Review Article

Evaluation of the Pollution by Toxic Elements around the Small-Scale Mining Area, Boroo, Mongolia

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Received 27 January 2012; Accepted 15 March 2012

Academic Editors: B. J. Birch, A. Lewenstam, S. Polesello, and A. Szemik-Hojniak

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The objective of this study is to investigate the contamination levels of toxic elements (TEs) in the vicinity of the small-scale mining Boroo area, Mongolia. Samples of surface soil, ground water and human hair were collected around the gold washing or milling places, grassland and village areas. After appropriate preparation, all samples were analyzed for major and toxic elements (TEs) by Particle-Induced X-ray Emission Spectrometry (PIXE). Soil texture, conductivity (EC), pH, total organic carbon (TC) and nitrogen (TN) contents were also measured. The enrichment factor (EF) was estimated to assess the level of the contamination and the possible anthropogenic impact in soils from the mining activity. The EF_{soil} for Cu, As and Pb were in the highest values around gold washing place, indicating that around mining area surface soils were highly enriched by those elements. The Mn, Fe and Ni concentrations of drinking waters exceed the WHO values. The mean concentrations of Ca, Ti, As and Sr were higher in hair of Mongolian miners than in the hair of normal people in Japan, Mongolia and Philippines. These results indicate that the area around gold washing or milling could be the main contamination sources of As and other toxic elements (TEs) in the surface soil samples.

1. Introduction

Small-scale mining (artisanal) with metallic mercury is practiced in many developing countries of the world. Small-scale mining is one of the most important sources of toxic elements (TEs) in the environment. Mining generally releases toxic elements such as arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg). The adverse effects of mining activity on the environment as well as human health have been observed in many countries [1–4]. Environmental pollution with TE originated by mines can become an important source of contamination both in soil and water [5–8]. Also the TEs contamination of soils and surface water or groundwater represents a great threat to human health

due to its high potential to enter into the food chain. Human scalp hair has been used as an alternative biological material for blood and urine in biomonitoring environmental and occupational exposures of various pollutants (papers), since its sampling is considered less invasive, more convenient to store and transport, and less hazardous to handle.

In Mongolia, small-scale mining started in 1998 with few small-scale miners, but the number has increased rapidly. The miners in these areas are almost rural people, who had lost their livestock and harvest during the winter and summer by a natural catastrophe, *zud* and drought, which happened in recent years. In the Boroo area of Mongolia, which is currently one of the active site for small-scale gold mining, test analyses on soils, river water, and human hair

FIGURE 1: Location of sampling sites in the Boroo area, Mongolia.

of toxic contaminants were primarily centered at mercury determination since 1994 (JICA project report). Many analytical methods have been developed for elemental analysis which can be used for environmental pollution analysis in selected areas. Among such methods, the particle-induced X-ray emission spectrometry (PIXE) has been widely used for elemental analysis along with neutron and proton activation analyses (NAA, PAA). Our previous studies had concerned with the development of a methodology for total elemental analyses [9–11] in environmental samples by PIXE method. The elemental analysis along Boroo River showed that [9] the contents of TE in the upstream river water were lower than those in the downstream river water, while the pH was also lower in the downstream river water compared to the upstream river water. It was indicating that the downstream waters might be influenced by gold mining activity in this area. But TEs concentrations in soil, ground water, and other tissues remained poorly documented and required further investigation for environmental assessment of Boroo small-scale mining area. Comparison of TEs

in water, soils, sediments, and human hairs (popular for monitoring environmental exposure) would give valuable information on the nature of TE intake (migration in soils, food chain, etc.) by human beings as well as their effect on health condition of selected cohort.

The objective of this study is to determine the heavy metal levels on soil, drinking water, and human hair at the area of the small-scale mining of Boroo area, Mongolia, to disseminate the acquired information to the local people and line ministries. We hope that this paper could be a valuable reference for counter actions to toxic elements pollution.

2. Materials and Method

2.1. Study Area. The Bornuur village of the Boroo area, where the study took place, is located in the southwestern part of the North Khentei gold belt at the distance about 120 km from the capital city Ulaanbaatar of Mongolia (north latitude 48.4°C, east longitude 106.1°C). In this area the illegal small-scale mining is conducted. Boroo River that

crosses the gold mining area is an important water supply for agricultural activity. From a geological point of view, in the studied area, the so-called Chara Group of early Paleozoic age (sedimentary rocks) and Boroo gold complex (igneous rocks) of Early Paleozoic age are outcropped. In the area hydrothermal and metasomatic alterations are widely developed, and the mineralized portion includes quartz-gold veins and gold dissemination zone. Constituent minerals of vein are mainly quartz and minor amount of electrum, argentite, pyrite, arsenopyrite, sphalerite, chalcocite, tetrahedrite, galena, tellurobismuthite, albite, and calcite. In our approach, around the gold mining area, when minerals were crushed, some TEs transferred by the wind into the soil under water of the studied area, originating from chiseling, wells, and explosions. The annual average temperature at the site is 17°C, and the precipitation does not exceed 300 mm with rainfall which usually occurs in the period between the end of summer and autumn.

2.2. Sampling. The sampling location points are sketched in the map of Figure 1. To provide a satisfactory geographical and human activity representation of the site, the sampling area for surface soils was subdivided into three areas: the grassland (G) representing the control area where 8 samples were taken, the village (V) representing human activities where 9 samples were taken, and the gold washing place of minerals (M) representing the influence of mining activity where 7 samples were taken. Additionally, three ground water samples were taken from the wells (GW1, GW2, and GW3).

Twenty-four soils were taken from a depth of 0.2–10 cm under the surface, in order to avoid contamination or removal of TEs from the surface by the wind. The soil samples were air-dried and sieved by 2 mm sieve prior to chemical analysis. The drinking water samples were taken from acting wells used by the local population (3 samples with duplicates in total, each of 100 mL, were taken). The water samples were taken in acid-washed plastic bottles and acidified (pH < 2) with HNO₃. All the samples were kept refrigerated at 4°C prior to the analysis. Human hair samples were asked from 25 miners and 20 local residents not involved in the mining activity. The samples thoroughly washed to remove external contamination by 1% soap solution, acetone, and distilled water prior chemical analysis.

2.3. Chemical Analysis. The chemical properties of soil samples were investigated by using the standard methods of analytical chemistry. The pH of the soil was measured by the use of a Toko, TPX-90i pH meter, made in Japan. In order to measure the pH of the soil, the soil was added in a 1 M-KCl solution, in ratio of 1 : 2.5. Electrical conductivity (EC) was measured by a Horiba, EC-51 conductivity meter, made in Japan. The TC and TN contents were analyzed using a CN analyzer (Sumika, Sumigraph NC-80). Particle size distribution (wt%) was investigated by using the pipette method [12]. Elemental analysis of soil samples after acid digestion was carried out by a previously developed USA methodology for PIXE [11]. The pH of water samples was measured by electrode method using a pH meter (Toko,

TPX-90i, Japan). Trace and TE contents in drinking water were determined according to the methodology described by Bolormaa et al. [9].

The dissolution of human hair samples was carried out by a mixture of nitric acid (HNO₃) and hydrogen peroxide (H₂O₂). The ratio of the acid should be given. The methodology of elemental analysis for human hair including the accuracy and precision is reported in Bolormaa O. et al. [10]. The relationship between metal concentration in soil and human hair samples was tested by Pearson's correlation coefficients (*r*). Coefficients of correlation were calculated at the significance level $P \leq 0.05$.

All the chemicals were supplied by Wako Pure Chemical Co.Ltd. (Osaka, Japan), and used without future purification.

2.4. The PIXE Technique. The targets prepared following the procedures above were irradiated by 2.5 MeV proton beam from the single-end-type Van de Graaff accelerator of Tokyo Institute of Technology (Tokyo, Japan). The proton beam was collimated to give a beam size of about 5 mm in diameter at the collimator exit. Emitted X-rays were analyzed using an ORTEC (Tennessee, USA) Si (Li) detector (active volume 0.45 cm³, effective diameter 10 mm, thickness 5.67 mm, distance from window 7 mm, FWTES of FeK_α X-Ray 180 eV) with a 1 mm polyethylene absorber and CANBERRA (Meriden, USA) Si (Li) detector (effective diameter 4 mm, thickness 3 mm, distance from window 5 mm, FWTES of FeK_α X-Ray 160 eV) without an absorber. The beryllium window was 25 μm in thickness for both detectors. The first detector was used for measurement of heavy elements ($Z > 25$) and the second for light or major elements ($Z < 25$). For soil samples we used Si (Li) detector (active volume 0.45 cm³, effective diameter 10 mm, thickness 5.67 mm, distance from window 7 mm, FWTES of FeK_α X-Ray 180 eV) with a 2 mm polyethylene absorber to avoid the effect of pile up associated with the FeK_α line in view of the high iron concentration in soil samples. The analysis of the X-rays was performed using PIXE2 software (<http://www.Labo-eq.co.jp/>). Major elements ($Z < 25$) of the samples (soil, water, human hair) analyzed by drop-and-dry method. For this procedure, a 20 μL aliquot of the mixture from sample was dropped onto Nuclepore Track-Etch Membrane (Whatman, New Jersey, USA), air-dried, and irradiated by proton beam. For TEs ($Z > 25$) the pre-concentration method (water samples treated to form metal dibenzylthiocarbamate (DBDTC) complexes) was applied and molybdenum (Mo) was used as an internal standard. Since the elements of low-energy region in the elemental spectra are covered by sulfur (S), which forms complexes with DBDTC, major elements could not be determined in water samples by preconcentration method. The X-ray spectra analysis was performed by using PIXE 2 software package, which is produced by the Laboratory Equipment Co., Ltd. (Japan) (<http://www.Labo-eq.co.jp/>). Since the energy of PbL_α and AsK_α X-rays is almost identical, As and Pb could not be distinguished in the spectrum. Therefore, AsK_β was used to correct the contribution of AsK_α to PbL_α.

TABLE 1: Physicochemical characteristics of soil samples.

Sampling site	Characteristics							
	pH (water)	EC, mS/m	Ng kg ⁻¹	C, g kg ⁻¹	C/N	Cly [%]	Silt [%]	Sand [%]
Grassland (<i>n</i> = 8)								
Range	7.76–8.74	3.53–10.1	1.03–2.13	15–25.8	10.80–16.84	10.53–21.54	10.89–23.54	55.71–78.57
Mean	8.45	6.95	1.61	20.95	13.3	16.92	15.81	67.28
Village (<i>n</i> = 9)								
Range	7.72–9.09	2.02–17.59	0.993–3.36	16.37–35.45	9.70–17.32	15.27–27.57	13.43–38.18	41.85–70.94
Mean	8.46	7.16	1.74	22.04	13.69	19.43	20.76	59.81
Gold washing place (<i>n</i> = 7)								
Range	7.86–8.76	2.59–28.2	0.37–5.30	5.40–57.61	10.86–14.70	11.79–21.31	15.58–36.97	47.18–70.14
Mean	8.20	13.29	2.41	29.13	12.86	17.07	21.29	61.61

n: number of samples, mean value: three replicates of each soil data.

3. Results and Discussion

3.1. Soil Properties and Elemental Concentrations. The mean and range of soil pH values are shown in Table 1. The results showed slightly alkaline values varying from 8.20 to 8.45. The EC values also varied between samples, in a range from 6.95 to 13.29 mS m⁻¹. The EC value near the gold washing place of ore was higher than grassland and village areas in effluent draining from the mine wastes. Total C and N contents varied from 5.40 to 57.6 g kg⁻¹ and from 0.37 to 5.30 g kg⁻¹, respectively. The relative percentage of clay, silt and sand in the soils were in the range 0.53–27.57%, 10.89–38.18%, and 41.85–78.57%, respectively. The texture of most of soils was classified as clay (C) and silty clay loam (SiCL). The contents of major elements in soil samples measured by PIXE are shown in Table 2. The results indicate that the soil samples have elevated Ca (24844–98334 mg kg⁻¹) and Cl (6092–33356 mg kg⁻¹) contents in the gold washing area. The soil samples in this study showed alkaline pH and high Ca content, suggesting that considerable amount of CaCO₃ might be present in these soils. The alkaline pH of the soils in the studied area and the high content of calcium play an important role in metal stability, in such a way that there is no transfer of soluble TEs and they are being only mobilized in particulate form. Consequently, the decreases in pH mobilize significant amount of the metals which are present in these soils since carbonates are one of the major sinks for TEs [13]. The behavior and bioavailability of the TEs in soil are primarily controlled by pH. Figure 2 shows the concentrations of Cu, Zn, As, Sr, and Pb in different sites of the three studied areas. In the grassland area, however, the average concentrations of Cu and Zn are close to the world average of 30 and 90 mg kg⁻¹ respectively, reported by Bowen [14], while the mean contents of Sr for the studied areas varied from 13.9 to 202 mg kg⁻¹ which are lower than world average of 240 mg kg⁻¹. The concentration of Cu in the site M6 of gold washing area was 407 mg kg⁻¹, exceeding the concentration of 35 mg kg⁻¹ reported by Bowen [14]. Elevated concentrations of Zn were found in surface soils at the gold washing area and village, ranging from 29.2 to 223 mg kg⁻¹. Soil texture has a major influence on

trace metal concentrations, with heavy textured soils higher concentrations of Cd, Co, Cr, Cu, Ni, and Zn than light soils having higher Pb concentrations than other soils [15]. In the contrast of this, in our study Pb concentration was higher in clay loam soils around the gold washing place. The Pb content was 637 and 257 mg kg⁻¹ at the sites V1 and V3. For village, Pb can increase through its various applications such as automobile components, solder in electronics and lead batteries. Near the gold washing place, at the sites M2 and M5 the Pb contents were 231, and 236 mg kg⁻¹. Arsenic contents were at the sites M2 M7, 573 and 245 mg kg⁻¹, respectively, around gold washing place. It illustrates that gold washing place was significantly affected by waste tailings from mining. The elevated concentrations of As and Pb are a matter of serious concern because they are carcinogenic, toxic at low concentrations, and bioaccumulative in organisms. The TEs analysis showed that near the gold washing place soils where polluted by TEs such as Cu, As, and Pb. The contents of Cu, Zn, As, and Pb in soil around gold washing place are separately 109, 146, 167, and 86 mg/kg in average, which are 3.6, 1.6, 27.8, and 2.5 times those of [14]. A major source of the TEs in the gold-mining area is the oxidation of sulphide minerals, since most of the TEs are known to occur mainly as sulphides and associated with gold minerals. In the process of mineral crushing and other mining activities, these TEs can be released in the environment.

Finally, enrichment factor (EF) was used to assess the level of the contamination and the possible anthropogenic impact in soils from the mining activity. To identify unexpected metal concentration, geochemical normalization of the TEs concentrations with respect data to a conservative element, such as Al, Fe, and Si, was employed. Several authors have successfully used iron to normalize heavy metal contaminants [16, 17]. In this study, the iron was used as an element to differentiate natural from anthropogenic components. The enrichment factor of soil (EF_{soil}) is relative abundance, with regards to iron, of one element (M) in a soil compared to its relative abundance (M/Fe) in control site: $EF_{soil} = (M/Fe)_{soil} / (M/Fe)_{control}$. Table 3 shows EF values of Cu, Zn, As, Sr, and Pb for grassland, village, and gold washing places. The EF_{soil} for Cu, As, and Pb had the highest around

TABLE 2: Major element contents in soil samples.

Sampling site	Concentration in dry sample, mg kg ⁻¹									
	Mg	Al	P	Cl	K	Ca	Ti	Mn	Fe	
Grassland (<i>n</i> = 8)	Range	2535–12328	9352–47542	834–2341	1277–23692	2293–19401	4979–3011	1789–2103	143–1540	1099–60423
	Mean	5636	18024	1435	6472	5402	19132	1317	723	8202
	SD	325	677	420	1043	651	708	153	177	1132
Village (<i>n</i> = 9)	Range	3073–13446	11822–36241	861–11015	1190–35661	3671–11918	4203–3860	2582–6789	287–1300	8204–61090
	Mean	7600	23000	2621	14332	7187	23860	2992	713	33106
	SD	405	1345	512	487	570	819	211	171	1498
Gold washing place (<i>n</i> = 7)	Range	9733–19753	14210–52998	895–3963	6092–33356	6403–29740	4844–9833	385–4774	62.3–937	6043–58201
	Mean	15025	36124	2168	23806	13993	50434	2713	555	35426
	SD	515	1563	534	504	678	1568	187	210	1679

n: number of samples, mean value: three replicates of each soil data, SD: standard deviation.

FIGURE 2: Contents of toxic elements in surface soil samples (a) for Zn, Cu, Sr and (b) for As and Pb.

TABLE 3: Enrichment factor of the determined toxic elements in the areas of grassland, village, and gold washing.

Element	Enrichment factor		
	Grassland	Village	Gold washing
Cu	1.29	1.56	4
Zn	1.21	1.14	1.79
As	3.26	3.34	30.52
Sr	0.39	0.28	0.51
Pb	3.49	4.08	4.37

the gold washing place, indicating that around mining area soil was highly enriched by those elements.

More interesting information was extracted from correlation analysis between contents of the determined elements for the different studied areas. A considerably significant high correlation was observed between Mn–Fe for the village ($r = 0.927$) and the gold washing ($r = 0.905$) places. In the gold washing place, there were significant correlations for Sr–Pb ($r = 0.847$), Mn–As ($r = -0.773$), Fe–As ($r = -0.774$) and As–Sr ($r = 0.778$) which reflects the presence of TEs in the minerals of these areas.

3.2. Concentrations of TEs in Ground Water. The concentration of major and TEs in drinking water are given in Table 4. The various TEs present in ground water are originated from the anthropogenic activities and compositions of the soils and rocks it comes into contact with. Also the mobility

TABLE 4: Analytical data of drinking water samples.

Parameters	UM	Sample ID		
		GW1	GW2	GW3
pH		8.34	8.64	8.71
Na	ppm	11.2 ± 0.2	10.3 ± 0.1	21.5 ± 1.2
Mg	ppm	5.11 ± 0.2	9.05 ± 0.2	24.6 ± 1.9
Al	ppm	0.982 ± 0.122	0.524 ± 0.145	0.481 ± 1.142
Si	ppm	8.12 ± 0.40	10.2 ± 0.5	11.8 ± 0.6
S	ppm	7.41 ± 0.24	16.9 ± 1.6	26.1 ± 2.1
Cl	ppm	11.0 ± 0.2	12.3 ± 0.3	25.7 ± 1.5
K	ppm	2.50 ± 0.17	1.35 ± 0.15	2.62 ± 0.21
Ca	ppm	31.0 ± 1.3	16.2 ± 1.2	32.3 ± 1.9
Mn	ppm	1.28 ± 0.21	2.79 ± 0.4	0.57 ± 0.07
Fe	ppm	65 ± 2.3	59 ± 2.1	14.5 ± 1.2
Ni	ppm	0.54 ± 0.08	0.65 ± 0.08	0.41 ± 0.1
Cu	ppm	8.6 ± 1.2	1.7 ± 0.3	1.3 ± 0.2
Zn	ppm	5.3 ± 0.4	39 ± 2.1	9.5 ± 0.9
As	ppm	<LOD (<0.002)	<LOD	<LOD
Pb	ppm	<LOD (0.001)	<LOD	<LOD

TABLE 5: Intercomparison of trace element concentrations (mg kg^{-1}) in human hair.

Element	UM	Our study (PIXE)		Japan (PIXE) normal [21]	Mongolia (PIXE) normal [20]	Philippines (PIXE) miners [19]
		Normal ($n = 20$)	Miners ($n = 25$)			
Ca	ppm	2096	524–2259	1890	411	383.1–5363
Ti	ppm	80.4	21.8–42.2	10.4	17.6	0–6.2
Mn	ppm	7.93	1.6–43.1	1.62	9.5	0.1–289.6
Fe	ppm	25.1	52.1–142	29.1	126	10.8–222.1
Ni	ppm	1.7	0.2–4.9	2.05	1.6	0–3.5
Cu	ppm	4.9	3.5–17.9	42.7	13.7	5.6–138.2
Zn	ppm	452	155–293	238	218	55–972.9
As	ppm	1.8	2.1–5.5	0.23	1.3	0–9
Pb	ppm	8	5.4–28.9	37.3	6.6	1.1–87.5
Sr	ppm	3.3	7.5–22.6	6.04	3.9	0

and behavior of TEs in natural waters largely depend on pH. The pH of the ground water samples is ranging from 8.34 to 8.71, showing slightly alkaline values due to natural and anthropogenic origin. The contents of Fe, Cu, and Zn are ranging in $14.5\text{--}65\text{ mg L}^{-1}$, $1.3\text{--}8.6\text{ mg L}^{-1}$, and $5.3\text{--}39\text{ mg L}^{-1}$, respectively. The samples GW1 and GW2 showed the highest Cu and Zn values 86 and 390.5 mg L^{-1} respectively, which were above the WHO maxima allowed concentrations 2 and 150.5 mg L^{-1} , respectively [18]. Further from Table 3, it is seen that the concentrations of Mn are ranging from 0.5 to 2.79 mg L^{-1} , while Ni concentration varies between 0.41 and $0.652.79\text{ mg L}^{-1}$. The Mn, Fe, and Ni concentrations of drinking waters exceed the WHO values: 0.5 mg L^{-1} , 0.1 mg L^{-1} , and 0.1 mg L^{-1} for Mn, Fe, and Ni, respectively. Among the TEs, As and Pb are highly toxic even

at very low levels and are classified as a possible human carcinogens. The concentrations of As and Pb in the potable water samples were lower than the maximum allowed values according to the US Environmental Protection Agency (EPA) level of $10\text{ }\mu\text{g L}^{-1}$ and $15\text{ }\mu\text{g L}^{-1}$ for As and Pb in drinking water.

3.3. TEs in Human Hair of Miners. After acid digestion of human hair samples, a $20\text{ }\mu\text{L}$ aliquot of the solution was dropped, air-dried on the Nuclepore track etch membrane (Whatman, New Jersey, USA), and irradiated by the 2.5 MeV proton beam from the single-end-type Van de Graaff accelerator. The elemental concentrations in biological samples vary considerably due to geographical differences, nutritional status, and environmental factors. The present results were

compared with other results reported by several researchers and the ranges of worldwide mean values. Table 5 shows the comparative results. The mean concentrations of Ca, Ti, As, and Sr in Mongolian miner's hair were higher than those in the hair of noncontaminated people in Japan, Mongolia, and Philippines [19, 20]. The level of Cu, Zn, and Mn is not higher in the hair samples from the people in these countries. The maximum concentrations for As and Pb, 5.5 and 38.9 ppm, are clearly higher than those normal Japanese people which are 0.25 and 29.2 ppm (Sera K. et al., [21]). It is shown that the difference of the average element contents in hair of healthy people and miners, in most cases, is statistically not significant. The positive correlations were observed for Zn and Fe; Mn and Ca; Sr and Zn of miners. The correlation matrix gives us information about relationships between two metals but does not take into consideration of the presence of other elements.

4. Conclusions

The present study evaluated the levels of TEs (Mn, Cu, Ni, Zn, As, Sr, and Pb) in surface soils, human hair, and potable water around the small-scale mining of Boroo area, Mongolia. The surface soil samples in this study showed alkaline pH and high Ca content, suggesting that considerable amount of CaCO_3 might be present in these soils. The soil of gold washing area is polluted by Cu, As, and Pb. The contents of Cu, Zn, As, and Pb in soil around the gold washing place were 109, 146, 167, and 86 mg/kg in average, respectively, which were 3.6, 1.6, 27.8, and 2.5 times those of the reference. Significant correlation was observed around gold washing place between Mn–Fe, Mn–As, As–Sr, and Sr–Pb reflecting the presence of TEs in the soils of these areas. Also, the EF_{soil} was for Cu, As, and Pb in the highest values around the gold washing area, indicating that the soils around mining area were highly enriched by those elements. The Mn, Fe, and Ni concentrations of drinking waters exceeded the maximum values allowed by the WHO: 0.5 mg L^{-1} , 0.1 mg L^{-1} , and 0.1 mg L^{-1} for Mn, Fe, and Ni, respectively. The mean concentrations of Ca, Ti, As, and Sr in hair of Mongolian miners were higher than those in the hair of noncontaminated people in Japan, Mongolia, and Philippines. Positive correlations were observed for Zn and Fe, Mn, and Ca, Sr and Zn in the hair samples of the miners. This study showed that environmental pollution around the small-scale mining gives a serious attention to local government and responsible environmental regulation bodies.

Acknowledgments

The authors wish to thank Professor Kh. Tsookhuu, National University of Mongolia, for his great help in field trip. B. Oyuntsetseg is most grateful to the Japanese Society for Promotion of Science (JSPS) for the financial support. The authors wish to thank the anonymous reviewers for constructive comments.

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