

# Lead Isotope and Trace Element Composition of Urban Soils in Mongolia<sup>1</sup>

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**Abstract**—Lead (Pb) pollution in and around Ulaanbaatar is of national concern, given that the Mongolian capital is home to nearly half of the country's entire population. By comparison, Mongolian countryside is a pristine environment because of its sparse population and low industrial activity. The concentration of Pb in urban soils (average of 39.1 mg kg<sup>-1</sup>) was twice the values found (average 18.6 mg kg<sup>-1</sup>) in background territories (i.e., Mongolian rural sites). Furthermore, Pb contamination was examined by using Pb stable isotopic composition, and covariance of Pb isotopic ratios showed two groups between rural and urban soils as pristine and disturbed sites. The <sup>206</sup>Pb/<sup>207</sup>Pb ratio, the most prominent fingerprint for Pb pollution, was 1.163–1.185 for the urban whereas values for rural soils (1.186–1.207) were analogue to the regional Pb isotopic signatures. Local coal sources and their combustion products, one of the potential Pb pollution sources in Ulaanbaatar, have significant radiogenic properties in terms of Pb isotopic composition and revealed an average of 1.25 for <sup>206</sup>Pb/<sup>207</sup>Pb and 19.551 for <sup>206</sup>Pb/<sup>204</sup>Pb ratios. Thus, contributions from coal firing activity to Pb pollution lower than it was assumed, and smaller range of these values measured in urban soils may be attributed to the mixing of less radiogenic Pb as a constituent of the leaded gasolines.

**Keywords:** lead isotope, multi-collector inductively coupled plasma mass spectrometry, soil, Ulaanbaatar

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## INTRODUCTION

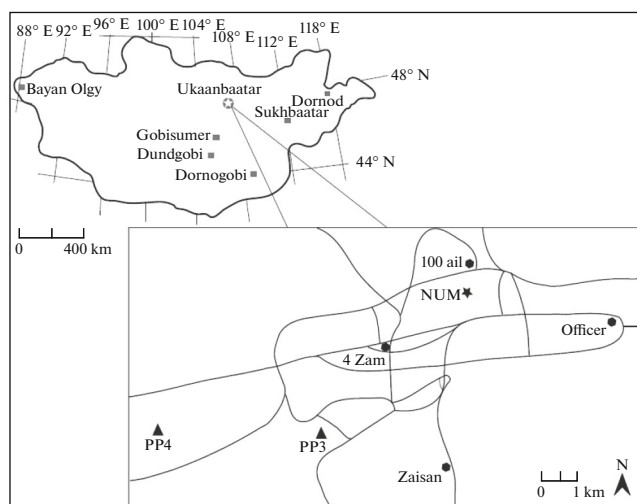
Because of the geological settings in Ulaanbaatar the elevated levels of heavy metals in its soil environment are not necessarily associated with anthropogenic sources. The city is located within a mineralized zone belonging to the Early Mesozoic Khentiy gold- and rare metal-bearing belt that is rich in tin (Sn) and tungsten (W) [15]. Accordingly, Ulaanbaatar soils were enriched in Sn [12]. Accumulations of some toxic and heavy metals from anthropogenic sources have also been measured in the atmosphere and topsoils within the Ulaanbaatar city [12, 30]. The sources of Pb pollution are multifaceted. However, automobile exhaust gases and emissions from coal combustion are the most prominent in the Ulaanbaatar environment. Further, the moderate pollution of Ulaanbaatar soils by Pb, coupled with the high Pb contamination of the city's atmosphere, was indicative for atmospheric transport as described by Sorokina and Enkh-Amgalan [30].

Atmospheric Pb is associated with sub-micrometer size whose airborne period increases with decreasing particle size. An example of atmospheric Pb transport

was the observation that the Pb isotopic composition in European aerosols was influenced by Pb carried by dust particles from the Sahara desert [9]. Likewise, aerosols from Asia have a marked influence on the chemical composition of the North Pacific atmosphere [8] since the Asian continent is a major source of natural dust and pollution [28].

Sources of Pb, such as lead-bearing products and ores, have a specific isotopic ratio, and only weak mass fractionation of Pb isotopes occurs through physicochemical and biological processes during transport [4, 14, 26]. Thus, the magnitude of this ratio can serve as a signature in tracing the sources for Pb pollution. Compositional variations can be found for the radiogenic <sup>206</sup>Pb, <sup>207</sup>Pb and <sup>208</sup>Pb isotopes, produced from the radioactive decay of <sup>238</sup>U, <sup>235</sup>U and <sup>232</sup>Th, respectively, whereas the abundance of <sup>204</sup>Pb remains constant at 1.4% [18]. Isotopic ratios can also vary according to the environmental input of Pb from anthropogenic sources. Literature on Pb isotopes in the environment has been reviewed by Komárek et al. [14], and also source signatures of Pb isotopes in the atmosphere over the Northern Hemisphere were published by Bollhöfer and Rosman [4].

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**Fig. 1.** Map showing soil sampling sites in rural and urban Mongolia. Enlarged inset shows the location of the 4 Zam, 100 Ail, Officer Palace and Zaisan sites, and lines signify main roads in Ulaanbaatar. The filled circles show the sites of soil sampling, while a star indicates aerosol sampling location, and triangles signify Thermal Power Plants PP3 and PP4.

It has been speculated and investigated, if the Gobi desert in southern Mongolia is one of the source regions to the Chinese Loess Plateau; therefore, the Pb stable isotopic approach suggested that southern Mongolia is the main source region together with northern China to the loess [34]. Moreover, Pb isotopic ratios preserved in different size fractions of the loess were an essential characteristic to trace the origin of this natural deposition.

Since information about stable Pb isotopic composition of environmental and source-related materials in Mongolia is limited, it is difficult to develop strategies for mitigating the risk of Pb contamination to the environment and human health. Therefore, one key objective of this research was to determine and report reliable information on trace elemental distribution and Pb stable isotope abundance in Ulaanbaatar soils. The isotopic compositions besides serving as a fingerprint for pollution source this parameter can provide an insight into contaminant distribution at the local, regional, and global scale.

## MATERIALS AND METHODS

**Soil sampling and source related materials.** Ulaanbaatar, the capital city of Mongolia, is home to 1.3 million people that account over 43% of the Mongolian population. The city is located on the banks of the Tuul River at an average altitude of 1350 m, occupying an area of ca. 4700 km<sup>2</sup>. The average annual evaporation is large and annual precipitation equals 267 mm. Random sampling was carried out in December 2012, when the ground was frozen and an

ambient daily mean temperature was  $-25^{\circ}\text{C}$ . Surface soils were taken from a depth of 0–5 cm, whilst sub-surface soils were collected at a depth of 5–8 cm and were collected from sites adjacent to the heavily used arterial roads in Ulaanbaatar, except the Salkhit site that is located about 18 km in the north of the city center. In Fig. 1, enlarged inset showed main roads and sampling locations, including the sites Baruun 4 Zam (Bayangol district), Officer Palace (Bayanzurkh district), Zaisan (Khan-uul district) and 100 Ail (Chingeltey district). 100 Ail site was characterized by densely distributed traditional dwellings as well as small family houses, and also construction material markets.

For Pb stable isotopic analysis, a total of 23 soil samples from urban (Ulaanbaatar) and rural areas were studied. Figure 1 showed a sketch map of the sampling sites for both rural and urban areas. The samples of 13 rural soils were collected from six different provinces covering western, central, southern, and eastern parts of Mongolia.

Isotopic composition of Pb in Mongolian raw coal and fly ash emitted by Power Plant 3 (PP3) and Power Plant 4 (PP4) were determined as pollution source related materials. PP3 and PP4 are the largest stations nationwide using brown coal from the Baganuur and Shivee-Ovoo deposits as their basic fuel. The fly ash samples from both power plants were taken from a furnace, declared to be fueled by Baganuur coal only. Atmospheric total suspended particulates (TSP) in Ulaanbaatar were sampled during winter between November, 2013 and January, 2014. TSP were collected on polycarbonate membrane filters (Whatman. Nuclerpore, Track-Etch Membrane, 0.4  $\mu\text{m}$ ) with flow rate of 1 L min<sup>-1</sup> using a Dust track-DRX aerosol monitor, operated by Prof. Lodoysamba at National University of Mongolia. Eight filter samples in total and were separated into two groups; therefore, each set consisted of 4 filter samples. The concentration of Pb was higher than a detection limit for ICP-MS isotopic analysis.

**Determination of soil properties, elemental and mineralogical composition.** The urban soils were air-dried, passed through a 2 mm sieve, and ground in an agate mortar and pestle. The soil pH was measured using an aqueous slurry (soil : water = 1 : 5) and a Sartorius Professional PP-50 pH meter. The soil water content was derived from the weight loss after oven-drying at 105°C. Total organic carbon (TOC) and total carbon (C) contents were determined using an element analyzer (PE2400 II, USA). The TOC was measured after removing carbonates with dilute HCl. The TOC was converted to total organic matter (TOM) on the assumption that organic matter contains 58% organic C. The inorganic C content was estimated by subtracting the TOC content from the total soil carbon.

Range of major and trace element (Al, Cl, K, Mg, Na, Sb, U etc.) concentrations were measured by

instrumental neutron activation analysis (INAA) using thermal neutrons as described elsewhere [24]. Determination of elements Cu, Ga, Ge, Mo, Nb, Sn, Pb and Tl was carried out by means of inductively coupled plasma mass spectrometry ICP-MS (Platform ICP, Micromass Instruments Corporation) after a total acid digestion. Analytical accuracy was estimated by analyzing Certified Reference Materials (soil GBW-07408 and sediment GBW-07305 from the Institute of Geophysical and Geochemical Exploration, China). Every sample was analyzed in duplicates. The mineralogical composition of the soils (oven-dried, ground, and passed through a 200 mesh sieve) was determined by X-ray diffraction (XRD) using a D/Max2200 diffractometer (Japan), and nickel filtered  $\text{CuK}\alpha$  radiation ( $\lambda = 1.54178 \text{ \AA}$ ). The XRD patterns were recorded in the  $2\theta$  range of  $3^\circ$ – $60^\circ$ , using a step-scanning mode with a step size of  $0.04^\circ$ , and a counting time of 5 s per step. Amorphous and crystalline mineral phases were identified, and their contents were calculated using a JADE 5 software. The crystalline phases in the samples were determined by comparing their XRD patterns with those of various mineral standards. The K-value method was employed to determine quantities of soil minerals [20].

**Pb isotope analysis.** A total of 23 samples were analyzed for stable Pb isotopic composition in a clean laboratory as described by Li et al. [17]. To this end, 200–300 mg of oven-dried and ground (200 mesh) soil samples were digested with concentrated acids ( $\text{HCl} : \text{HNO}_3 = 3 : 1$ ) and addition of HF in capped perfluoroalkoxy (PFA) tubes (SAVILEX). The coals with and without combustion (at  $600^\circ\text{C}$ ), fly ash (as received from the power plants), and aerosol filter samples were digested in the same manner. MilliQ-water (18M $\Omega$ m) and sub-boiled acids were used throughout digestion and separation (explained below) procedures. The digestion was performed on a hot plate at a temperature of  $100$ – $110^\circ\text{C}$  for about 4 days with periodic stirring. The leachate in the PFA tube was evaporated to dryness, 1 mL of concentrated HCl was added, and again evaporated to dryness. This procedure was repeated after adding 1 mL of 2M HCl to the residues, and dissolving in 2 mL of 2 M HCl : 1M HBr (2 : 1) solution. The solid phases were separated from leachate by centrifugation at 3000 rpm for 25 min.

Separation of Pb from the matrix was carried out using a preconditioned Dowex  $1 \times 8$  resin (200–400 mesh) which was inserted into a pre-cleaned glass column and washed three times with concentrated HCl, followed by water. Then, the centrifuged soil leachates were loaded into the column and eluted at an average flow rate of  $1 \text{ drop} \times 1.5 \text{ min}^{-1}$ . Matrix related impurities were washed from the resin first with 1M HBr and subsequently by 2M HCl. Finally, the Pb was eluted with 1.5 mL 6M HCl into polytetrafluoroethylene (PTFE) vessels and dried at  $100$ – $110^\circ\text{C}$ . The dry residues were dissolved in a  $\text{HNO}_3 : \text{HCl}$  (1 : 3)

mixture and again evaporated to dryness. This procedure was repeated using 2 drops of  $\text{HNO}_3$ . The dry residues were dissolved in 2%  $\text{HNO}_3$  solution and analyzed for Pb isotope ratios using a multi-collector inductively coupled plasma-mass spectrometer (MC-ICP-MS; Nu Plasma, Nu Instruments Ltd). The instrumental mass fractionation of Pb was corrected by adding thallium (Tl) spike Tl : Pb (1 : 5) as an internal isotopic standard (NIST SRM 997). The accuracy and reproducibility of measurements were controlled by analyzing the Pb isotope standard (NIST SRM 981) throughout the measurements with 5 sample intervals. To control leaching efficiency the Columbia River Basalt BCR-2 (U.S. Geology Survey) reference material was analyzed, yielding the following results:  $^{206}\text{Pb}/^{204}\text{Pb} = 18.761 \pm 0.030$ ;  $^{207}\text{Pb}/^{204}\text{Pb} = 15.639 \pm 0.041$ ;  $^{208}\text{Pb}/^{204}\text{Pb} = 38.757 \pm 0.108$ ;  $^{206}\text{Pb}/^{207}\text{Pb} = 1.200 \pm 0.001$ .

## RESULTS AND DISCUSSIONS

**Soil characteristics and elemental distributions.** The intermountain Tuul River valley is one the fertile regions in the Mongolian landscape. The urban soils from Ulaanbaatar city have an average of 4.6% of TOM contents (Table 1). Properties and composition of the rural soils can be found in Tserenpil et al., 2013. The TOM contents (2–8.2%) of randomly collected soils from the northern, southern, eastern and central parts of Ulaanbaatar were also higher than values previously reported by Kasimov et al. for soils along the Tuul River and its tributaries, but were similar in range to the TOM values (2–12%) for soils within city districts [1]. Background chestnut soils were also rich in organic matters (3.3–9.9% given as humus content) [10]. The soils analyzed by Kasimov et al. [12] contained 0.7–5.0% of  $\text{CaCO}_3$  and had a pH of 7.5–9.0. We measured a similar pH range (7.3–8.4) for the urban soils (Table 1), and their mineralogical composition indicate the abundant presence of carbonate minerals (Table 2).

Besides having high organic matter contents, the urban soils differed in their mineralogical composition from soils taken from the countryside. Although the mineralogy of the urban soils (as given in Table 2) was dominated by the presence of primary minerals (67.7–92.7%), contribution of the secondary minerals in making up soil structure was rather greater (in this case montmorillonite and kaolinite) and uniform (total of 6–16.6% except 1% content) in urban samples than those in the rural [31]. The total content of clay minerals was highest (16.6%) in the subsurface soil from the Officer Palace site, and lowest (1%) in the 100 Ail subsurface sample. It is noteworthy that all the soils collected within city districts contained carbonate minerals (i.e., calcite and dolomite); e.g., the surface soil from Salkhit contained as much as 15.3%. On the other hand, carbonate minerals were not detectable in

**Table 1.** Some properties of the urban soils from Ulaanbaatar city

	Sample designation	Moisture, %	pH	TOM <sup>a</sup> , %	TIC <sup>b</sup> , %	LOI <sup>c</sup> , %
1	4 Zam surface	10.3	7.9	3.1	1.1	92.1
2	4 Zam subsurface	2.9	7.9	3.8	0.5	92.4
3	100 Ail surface	2.9	8.4	5.3	0.9	88.3
4	100 Ail subsurface	3.9	8.2	6.1	0.9	88.6
5	Salkhit surface	10.5	8.1	4.6	1.0	91.4
6	Salkhit subsurface	11.0	8.0	4.0	1.3	90.6
7	Officer Palace surface	5.5	8.4	3.1	1.2	91.8
8	Officer Palace subsurface	2.6	8.1	2.0	1.9	91.9
9	Zaisan surface	9.7	7.3	8.2	0.9	87.1
10	Zaisan subsurface	5.0	7.7	6.1	0.5	89.8

<sup>a</sup> Total Organic Matter. <sup>b</sup> Total Inorganic Carbon. <sup>c</sup> Loss On Ignition at 550°C

**Table 2.** Mineralogical composition of soils in Ulaanbaatar

Sample designation	Quartz	Anorthite	Illite	Montmorillonite	Kaolinite	Hornblende	Calcite	Dolomite	Fe containing mineral
4 Zam surface	63.2	25.0	2.1	4.6	–	1.7	0.9	1.6	0.9
4 Zam subsurface	68.2	21.8*	2.8	2.0	1.5	3.1	+	+	0.6
100 Ail surface	52.8	29.4	2.9	2.4	1.7	0.7	8.2	1.2	0.9
100 Ail subsurface	68.3	24.3	–	1.0	–	–	5.7	+	+
Salkhit surface	53.0	19.4	2.5	4.2	4.6	1.1	14.3	1.0	+
Salkhit subsurface	52.1	32.4*	4.6	4.9	+	2.6	2.4	–	1.0
Officer Palace surface	54.7	14.5	6.6	5.7	4.0	2.2	8.8	–	2.3
Officer Palace subsurface	51.7	16.0	4.8	8.5	3.2	2.2	8.2	3.8	1.6
Zaisan surface	43.2	45.8	1.6	3.0	1.3	2.0	1.2	0.8	1.0
Zaisan subsurface	65.6	11.4	6.2	3.5	4.9	2.6	4.8	+	1.1

\* Trace amount of K-feldspar; + trace amount; – not detected.

randomly selected rural soils from 6 different provinces. Most rural and urban soils contained moderate amounts of hornblende and other iron-bearing minerals. Anatase was detected (from trace amount up to 1.7%) in some rural samples as well as in urban soils, denoted as 100 Ail subsurface, Salkhit surface, Zaisan and Officer Palace surface and subsurface (content in the surface was 1.2%).

The concentrations of 47 elements in Ulaanbaatar soils were tabulated in Table 3, in comparison to the soil average values quoted in Vinogradov [33] and Bowen [5]. The data pertaining to rural soils have been reported by Tserenpil et al. [31]. Since the contents of various elements in rural soils of Mongolia fell within the range of values for soils worldwide, they could serve as a reference for uncontaminated soils in the region. Concentrations of trace elements were in the similar range for urban and rural soils for elements including Co, Cr, Cs, Cu, Mn, Nb, Ta, Tl, U and V and were comparable with the soil average. The average Zn concentration for both

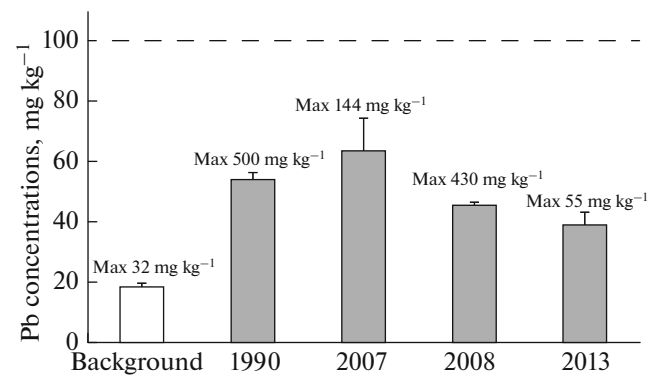
rural (average of 115 mg kg<sup>-1</sup>) and urban (average of 148 mg kg<sup>-1</sup>) soils was similar in magnitude but 2 and 3 times higher than the world average, respectively. As high as 259 mg kg<sup>-1</sup> value was also reported in another investigation in rural soils [19]; hence, this finding may be ascribed to regional natural abundance although anthropogenic pollution may also have contributed. Kasimov et al. [12], for example, reported a background value of 41 mg kg<sup>-1</sup> for soils from the Gatsurt and Songino areas, located just outside the centralized urban sites. However, average concentration of W was 6.8 mg kg<sup>-1</sup> and 3.5 times greater than the rural background, which may be associated with its local geological feature.

We might add that Ulaanbaatar soils accumulated some chalcophile elements, namely, As, Cd (at some sites), Hg, Pb, Sb and Zn; those tend to be enriched in fossil fuels. Although slightly elevated concentrations for As (9.4–21.7 mg kg<sup>-1</sup>), Cd (up to 2.1 mg kg<sup>-1</sup>), Pb (20.4–54.7 mg kg<sup>-1</sup>) and Sb (1.8–4.7 mg kg<sup>-1</sup>) were

observed, the Hg level was approximately 40 times higher than its abundance in the earth's upper crust. It should be investigated further if it was affected by a local gold mining, where Hg was used to recover Ag through amalgamation [27]. Between the sampling points concentrations of elements with environmental interest were differed by showing more accumulations of As, Cl, Hg and Pb at 100 Ail, Officer and Zaisan than at other sites. At Salkhit site concentrations were lower, which may be correlated with its windward location in the city. Concentration of Cl in urban soils was greater (an average 16 times) than its abundance in the soils, particularly at 100 Ail location the Cl accumulation was extremely high both in surface ( $6160 \text{ mg kg}^{-1}$ ) and subsurface sample ( $4160 \text{ mg kg}^{-1}$ ). Deicing agents may be a common Cl source due to the extremely continental climate conditions along with other Cl containing products.

A survey 2008 [13], on different functional zones in Ulaanbaatar indicated that the soil cover was polluted weakly, meantime the pollution index decreased in relation to the soil ecological-geochemical state in 1990. These observations were explained in terms of the reduction in the industrial activity, natural attenuation in soil over time ('self-purification'), and frequent wind gusts blowing away contaminants from bare and loose soil covers. On the other hand, Table 3 showed that the average concentrations of trace elements in Mongolian soils were often in a lower end of the concentration ranges for the elements, including,  $6.2\text{--}11.0 \text{ mg kg}^{-1}$  for Co,  $32.2\text{--}77.5 \text{ mg kg}^{-1}$  for Cr,  $3.2\text{--}5.4 \text{ mg kg}^{-1}$  for Cs,  $21.7\text{--}36.5 \text{ mg kg}^{-1}$  for Cu,  $539\text{--}646 \text{ mg kg}^{-1}$  for Mn,  $12.1\text{--}21.8 \text{ mg kg}^{-1}$  for Ni,  $0.64\text{--}1.17 \text{ mg kg}^{-1}$  for Ta,  $0.4\text{--}0.5 \text{ mg kg}^{-1}$  for Tl and  $1.9\text{--}3.6 \text{ mg kg}^{-1}$  for U. Similarly, Cheng [7] has concluded that Chinese soils have low background levels of metals, including Pb.

**Concentrations of lead and its stable isotopic composition.** By the 1990s, the vast majority of soils in the industrial and residential areas of Ulaanbaatar were already polluted by Pb even when the city's population was lower, and the number of motor vehicles was much smaller than it is at present [12]. The soil Pb concentration exceeded twice the average value, being 5–7 times higher for soils from central areas than soils from the outer regions. A maximum Pb concentration of  $430 \text{ mg kg}^{-1}$  was recorded with an average value of  $46 \text{ mg kg}^{-1}$  for 90 topsoil samples taken in 2008 from different zones in Ulaanbaatar [13]. Figure 2 showed the variations in Pb concentration for Ulaanbaatar soils as reported by different authors during sampling campaigns in years 1990, 2007, 2008 and 2013. Analysis of variance (ANOVA) between data sets revealed a statistical significance at a lower level ( $p$ -value 0.064) [29]. Moreover, continuous increase in the average Pb concentration was not observed with time, when data sets were compared. In the present investigation, we measured a maximum Pb concentration of  $54.7 \text{ mg kg}^{-1}$



**Fig. 2.** Variations in average Pb concentration of Ulaanbaatar soils since 1990 in relation to the background value for uncontaminated soils and the maximum permissible limit set for Mongolia. Gray bars refer to values measured in 1990 [12]; 2007 [1]; 2008 [13] and 2013 (present study). White bar denotes background concentration [31]; and dashed line represents permissible limit (PL) in the country (MNCSSM, MN 11047:2007). Values of maximum Pb concentrations from separate surveys were shown on top of each bar.

for the Zaisan topsoil while the values for rural soils varied from 16 to  $32 \text{ mg kg}^{-1}$ , comparable with the range of  $24\text{--}33 \text{ mg kg}^{-1}$  reported by Kasimov et al. [13].

Table 4 lists the Pb isotopic composition, expressed as ratios of  $^{208}\text{Pb}/^{204}\text{Pb}$ ,  $^{207}\text{Pb}/^{204}\text{Pb}$ ,  $^{206}\text{Pb}/^{204}\text{Pb}$ ,  $^{208}\text{Pb}/^{206}\text{Pb}$  and  $^{206}\text{Pb}/^{207}\text{Pb}$  for the soils, TSP and coals as well as for coal ash. Based on the multi element composition, the rural soils were considered to be unpolluted, hence their Pb contents may serve as a baseline for the country. By the same token, the Pb isotopic composition of rural soils would reflect the natural abundance of the isotopes in question.

The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio for rural soils ranged from 1.186 to 1.207, and the ratio for soils from Ulaanbaatar city ranged from 1.163 to 1.185. Similarly, the  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio for the randomly collected rural soils (18.470–18.840) was appreciably higher than the range of values observed for urban soils (18.128–18.316), especially those adjacent to busy arterial roads (Table 4). By contrast, the  $^{208}\text{Pb}/^{206}\text{Pb}$  ratio for rural soils was lower than that for urban soils. This observation was consistent with the radiogenic origin of Pb much of which was derived from weathered bedrocks. In other words, the isotopic ( $^{208}\text{Pb}/^{206}\text{Pb}$ ) ratio for uncontaminated soils was essentially influenced by the radioactive decay of  $^{238}\text{U}$  yielding  $^{206}\text{Pb}$  as the end product. Further evidence showed that the isotopic composition for the rural soils are indicative as a regional phenomenon. For instance, Gobi Desert in southern Mongolia exhibited the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio as 1.200–1.211 [3], and it was reported to be 1.183–1.203 for the China Loess Plateau to which the Mongolian Gobi contributes significantly [34]. Similar values of 1.187–1.191 (18.409–18.521 for the  $^{206}\text{Pb}/^{204}\text{Pb}$ ) were

**Table 3.** Concentrations of major and trace- elements in soils from Ulaanbaatar city ( $\text{mg}\cdot\text{kg}^{-1}$ ) (unless stated otherwise)

Elements §	100 AIL		4 ZAM		ZAISAN		OFFICER	SALKHIT	
	surface	subsurface	surface	subsurface	surface	subsurface	Surface	surface	subsurface
Na, %	2.72 ± 0.16	2.52 ± 0.15	2.64 ± 0.16	2.54 ± 0.15	1.75 ± 0.11	2.01 ± 0.12	2.01 ± 0.12	1.95 ± 0.12	2.02 ± 0.12
Mg, %	1.07 ± 0.11	1.06 ± 0.12	1.03 ± 0.10	1.19 ± 0.12	0.97 ± 0.96	1.07 ± 0.11	1.08 ± 0.11	1.12 ± 0.11	1.20 ± 0.12
Al, %	6.07 ± 0.30	5.75 ± 0.29	6.23 ± 0.31	7.46 ± 0.37	6.01 ± 0.30	6.48 ± 0.32	6.92 ± 0.35	6.61 ± 0.33	7.13 ± 0.36
K, %	2.72 ± 0.27	2.40 ± 0.24	2.99 ± 0.30	2.75 ± 0.28	2.28 ± 0.23	2.53 ± 0.25	2.09 ± 0.21	2.29 ± 0.23	2.32 ± 0.23
Fe, %	2.22 ± 0.11	2.04 ± 0.10	2.56 ± 0.13	2.61 ± 0.13	2.06 ± 0.10	3.19 ± 0.16	2.85 ± 0.14	3.06 ± 0.15	2.95 ± 0.15
Ca, %	4.99 ± 0.50	4.65 ± 0.47	1.67 ± 0.17	1.83 ± 0.18	1.72 ± 0.17	1.46 ± 0.15	5.66 ± 0.57	2.82 ± 0.28	3.00 ± 0.30
Cl	6160 ± 1540	4160 ± 1040	506 ± 127	200 ± 50	795 ± 199	724 ± 181	701 ± 175	868 ± 217	840 ± 210
Sc	6.8 ± 0.8	6.5 ± 0.8	9.0 ± 1.1	9.3 ± 1.1	6.1 ± 0.7	10.5 ± 1.3	9.6 ± 1.2	11.2 ± 1.3	10.7 ± 1.3
Ti	3200 ± 384	2400 ± 288	3530 ± 424	4220 ± 506	2990 ± 359	3660 ± 439	3760 ± 451	3370 ± 404	4060 ± 487
V	53.5 ± 5.4	48.2 ± 4.8	53.2 ± 5.3	67.6 ± 6.8	50.1 ± 5.0	66.5 ± 6.7	76.1 ± 7.6	74.6 ± 7.5	69.3 ± 6.9
Cr	76.7 ± 7.7	67.5 ± 6.8	45.6 ± 4.6	50.9 ± 5.1	32.3 ± 3.2	77.5 ± 7.8	51.2 ± 5.1	70.1 ± 7.0	63.6 ± 6.4
Mn	547 ± 55	539 ± 54	573 ± 57	629 ± 63	560 ± 56	646 ± 65	577 ± 58	600 ± 60	633 ± 63
Co	6.2 ± 0.3	6.4 ± 0.3	8.5 ± 0.4	8.7 ± 0.4	6.9 ± 0.3	11.0 ± 0.6	9.7 ± 0.5	10.0 ± 0.5	9.5 ± 0.5
Ni	12.1 ± 1.8	15.7 ± 2.4	21.8 ± 3.3	20.6 ± 3.1	17.2 ± 2.6	13.9 ± 2.1	15.3 ± 2.3	20.8 ± 3.1	19.6 ± 2.9
Cu*	33.5 ± 0.6	30.5 ± 2.6	29.7	23.9 ± 1.5	36.5 ± 0.1	34.5 ± 1.7	22.9 ± 0.4	23.4 ± 1.0	21.7 ± 1.4
Zn	189 ± 9.45	178 ± 8.9	168 ± 8.4	134 ± 6.7	180 ± 9	180 ± 9	92 ± 4.59	107 ± 5.35	104 ± 5.2
Ga*	9.9 ± 0.9	10.1 ± 0.7	13.1	14.1 ± 1.1	11.7 ± 0.5	13.8 ± 1.2	11.5 ± 0.0	14.9 ± 0.5	14.1 ± 0.2
Ge*	1.5 ± 0.1	1.5 ± 0.1	1.6 ± 0.0	1.7 ± 0.1	1.8 ± 0.0	2.2 ± 0.0	1.7 ± 0.0	1.5 ± 0.0	1.4 ± 0.0
As	11.1 ± 0.6	11.4 ± 0.6	10.0 ± 0.5	13.5 ± 0.7	9.4 ± 0.5	15 ± 0.8	21.7 ± 1.1	11 ± 0.6	11.7 ± 0.6
Rb	82 ± 16	79 ± 16	99 ± 20	95 ± 19	68 ± 14	97 ± 19	85 ± 17	91 ± 18	90 ± 18
Sr	339 ± 34	334 ± 33	306 ± 31	302 ± 30	242 ± 24	311 ± 31	489 ± 49	329 ± 33	332 ± 33
Zr	419 ± 126	328 ± 98	486 ± 146	527 ± 158	274 ± 82	383 ± 115	464 ± 139	357 ± 107	379 ± 114
Nb*	9.1 ± 0.1	9.2 ± 0.4	11.8 ± 0.0	12.9 ± 0.6	9.8 ± 0.3	11.2 ± 0.1	10.5 ± 0.5	11.8 ± 0.2	11.6 ± 0.8
Mo*	1.4 ± 0.0	1.6 ± 0.0	1.2 ± 0.0	1.1 ± 0.0	2.2 ± 0.0	2 ± 0.1	1.2 ± 0.0	0.8 ± 0.1	1.1 ± 0.3

Table 3. (Contd.)

Elements §	100 AIL		4 ZAM		ZAISAN		OFFICER		SALKHIT	
	surface	subsurface	surface	subsurface	surface	subsurface	Surface	subsurface	surface	subsurface
Cd	0.2 ± 0.06	0.1 ± 0.03	0.3 ± 0.08	0.0	0.2 ± 0.07	2.1 ± 0.64	2.1 ± 0.63	1.9 ± 0.58	1.9 ± 0.58	1.9 ± 0.58
Sn*	5.4 ± 0.6	4.2 ± 0.7	4.3 ± 0.0	3.4 ± 1.0	4.0 ± 0.1	4.0 ± 0.5	3.3 ± 0.8	3.3 ± 0.6	3.3 ± 0.6	2.7 ± 0.1
Sb	4.65 ± 0.47	2.91 ± 0.29	2.48 ± 0.25	2.71 ± 0.27	2.66 ± 0.27	3.01 ± 0.30	2.72 ± 0.27	1.75 ± 0.18	1.75 ± 0.18	1.8 ± 0.18
Cs	3.49 ± 0.18	3.35 ± 0.17	4.29 ± 0.22	4.55 ± 0.23	3.22 ± 0.16	5.26 ± 0.26	5.41 ± 0.27	4.96 ± 0.25	4.96 ± 0.25	4.74 ± 0.24
Ba	626 ± 63	609 ± 61	695 ± 69	659 ± 66	557 ± 56	696 ± 70	729 ± 73	641 ± 64	641 ± 64	623 ± 62
La	25 ± 1.3	24.6 ± 1.2	30.8 ± 1.5	38 ± 1.9	21.5 ± 1.1	34.3 ± 1.7	30.7 ± 1.5	33.3 ± 1.7	33.3 ± 1.7	32.2 ± 1.6
Ce	55 ± 8.3	55 ± 8.2	66 ± 9.9	75 ± 11.2	49 ± 7.4	70 ± 10.5	60 ± 9	66 ± 9.9	66 ± 9.9	64 ± 9.6
Nd	48.4 ± 14.5	50.8 ± 15.2	59.4 ± 17.8	69.7 ± 20.9	17.9 ± 5.4	35.5 ± 10.7	26 ± 7.8	33 ± 9.9	33 ± 9.9	32.4 ± 9.7
Sm	4.01 ± 0.48	3.59 ± 0.43	2.58 ± 0.31	2.89 ± 0.35	3.02 ± 0.36	3.64 ± 0.44	3.5 ± 0.42	2.04 ± 0.25	2.04 ± 0.25	2.01 ± 0.24
Eu	0.07 ± 0.02	0.21 ± 0.06	0.20 ± 0.06	0.24 ± 0.07	0.19 ± 0.06	0.62 ± 0.19	0.52 ± 0.16	0.64 ± 0.19	0.64 ± 0.19	0.59 ± 0.18
Gd	7.4 ± 1.1	8.7 ± 1.3	7.7 ± 1.2	10.1 ± 1.5	0.8 ± 0.1	6.7 ± 1.0	5.8 ± 0.9	6.7 ± 1.0	6.7 ± 1.0	8.0 ± 1.1
Tb	0.58 ± 0.09	0.61 ± 0.09	0.70 ± 0.11	0.85 ± 0.13	0.56 ± 0.08	0.74 ± 0.11	0.70 ± 0.11	0.70 ± 0.11	0.70 ± 0.11	0.72 ± 0.11
Dy	0.7 ± 0.2	1.4 ± 0.4	0.7 ± 0.2	1.8 ± 0.5	0.9 ± 0.3	0.7 ± 0.2	0.7 ± 0.2	0.9 ± 0.3	0.9 ± 0.3	0.4 ± 0.1
Yb	0.01 ± 0.00	0.26 ± 0.04	0.94 ± 0.14	2.13 ± 0.32	2.05 ± 0.31	2.40 ± 0.36	2.87 ± 0.43	0.38 ± 0.06	0.38 ± 0.06	3.08 ± 0.46
Hf	8.82 ± 2.65	7.48 ± 2.24	10.5 ± 3.15	11.6 ± 3.48	5.93 ± 1.78	8.59 ± 2.58	9.46 ± 2.84	7.21 ± 2.16	7.21 ± 2.16	7.26 ± 2.18
Ta	0.87 ± 0.26	0.72 ± 0.22	1.02 ± 0.31	1.00 ± 0.30	0.64 ± 0.19	0.98 ± 0.30	1.17 ± 0.36	1.16 ± 0.35	1.16 ± 0.35	1.15 ± 0.35
W	5.89 ± 1.77	5.98 ± 1.79	6.07 ± 1.82	8.55 ± 2.57	9.01 ± 2.70	14.2 ± 4.26	6.63 ± 1.99	2.24 ± 0.67	2.24 ± 0.67	2.15 ± 0.65
Au	0.021 ± 0.006	0.016 ± 0.005	0.006 ± 0.002	0.007 ± 0.002	0.015 ± 0.005	0.022 ± 0.007	0.009 ± 0.003	0.004 ± 0.001	0.004 ± 0.001	0.004 ± 0.001
Hg	2.93 ± 0.73	3.53 ± 0.88	3.87 ± 0.97	3.68 ± 0.92	2.71 ± 0.68	5.47 ± 1.37	4.73 ± 1.18	1.4 ± 0.35	1.4 ± 0.35	0.336 ± 0.08
Tl*	0.4 ± 0.0	0.4 ± 0.0	0.5 ± 0.0	0.5 ± 0.0	0.4 ± 0.0	0.5 ± 0.0	0.4 ± 0.0	0.5 ± 0.0	0.5 ± 0.0	0.4 ± 0.0
Pb*	45.87 ± 0.55	46.71 ± 9.10	33.18	31.11 ± 0.38	54.71 ± 0.03	45.66 ± 1.07	26.99 ± 1.27	20.35 ± 0.45	20.35 ± 0.45	20.38 ± 0.25
Th	8.45 ± 0.42	7.98 ± 0.40	9.87 ± 0.49	12.2 ± 0.61	3.38 ± 0.17	10.3 ± 0.52	9.74 ± 0.49	9.74 ± 0.49	9.74 ± 0.49	10.8 ± 0.54
U	2.98 ± 0.30	2.99 ± 0.30	2.79 ± 0.28	3.61 ± 0.36	2.45 ± 0.25	3.06 ± 3.06	2.51 ± 0.25	1.87 ± 0.19	1.87 ± 0.19	1.97 ± 0.20

\* Concentrations of those elements were measured by ICP-MS after acid digestion. § Soil average given by Bowen (1979), and by (Vinogradov, 1959) if not noted.

**Table 4.** Lead isotopic ratios and total Pb contents for rural and urban soils in Mongolia and source related materials

Sample assignation	Pb content, mg kg <sup>-1</sup>	<sup>208</sup> Pb/ <sup>204</sup> Pb <sup>a</sup>	<sup>207</sup> Pb/ <sup>204</sup> Pb <sup>b</sup>	<sup>206</sup> Pb/ <sup>204</sup> Pb <sup>b</sup>	<sup>208</sup> Pb/ <sup>206</sup> Pb <sup>c</sup>	<sup>206</sup> Pb/ <sup>207</sup> Pb <sup>c</sup>
<b>Rural soils</b>						
1. Bayan Olgy	19.15	38.465	15.595	18.585	2.070	1.192
2. Gobisumber 3	26.30	38.486	15.592	18.630	2.066	1.195
3. Gobisumber 28	23.70	38.476	15.592	18.626	2.066	1.195
4. Gobisumber 41	24.70	38.440	15.593	18.593	2.067	1.192
5. Gobisumber 52	25.35	38.327	15.598	18.597	2.061	1.192
6. Sukhbaatar 1	23.00	38.492	15.596	18.635	2.066	1.195
7. Sukhbaatar 2	22.10	38.525	15.593	18.641	2.067	1.195
8. Dornod 1	15.50	38.365	15.580	18.470	2.077	1.186
9. Dornogobi 1	17.13	38.323	15.573	18.493	2.072	1.187
10. Dundgobi 1	19.35	38.383	15.572	18.548	2.069	1.191
11. Dundgobi 2	32.03	38.114	15.608	18.840	2.023	1.207
12. Dundgobi 3	18.55	38.334	15.581	18.585	2.063	1.193
13. Dundgobi 4	19.00	38.339	15.576	18.608	2.060	1.195
<b>Median</b>	<b>18.55</b>	<b>38.390</b>	<b>15.588</b>	<b>18.604</b>	<b>2.064</b>	<b>1.194</b>
<b>Urban soils</b>						
14. 100 Ail surface	45.87	38.206	15.597	18.247	2.094	1.170
15. 100 Ail subsurface	46.71	38.182	15.593	18.266	2.090	1.171
16. 4 Zam surface	33.18	38.166	15.576	18.218	2.095	1.170
17. 4 Zam subsurface	31.11	38.194	15.574	18.257	2.092	1.172
18. Zaisan surface	54.71	38.108	15.585	18.128	2.102	1.163
19. Zaisan subsurface	45.66	38.161	15.581	18.175	2.100	1.167
20. Officer surface	26.99	38.242	15.576	18.276	2.093	1.173
21. Officer subsurface	28.60	38.252	15.567	18.316	2.089	1.177
22. Salkhit surface	20.35	38.397	15.573	18.448	2.081	1.185
23. Salkhit subsurface	20.38	38.350	15.571	18.407	2.083	1.182
<b>Median</b>	<b>39.10</b>	<b>38.226</b>	<b>15.579</b>	<b>18.274</b>	<b>2.092</b>	<b>1.173</b>
<b>Related materials</b>						
24. Ash PP4‡		38.876	15.646	19.614	1.982	1.254
25. Ash PP3‡		39.202	15.654	19.716	1.988	1.260
26. Coals (average)		39.240	15.630	19.324	2.031	1.236
27. TSP1§		38.206	15.596	18.261	2.092	1.171
28. TSP2§		37.615	15.424	18.064	2.082	1.171

<sup>a</sup> SD for given values are  $1 \times 10^{-3}$ ; <sup>b</sup>  $1 \times 10^{-4}$  and <sup>c</sup>  $1 \times 10^{-5}$  orders of magnitude; † median value excluding data at Salkhit site; ‡ PP-Power Plant; § TSP1 – Total Suspended Particulates from Ulaanbaatar atmosphere between November and December, 2013; § TSP2 – Total Suspended Particulates from Ulaanbaatar atmosphere between December, 2013 and January, 2014.

found also in volcanic materials sampled from a volcanic field in a border area between China and Mongolia [11].

It was noteworthy that the surface and subsurface soils from urban districts had similar Pb isotopic ratios along with similar Pb concentration range. For example, the <sup>206</sup>Pb/<sup>207</sup>Pb ratio of the soils from the Salkhit site was 1.185 for the surface samples and 1.182 for the

subsurface materials; these values were slightly higher than those measured at other sites in the city. The lowest <sup>206</sup>Pb/<sup>207</sup>Pb ratio was obtained for the Zaisan, leeward location by prevailing wind direction (Fig. 1), being 1.163 for the surface and 1.167 for the subsurface sample. It would appear that Pb isotopic composition for both natural and human-altered environments are not sensitive to weathering and processing although

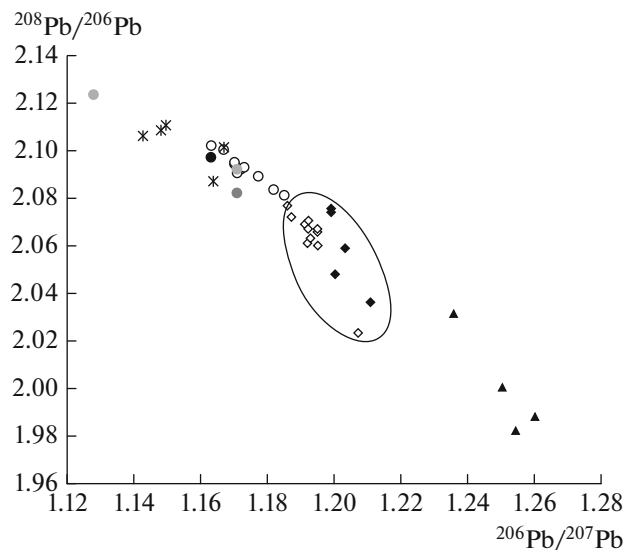


their values can change through receiving inputs from secondary Pb sources. Thus, Pb isotopic ratios can be used to pinpoint the source of pollution.

Depressed values of  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio for Ulaanbaatar soils reflect the “mixing” of Pb pollution sources, notably coal combustion and also car emission. Several approaches and models are available to differentiate Pb sources. Figure 3 shows a three-isotope plot for the Mongolian soils investigated and other related materials. Two areas, corresponding to urban and rural soils, were distinguished by analysis of variance ( $p = 0$ ). The Pb isotope composition for the urban Salkhit site was close to that for rural (uncontaminated) soils, its value being at the border between the urban and rural sites. This finding was consistent with this site being located away from busy roads and upwind direction from the city center.

Mongolia uses coal as the main fuel for generating heat and electricity, with ca. 5 million tons being consumed annually in Ulaanbaatar city [2]. Little information, however, is available on the Pb isotopic composition of source related materials. As such, at this time it was not possible to interpret temporal changes in Pb pollution in terms of specific sources. The high  $^{208}\text{Pb}/^{204}\text{Pb}$  (38.876–39.202) and  $^{207}\text{Pb}/^{204}\text{Pb}$  (15.646–15.654) ratios for fly ash determined in this investigation may be related to the geological age of the parent coal deposits. The average  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of 1.236 measured for Mongolian coals (Table 4) would also indicate that Mongolian coals was more radiogenic than either coals ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.206$ ) used in Irkutsk, Russia [22]), or the coals ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.17$ –1.18) in China [6, 23]. Although the radiogenic emission of Pb from coal combustion does not constitute the sole input of Pb into the urban soils in Ulaanbaatar, it has the effect of increasing the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio with respect to natural fractionations. If coal combustion was a significant source to the soil Pb pollution, its contribution to the Pb content in the urban samples would be rather pronounced in mid-winter, when large quantities of coal are used for heating. Nonetheless, aerosol samples taken between November, 2013 and January, 2014 disclosed the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of 1.171 (a sampling location NUM as depicted in Fig. 1), which resembled to the urban soils. However, dispersions of pollutants could be different day-to-day depending on weather conditions and sampling locations. In this point aerosol samples need to be monitored with high frequency and in different locations around Ulaanbaatar.

Natural non-ore materials, such as soil and coal, generally tend to be more radiogenic in terms of their Pb isotopic composition as compared to aged mineral Pb deposits. As such, the input into urban soils of Pb from leaded gasoline would diminish the radiogenic character of the samples [23]. Thus, leaded gasoline (as a source of Pb pollution) was a factor affecting the Pb isotopic ratio of urban soils in Mongolia. Unfortu-



**Fig. 3.** Three-isotope plot for rural and urban soils in Mongolia. Open circles denoted isotopic ratios in urban soils; whereas filled dark circles for atmospheric aerosols in 1994 and 1995 [4] and grey circles correspond to winter aerosols in 2013–2014 (this investigation). Stars are referring to the atmospheric depositions from Russian Far east and Baikal region. Open diamonds refer to rural soils, and filled diamonds to samples from Chinese Loess [34], and to Mongolian Gobi [3]; Filled triangles signify Mongolian coal and Power Plant fly material.

nately, information is lacking on Pb contents and its isotopic composition in the fuels imported into Mongolia. To the first approximation, however, we may use published data on the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio for leaded gasoline from Russia and China, the principal suppliers of fuel to the Mongolian market. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of 1.134–1.149 was reported for Russian fuel [22] and  $1.157 \pm 0.006$  for vehicle exhaust [16], and were close to the range of values measured for ores from Kazakhstan. In larger extent of its territory, the Russian Far East and the Baikal region showed the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio from 1.140 to 1.167 in its atmospheric depositions between 2005 and 2007 [25]. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of Chinese ores varied from 1.081 to 1.176, with lower values being recorded for ores from Northern China by Mukai et al. [22]. Relatively low values (1.098–1.116) in Chinese cities have also been measured for vehicle exhaust due to the use of imported alkyl-lead additives with a characteristically low  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio [6]. It would therefore appear that the low  $^{206}\text{Pb}/^{207}\text{Pb}$  signatures of leaded gasoline from Russia and China lie behind the depression in the Pb isotopic ratio for Ulaanbaatar soils, notably those sampled along the traffic roads.

The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio was examined in aerosol specimens those collected in June 1994 and August 1995 in Ulaanbaatar and was found 1.128 and 1.163, respectively [4]. As it is indicated aerosols were sam-

pled during the non-heating months in Ulaanbaatar; therefore, changes in the Pb isotopic ratio dominantly influenced by the automobile exhaust and a difference in values from 1994 and 1995 years indicating a change in sources. It may be timely coincident with opening of extensive trade of the country with the southern neighbor i.e., China. The aerosol value found in 1995 was same as values for the soils adjacent to the main traffic roadsides and aerosol samples in this investigation. The Pb pollution source seems to remain nearly constant and locally generated. Moreover, it raises skepticism about whether or not Mongolia completely phased out the use of the leaded fuels [32].

## CONCLUSIONS

Soils within the Ulaanbaatar city were differed with high total organic matter content; and mineralogical composition also varied compared to the Mongolian rural soils. Total of 47 elements were determined for their distributions by using nuclear analytical and plasma based techniques, latter one requires acid digestions. Insignificant enrichment of elements such as As, Pb, Sb, W and Zn was observed, but not all of them are regarded as anthropogenic pollutants in this context. However, very high accumulation of Hg and Cl obviously is associated with character of the land use.

Up to several decades ago, the soils in Ulaanbaatar were polluted moderately by some heavy and toxic metals (e.g., Pb), but this situation is not worsened over time. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of 1.163–1.185 for urban soils was appreciably lower than the value of 1.186–1.207 measured for rural soils. The latter range of values was indicative of natural fractionation between stable isotopes and corresponding to a regional natural fingerprint. By comparison, the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of 1.236 was recorded for the coals and 1.257 for fly ash materials collected from the two main coal-fired power plants in the city. The contribution of radiogenic Pb emissions from coal combustion did not appear to be dominant. The relatively low  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios for urban soils would indicate an input of Pb (from lead ore) used as an additive in gasoline. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios for roadside soils in Ulaanbaatar soils, however, were higher than those measured for Russian and Chinese leaded gasoline and vehicle exhausts. Thus, vehicle exhaust and coal combustion appear to have a competing influence on the Pb isotopic composition of urban soils. As a result, the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio for Ulaanbaatar soils deviates from that of leaded gasoline or coal, and would indicate only moderate pollution by Pb. Literature data on Ulaanbaatar aerosol from 1995 were comparable to the urban soil  $^{206}\text{Pb}/^{207}\text{Pb}$  composition with value of 1.163. The ratio resembled in recent urban aerosol samples and was 1.171. Therefore, it may be assumed that the Pb pollution source in Ulaanbaatar was nearly constant. However, to verify this observations more

experiments will be needed. The present investigation provided valuable baseline data of Mongolian Pb pollution and isotopic signatures under the condition of continuous atmospheric transport.

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## REFERENCES

1. T. Batjargal, E. Otgonjargal, K. Baek, and J.-S. Yang, "Assessment of metals contamination of soils in Ulaanbaatar, Mongolia," *J Hazard. Mater.* **184**, 872–876 (2010).
2. L. Batnum and S. Enhmaa, *Ulaanbaatar Hotiin Agaar Bohirduulah eh Uusveriin Uzleg, Toollogo Yavuulsan Tuhai Ajliih Tailan* (Ulaanbaatar, 2008), pp. 40–41.
3. P. E. Biscaye, F. E. Grousset, M. Revel, S. van der Gaast, G. A. Zielinski, A. Vaars, and G. Kukla, "Asian provenance of glacial dust (stage 2) in the Greenland Ice Sheet Project 2 Ice Core, Summit, Greenland," *J. Geophysical Res.* **102**, 26 765–26 780 (1997).
4. A. Bollhöfer and K. J. R. Rosman, "Isotopic source signatures for atmospheric lead: the Northern Hemisphere," *Geochim. Cosmochim. Acta* **65**, 1727–1740 (2001).
5. H. J. M. Bowen, *The Environmental Chemistry of the Elements* (Academic, London, 1979).
6. J. Chen, M. Tan, Y. Li, Y. Zhang, W. Lu, Y. Tong, G. Zhang, and Y. Li, "A lead isotopic record of Shanghai atmospheric lead emissions in total suspended particles during the period of phasing out of leaded gasoline," *Atmos. Environ.* **39**, 1245–1253 (2005).
7. S. Cheng, "Heavy metal pollution in China: origin, pattern, and control," *Environ. Sci. Pollut. Res.* **10**, 192–198 (2003).
8. A. D. Clarke, Y. Shinozuka, V. N. Kapustin, S. Howell, B. Huebert, S. Doherty, T. Anderson, D. Covert, J. Anderson, X. Hua, K. G. Moore, C. McNaughton, G. Carmichael, and R. Weber, "Size distributions and mixtures of dust and black carbon aerosol in Asian outflow: physiochemistry and optical properties," *J. Geophys. Res.: Atmos.* **109**, 15S09 (2004). doi 10.1029/2003JD004378
9. F. J. Doucet and J. Carignan, "Atmospheric isotopic composition and trace metal concentration as revealed by epiphytic lichens," *Atmos. Environ.* **35**, 3681–3690 (2001).
10. I. P. Gerasimov and N. A. Nogina, *The Soil Cover and Soil of Mongolia* (Nauka, Moscow, 1984), pp. 109–112.
11. K.-S. Ho, Y. Liu, J.-C. Chen, and H.-J. Yang, "Elemental and Sr-Nd-Pb isotopic compositions of late Cenozoic Abaga basalts, Inner Mongolia: implications

- for petrogenesis and mantle process,” *Geochem. J.* **42**, 339–357 (2008).
12. N. S. Kasimov, M. Yu. Lychagin, A. K. Evdokimova, D. L. Golovanov, and Yu. I. Pikovskii, “Ulaanbaatar, Mongolia (heat and power engineering): intermountain depression,” in *Ecogeochemistry of Urban Landscapes*, Ed. by N. S. Kasimov (Moscow State University, Moscow, 1995), pp. 231–248.
  13. N. S. Kasimov, N. E. Kosheleva, O. I. Sorokina, S. N. Bazha, P. D. Gunin, and S. Enkh-Amgalan, “Ecological-geochemical state of soils in Ulaanbaatar (Mongolia),” *Eurasian Soil Sci.* **44** (7), 709–722 (2011). doi 10.1134/S106422931107009X
  14. M. Komárek, V. Ettler, V. Chrastny, and M. Mihaljevič, “Lead isotopes in environmental sciences: a review,” *Environ. Int.* **34**, 562–577 (2008).
  15. V. I. Kovalenko, *Rare Metal Metallogeny of People’s Republic of Mongolia Based on Potential Ore Resources on Magma Minerals. Ore Deposits of Magma Associations* (Nauka, Moscow, 1988), pp. 114–159.
  16. D. V. Ladonin and O. V. Plyaskina, “Isotopic composition of lead in soils and street dust in the southeastern administrative district of Moscow,” *Eurasian Soil Sci.* **42** (1), 93–104 (2009). doi 10.1134/S1064229309010128
  17. F.-L. Li, C.-Q. Liu, Y.-G. Yang, X.-Y. Bi, T.-Z. Liu, and Z.-Q. Zhao, “Natural and anthropogenic lead in soils and vegetables around Guiyang city, southwest China: a Pb isotopic approach,” *Sci. Total Environ.* **431**, 339–347 (2012).
  18. L. Long, “Lead isotopes,” in *Encyclopedia of Geochemistry*, Ed. by C. P. Marshall and R. W. Fairbridge (Kluwer, Amsterdam, 1999).
  19. A. Markwitz, B. Barry, and D. Shagijamba, “PIXE analysis of sand and soil from Ulaanbaatar and Karakorum, Mongolia,” *Nucl. Instrum. Methods Phys. Res.* **266**, 4010–4019 (2008).
  20. Y. Meng, G. Gong, Z. Wu, Z. Yin, Y. Xie, and S. Liu, “Fabrication and microstructure investigation of ultra-high-strength porcelain insulator,” *J. Eur. Ceram. Soc.* **32**, 3043–3049 (2012).
  21. *MNCSSM, MN 11047:2007: Soil Pollutants Standard of Mongolia* (Mongolian National Center of Standardization and Metrology, Ulaanbaatar, Mongolia, 2007).
  22. H. Mukai, A. Tanaka, T. Fujii, Y. Zeng, and Y. Hong, “Regional characteristic of sulfur and lead at several Chinese urban sites,” *Environ. Sci. Technol.* **35**, 1064–1071 (2001).
  23. H. Mukai, T. Machida, A. Tanaka, Y. P. Vera, and M. Uematsu, “Lead isotope ratios in the urban air of eastern and central Russia,” *Atmos. Environ.* **35**, 2783–2793 (2001).
  24. V. V. Nikonov, N. V. Lukina, and M. V. Frontasyeva, “Trace elements in Al-Fe humus podzolic soils subjected to aerial pollution from the apatite-nepheline production industry,” *Eurasian Soil Sci.* **32** (12), 1331–1339 (1999).
  25. N. A. Onischuk, T. V. Khodzher, and E. P. Chebykin, “Chipanina lead and its isotopic ratios in atmospheric precipitation in the Baikal region and Primorski krai,” *Geogr. Nat. Resour.* **30**, 345–349 (2009).
  26. T. V. Pampura, A. Probst, D. V. Ladonin, and V. A. Demkin, “Lead content and isotopic composition in submount and recent soils of the Volga Upland,” *Eurasian Soil Sci.* **46** (11), 1059–1075 (2013). doi 10.1134/S1064229313090020
  27. M. Satoshi, N. Kazuki, D. Gunchin, and H. S. Soey, “Mercury content in electrum from artisanal mining site of Mongolia,” *Nucl. Instrum. Methods Phys. Res., Sect. B* **249**, 556–560 (2006).
  28. B. R. T. Simoneit, M. Kobayashi, M. Mochida, K. Kawamura, M. Lee, H. J. Lim, B. J. Turpin, and Y. Komazaki, “Composition and major sources of organic compounds of aerosol particulate matter sampled during the ACE-Asia campaign,” *J. Geophys. Res.: Atmos.* **109**, 19S10 (2004). doi 10.1029/2004JD004598
  29. D. S. Soper, *Analysis of Variance (ANOVA) Calculator*, 2015. <http://www.danielsoper.com/statcalc>
  30. O. I. Sorokina and S. Enkh-Amgalan, “Lead in the landscapes of Ulaanbaatar city (Mongolia),” *Arid Ecosyst.* **2** (1), 61–67 (2012).
  31. Sh. Tserenpil, O. D. Maslov, N. Norov, C.-Q. Liu, M. F. Phillipov, B. K. G. Theng, and A. G. Belov, “Chemical and mineralogical composition of the Mongolian rural soils and their uranium sorption behavior,” *J. Environ. Radioact.* **118**, 105–112 (2013).
  32. UNEP, *Target 2008: Global Elimination of Leaded Petrol. A Report of the Partnership for Clean Fuels and Vehicles (PCFV) United Nations Environment Program* (Nairobi, Kenya, 2008).
  33. A. P. Vinogradov, *The Geochemistry of Rare and Dispersed Chemical Elements in Soil* (Consultants Bureau, New York, 1959).
  34. F. Wu, H. S. S. Hang, Q. Sun, and I. S. H. Sai, “Provenance of Chinese loess: evidence from stable lead isotope,” *Terr., Atmos. Ocean. Sci.* **22**, 305–314 (2011).