diorites rich in V–Co–Cr–Ni.

The third association is made up of chalcophile elements (Greenwood and Ernsho 1997) concentrated in granites and sienogranites (Gavrilova et al. 2010) that are ubiquitous within the study area. Combustion of gas and coal along with contamination linked to domestic waste from a landfill are other contributing factors. The latter is particularly important for Cd and Sn distribution. Among the elements in this group, redox behavior is very similar: weakly mobile in oxidizing and reducing environments, immobile in sulfidic environments.

Paragenetic associations involved 13 of the 18 elements studied. Their origins are related to lithologic factors (distinct parent rocks with petrochemical characteristics typical for the Erdenet ore complex) and anthropogenic factors such as emissions from petrol and brown coal combustion.

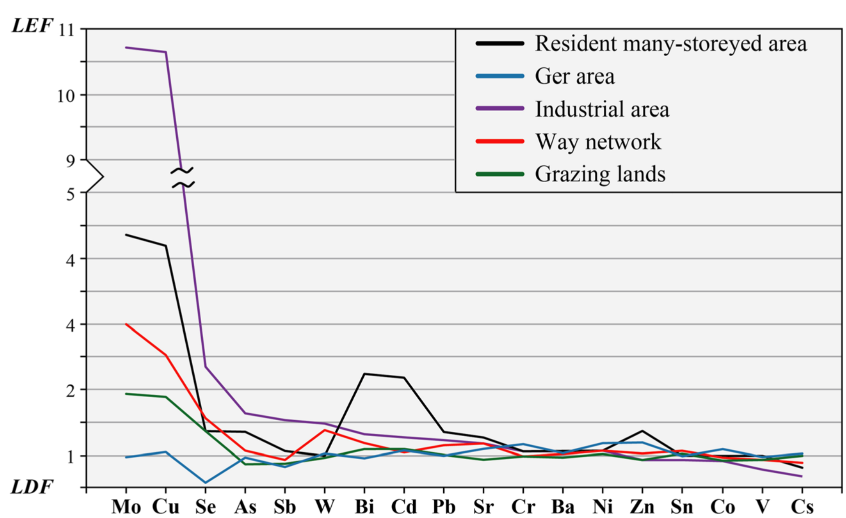


Fig. 3 Local enrichment (LEF) and depletion (LDF) factors for urban topsoil samples located in different land-use zones of the city

4.4 Spatial patterns in element distributions

HM and metalloid associations showed highly analogous spatial distributions in urban soil cover. The elements of the first association (Cu–Mo–As–Sb–Bi) were in two accumulation zones: one in the northern part of the city and one in the eastern part. The northern zone, with local enrichment factors ranging from 1.7 (Bi) to 56.8 (Mo), was restricted to the tailing area. It originated and is gradually expanding due to waste disposal from EMC. The eastern zone is an anomaly, with higher element concentrations characteristic of the processing plant territory. The levels of Cu and Mo in surface soil horizons were, respectively, 982 and 460 times higher than the reference values; As, Sb, and Bi levels were 44, 42, and 12.6 times higher. This implies that the anomaly is due to atmospheric impacts from large quantities of dust produced through grinding and transport operations.

Relatively high concentrations of all elements except Cu–Mo and Zn–Cd–Pb–Sn were found in the city center. Local enrichment factors varied from 1.3 (Sb and Zn) to 4.7 (As) on the northern boundary of the thermal power plant. Elements are released from a huge pile of coal that serves as fuel for the city. This coal from the Baganur strip mine is enriched with As (REF=17.1), Sn (9.9), Zn (7.8), Pb (6.1), Sb (4.2), Cd (3.2), and Bi (2.3) (Ketris and Yudovich 2009). Pollution is linked to the transfer of coal material via surface runoff, linear erosion, and deflation. The same type of anomaly, though with lower HM and metalloid levels, occurs in the southeast *ger* district, where coal is used for heating homes.

The elements of the third association accumulate in the multistory residential area, where local enrichment factors varied from 1.3 (Cd) to 3.1 (Pb). Soil pollution in this area is likely due to vehicle emissions. The city landfill is another highly polluted area, with concentrations of Sn, Cd, Zn, and Pb that were, respectively, 2.1, 2.0, 1.7, and 1.4 times higher than in reference soils.

In comparison with the first and third associations, elements in the second association (V–Co–Cr–Ni) showed much less variance and were more similar to the background samples. Their levels were lower in the tailing zone than in average world soils, which implies reduced concentrations in the porphyry complex of the ore deposit and depletion in regional soils (RDF for Cr=9.2, V=4.2, Ni=3.0 and Co=1.5).

Analysis of Z_c values (Fig. 4d) revealed that 50 % of the study area can be classified as "uncontaminated" (Z_c<8) while 16 % shows low contamination (Z_c varies from 8 to 16). The eastern part of the city, where the industrial zone is located, had a very high level of soil cover contamination (Z_c ranging from 64 to 128). The most distinct anomalies were in the tailing area and at the Erdenet and Erdmin plants (with local maximum Z_c reaching 1,560).

4.5 The impact of natural and anthropogenic factors on HM and metalloids concentrations in urban soils

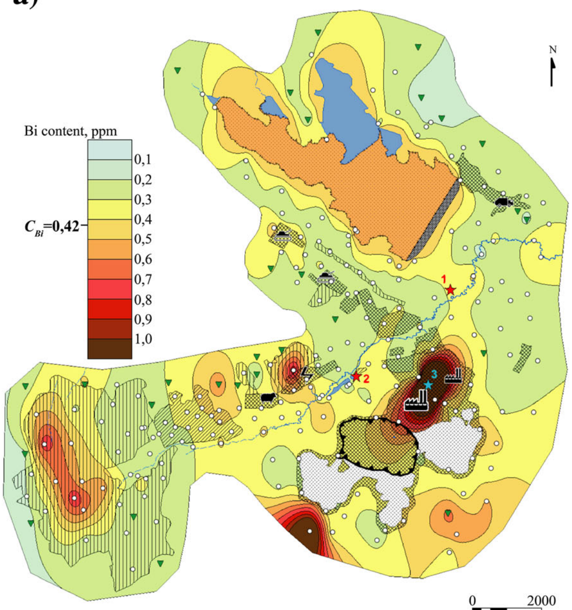
Multivariate regression analysis revealed the natural and anthropogenic factors responsible for spatial differences among HM and metalloid concentrations in soil surface horizons. The impact of the following factors were analyzed: (1) land-use assignment of territories corresponding to different anthropogenic loads, (2) parent rock types that give rise to natural geochemical soil heterogeneity, (3) elevations that indicate the position of sites within the landscape-geochemical (relief) system, and (4–10) various physical and chemical properties that may influence the mobilization or retention of elements, including soil solution reaction; levels of Al, Fe, and Mn oxides; humus content; and soil granularity (proportion of sand and clay).

Concentrations of Cu, Mo, As, Sb, Zn, Cd, and Pb were strongly influenced by technogenic factors while levels of Ni, Bi, Ba, W, Se, V, Co, Sn, Cr, Cs, and Sr were based on soil properties (Table 3). For the first group of elements, increased technogenic loads in the industrial and residential areas accompanied 1.4- to 5-fold growth of element concentrations in comparison with other land-use zones. For several decades, the Erdenet processing plant, the tailing area, and the thermal power plant have been sources of Cu, Mo, and As. Disproportionately high concentrations of Zn, Cd, and Pb are linked to vehicle emissions and domestic waste combustion. Within the land-use zones in each case, accumulation of elements was conditioned by soil properties such as pH (for Cd and Pb), humus content (for Zn), levels of Fe and Mn oxides (for As, Cu, Mo, As, and Zn), and proportion of clay fraction (for Cd). Concentrations of Sb depend on elevation and parent rock type.

The second group of HMs and metalloids is associated mainly with Fe oxides (Ni, Bi, Ba, W, and Se), Mn oxides (V, Co, and Sn), and Al oxides (Cr and Cs). The relationship between oxide content and element concentration is positive. Enrichment of soil samples with oxides results in a 1.2- to 1.4-fold increase in metal concentrations and a 2.6-fold increase in Se concentrations. Trace element accumulation often occurs in Fe- and Mn-rich nodules (Vodyanitskyi 2014) due to intense sorption and co-precipitation with oxide phases. It is found that the Fe oxides are added to soils from the waste rock pile produced by the "Erdenet" processing plant. This finding coincides with the data reported by other authors (Durov et al. 2009).

Fig. 4 Distribution of Bi (**a**), As (**b**), Ni (**c**) and index of total pollution (**d**) for topsoil samples. At the scale, C_i is element clark (Kabata-Pendias 2011). **Highlighted in red* element concentrations in excess of clark ▶

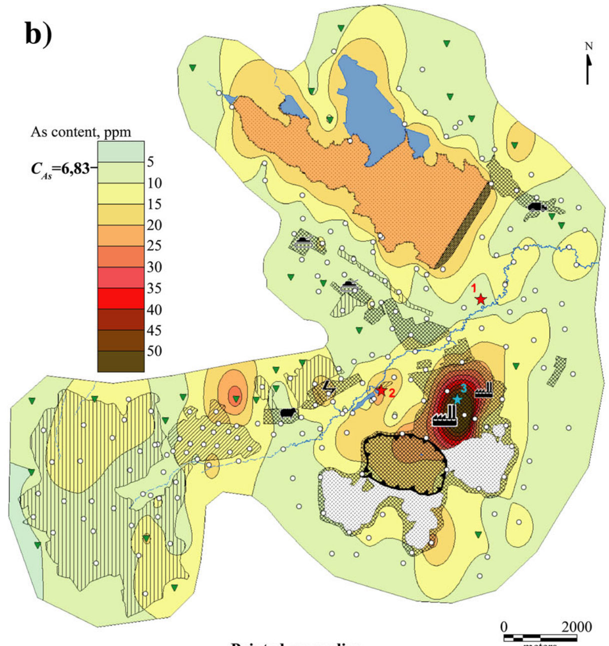
a)



Pointed anomalies

☆	Location	W	Bi	Cd	Zn	Sn	Pb
1	Municipal drainage basin	7,9*	3,0	1,6	770	9,2	84,0
2	Thermal power plant waste pile	39,0	1,5	0,5	96,0	5,1	24,0
3	Processing "Erdenet" Plant	2,8	3,2	2,7	500	2,6	52,0

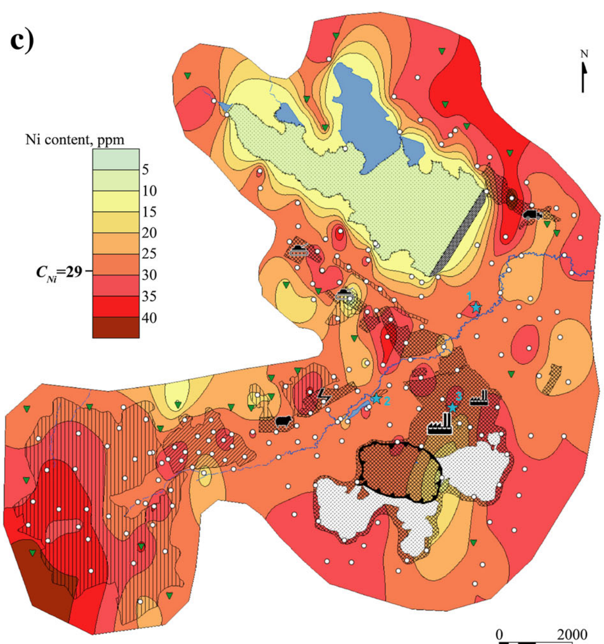
b)



Pointed anomalies

☆	Location	Cu	As	Sb	Mo
1	Municipal drainage basin	660	5,4	3,4	130
2	Thermal power plant waste pile	770	110	1,9	23,0
3	Processing "Erdenet" Plant	42000	350	870	48,0

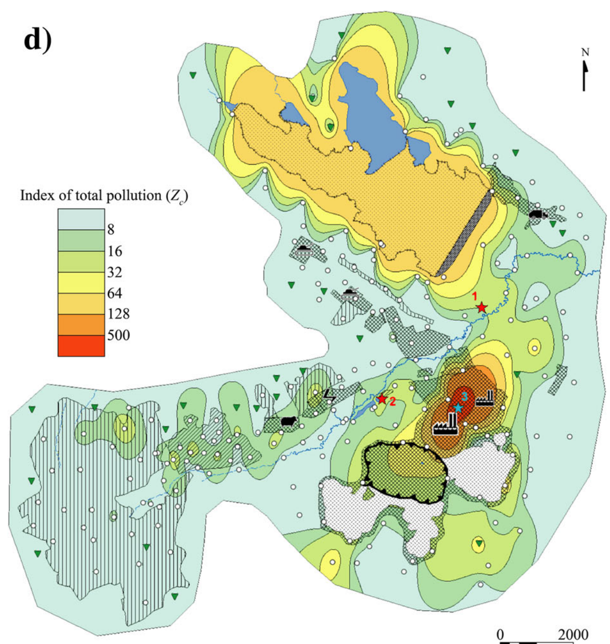
c)



Pointed anomalies

☆	Location	V	Co	Sr	Cr	Ni
1	Municipal drainage basin	74,0	9,7	270	67,0	31,0
2	Thermal power plant waste pile	190	31,0	740	62,0	48,0
3	Processing "Erdenet" Plant	130	34,0	420	42,0	34,0

d)



Pointed anomalies

☆	Location	Z_c	*IIR*
1	Municipal drainage basin	111	209
2	Thermal power plant waste pile	56,6	108
3	Processing "Erdenet" Plant	1559	2102

Table 3 The significance of anthropogenic and natural factors in HMs' and metalloids' distributions in the surface soil horizons of Erdenet

Factors		Element group I							Element group II											Sr	Zc
		Cu	Mo	As	Sb	Zn	Cd	Pb	Ni	Bi	Ba	W	Se	V	Co	Sn	Cr	Cs			
Functional zone		**1**	**1**	**1**	**1**	**1** **3**	**1**	**1**		**5**	**3**	**3**	**2**	***3*** ***4***			**4**			**1** **6**
Parent rocks		4			2		5				3			3						
Elevations				**3** ***4***	**2** ***3***			**4**			**3** **4**	***3***		***3*** ***4***		***3***	**3**	**2** **5**		***6***
Soil physical and chemical properties	pH	***5***	***3***				***2***	***2***		***3***			**3**			***2*** **4**		***3***	**1** ***3***	
	Humus content, %		***3***		***3***	***2*** **4**	**3** **4**	***3***	***4*** ***5***	***3***							**3**	***4***	***2***	**7**
	Al_2O_3, %			**3**	**4**		**5**		**1** **5**	**1**	**1** **2**	**1** **2** **4**	**1** **4**		**2**		**2** **4**			**2** **3** **4**
	MnO, %	***2*** ***3***	***2***	***2*** ***4***		**2**	**4**	***2***	**2** **5**	***4***			***3***		**3**	**3**	**1**	**1** **3**	***4***	***2*** ***3*** ***5***
	Fe_2O_3, %	**4**	***4***	**2**				***3***	**4**	***2***		***2***	**3** **5**	**1** **2**	**1** **2** **4**	**1** **2**	**2**		**2** **5**	***3***
	clay, %			***5***			**2** **3**	**3**							**3**		**3**		***3***	
	sand, %			***3***		**3**			***3***	***2***					***5***		**4**	***2***	**4**	**3**

The range of values from 1 to 7 indicate the decrease in the factor significance, and the additional signs such as «**1**» or «***1***» show respectively positive or negative relationship

The only element not part of an association was Sr, because its distribution in the surface horizons of urban soil is largely dependent on soil reaction and less influenced by factors such as humus, Fe oxide, and clay content. The relation with pH is positive, confirming substantial accumulation of this element in alkaline soils.

The Integrated Pollution Index Z_c is dependent on anthropogenic factors expressed by assigning each territory to a specific land-use zone. Increased technogenic loads found in the industrial zone were accompanied by a higher Z_c value. Levels of Al, Fe, and Mn oxides were of less importance. Maximum pollution was found in samples collected in the tailing area and in the industrial grounds of the Erdenet processing plant, where Al_2O_3 was above 14.75 % and MnO was below 0.087 %. Such oxide proportions are typical for rocks in the Erdenet ore complex that are high in Al_2O_3 and low in MnO (Gavrilova et al. 2010).

4.6 Assessment of environmental risks associated with soil cover pollution in Erdenet

Comparison between the measured concentrations of V, Cr, Co, Ni, Cu, Zn, As, Se, Sr, Mo, Cd, Sn, and Pb in soil surface horizons and the data on the elements' maximum permissible concentrations (MPC), taking into account soil granulometry and specified in Mongolia Soil Quality Standards, evaluates the potential environmental risk that may exist as a result of soil cover pollution. The granulometric analysis of the samples showed that surface soil horizons in most cases have loamy texture except of technogenic layers (with higher content of sand) sampled at the surface in the tailing area.

Comparison of V, Cr, Co, Ni, Cu, Zn, As, Se, Sr, Mo, Cd, Sn, and Pb concentrations in surface horizons with data on Maximum Permissible Concentration (MPC) for such elements—with attention to granulometry and Mongolia Soil Quality Standards (Х рсний 2008) —illuminates potential environmental risk due to soil cover pollution. Granulometric analysis of samples showed that most surface soil horizons have a loamy texture, except technogenic layers (with higher sand content) sampled in the tailing area.

Calculating ratios of HM and metalloid concentrations to MPC ($K_o = C_a$/MPC) revealed that the most polluted sites were in the industrial zone (Fig. 5). Average K_o values in this area for Mo, Cu, and As were 8.6, 5.7, and 3.8, respectively. Maximum K_o values were calculated for Cu (525), Mo (290), and As (218). Each site with an extremely high K_o value was located at the Erdenet processing plant.

All land-use zones were characterized by high K_o values for As (2.6–3.0). The distribution of K_o estimates for Mo and

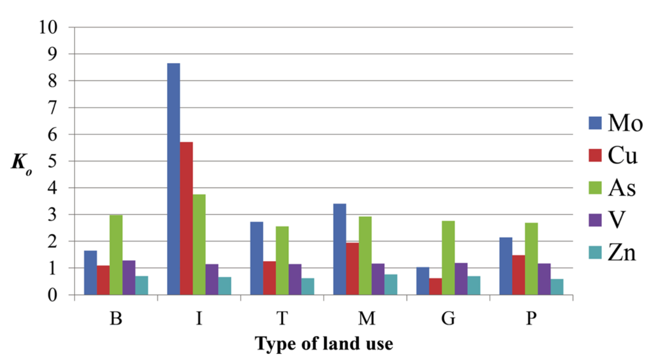


Fig. 5 The ratios (K_o) between the observed concentrations in for soil samples and *MPC* in reference sites (*B*), industrial area (*I*), way network (*T*), resident many-storeyed area (*M*), *ger* area (*G*), grazing lands (*P*)

Cu depended on the degree of anthropogenic load: Maximum values were found in the multistory residential area (3.4 and 1.9), and minimum values in the *ger* area (1.03 and 0.62). Concentrations of Zn, Cr, Sr, Se, Pb, Co, Ni, Cd, and Sn in the surface horizons of urban soils did not exceed MPCs, and K_o varied within 7–77 % of MPC as required by Mongolian law. There was, thus, no significant environmental risk associated with these elements.

Because environmental contamination is multivariate, an Integrated Index of Risk (IIR) was used to incorporate a variety of pollutants in site assessment. IIR analysis revealed variation from 0.4 to 2012, with an average of 45. The spatial distribution of IIR was similar to that of Integrated Pollution Index Z_c (Fig. 4d). Maximum values were identified at the Erdenet processing plant, while minimum values were in the *ger* area and pastureland. The clear difference between IIR and Z_c distributions was that the former showed not low but acceptable environmental risk (IIR=16–32) in the multistory residential area. Higher contamination was found near waste rock piles between the Erdenet processing plant and the tailing area in the Erdenetii-Gol river valley. Figure 6 displays proportions of the land-use area with different classes of contamination risk. As represented in this diagram, environmental risk from soil pollution was low (IIR<16) in 39 % of the total area, acceptable (IIR=16–32) in 26 %, moderate (IIR=32–64) in 15 % and high (IIR>64) in nearly 20 %.

The above findings justify the need for remedial action on 1/5 of the total area of Erdenet (S~36 km^2) where there are high levels of multi-element contamination in soil surface horizons. Special attention should be paid to the Erdenet processing plant and the Erdmin plant, since the people who work there are consistently exposed to the harmful influence of high HM and metalloid concentrations.

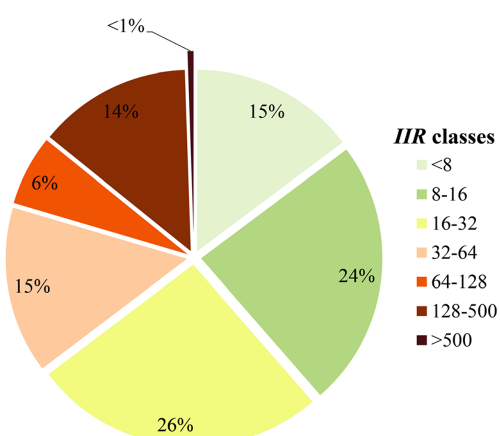


Fig. 6 The proportions of the area with different classes of the integrated index of environmental risk (IIR)

5 Conclusions

1. HM and metalloid concentrations in background soils correspond with the geochemical features of underlying parent rocks. Compared with average levels of the elements in uncontaminated world soils, all reference samples were rich in Se (5.2–7), Mo (1.7–5.1), Sb (1.2–2.4), and Sr (1.8–2.3). Additionally, higher levels of W and As have been recorded in soils associated with Early Mesozoic porphyry intrusions, and higher levels of W and Ba have been found in soils derived from Triassic volcanic suite and subvolcanic intrusions. The variability of V, Cr, Co, Ni, Zn, Cd, Sn, Cs, Ba, and Pb concentrations is rather low (C_v<31 %). By contrast, Cu, Mo, Sb, and W are distributed unevenly to an extreme, C_v=107–287 %. Highest levels of V, Cr, Co, Ni, Zn, and Sr occur in soils associated with granodiorite and granite suites of the Permian Selenga complex, and lowest concentrations are in soils derived from Triassic volcanic suite and subvolcanic intrusions.
2. In all Erdenet land-use zones except the *ger* settlement, upper soil horizons were contaminated with Mo, Cu, and Se. These elements were classified as first-order contaminants in the study area. The highest levels of contamination were in the industrial zone, where concentrations of Mo–Cu–Se–As–Sb–W were, respectively, 10.7, 10.6, 2.3, 1.6, and 1.5 times higher than in the reference samples, and where the Integrated Pollution Index Z_c was 74.8. Evaluation of cumulative technogenic impact on Erdenet soil cover based on Z_c analysis revealed two distinct anomalies: One was restricted to the Erdenet processing plant (where average Z_c=106 and maximum Z_c=1,560), and another was in the tailing zone (where Z_c=109).
3. Three stable associations of elements (Cu–Mo–As–Sb–Bi, V–Co–Cr–Ni, and Zn–Cd–Pb–Sn) were identified in the urban soil surface horizons. Spatial distribution of elements occurred in similar patterns. The origins of these associations are closely related to petrochemical characteristics of the Erdenet ore field and technogenic influence. The first and third associations represent distinct anomalies within the industrial zone. However, the third shows homogeneous spatial distribution of elements (C_v= 18–24 %) and minimum load in the tailing zone.
4. The anthropogenic factor, expressed by assigning land-use zones, was most influential on spatial patterns of HM and metalloid distribution. Of secondary importance was the retention capacity of different soils. Other natural landscape–geochemical features of the study area (parent rocks and position in the relief) appeared to be of less importance.
5. The highest K_o ratios between metal concentrations and Mongolian sanitary standards were found in the industrial zone. For Mo, Cu, and As, average K_o values were 8.6,

5.7, and 3.8, respectively. Integrated Index of Risk (IIR) calculations showed high environmental risk from soil pollution in over 35 km^2 of territory—approximately 1/5 of the city. Maximum IIR values (>500) were found at the Erdenet processing plant. These results underscore the need for remedial action to reduce negative impacts of HMs and metalloids on workers' health. Such remediation should be aimed at diminishing concentrations of toxic HMs and metalloids in soils.

6. Our study confirms the utility in applying integrated indices Z_c and IIR to evaluate cumulative technogenic impact on urban soils in mining regions. The Integrated Pollution Index Z_c highlights the degree of geochemical transformation of soil cover induced by anthropogenic factors, and the IIR gauges risk associated with multi-element pollution in conditions with high background levels. IIR revealed distinct spatial patterns in this case, helping to prioritize large concentrations of the most toxic elements as first-order hazards.

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Internet-resources

www.erdenetmc.mn–the official site of Erdenet mining Corporation

www.worldweather.wmo.int–World Meteorological Organization