RESEARCH ARTICLE



Arsenic distribution and pathway scenarios for sediments and water in a peri-urban Mongolian small-scale coal mining area (Nalaikh District, Ulaanbaatar)

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Abstract

The distribution of arsenic (As) in environmental compartments is investigated in the Nalaikh Depression of N-Mongolia. In Nalaikh, lignite coal is mined by artisanal small-scale mining (ASM) approaches. Because As is often associated with sulfuric minerals in coal, it was hypothesized that enrichment of As is related to coal ASM. A second hypothesis considered coal combustion in power plants, and stoves are a key source of As in the local environment. Three mobilization and distribution scenarios were developed for potential As pathways in this semiarid environment. About 43 soil and 14 water sites were analyzed for As concentrations and meaningful parameters in soil and water. About 28 topsoil samples were analyzed in surface-subsurface pairs in order to identify potential eolian surface enrichment. Additionally, fluvial-alluvial sediments and geogenic and anthropogenic deposits were sampled. Water was sampled as surface water, groundwater, precipitation, and industrial water. Results show that As does not pose a ubiquitous risk in the Nalaikh Depression. However, locally and specifically in water, As concentrations may exceed the WHO guideline value for drinking water by up to a factor of 10. A carefully selected sampling strategy allows the evaluation of the distribution scenarios, which reveals a combination of (a) geogenic As in groundwater and distribution via surface water with (b) anthropogenic As redistribution via eolian pathways. An immediate linkage between As redistribution and coal mining is not evident. However, As distribution in fly ash from coal combustion in the local power plant and yurt settlements is the most likely As pathway. Hence, the results indicate the potential influence of diffuse, low-altitude sources on As emission to the environment. As such, this study provides a good example for As distribution under semiarid climate conditions influenced by geogenic and anthropogenic factors.

Keywords Arsenic · Water · Soil · Sediment · Mongolia · Coal mining · Artisanal small scale mining · Fly ash

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Introduction

Mining has become a major industrial branch of the Mongolian economy since extensive coal and ore deposits were discovered during the previous century and particularly since the year 2000 (Cane et al. 2015; McIntyre et al. 2016). In recent years, extractive industries made up about 20% of the Mongolian GDP and about 90% of Mongolia's exports (Bataarzorig et al. 2018). Mine sites and related processing facilities exert unquestionable influence on the surrounding landscape, while also exposing the population to potentially hazardous mining impacts. Additionally, Mongolia experienced a rapid population growth of almost more than 47% during the past 25 years (1992 to 2017; 2,158,362 and

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3,177,899, respectively; MSIS 2018), increasing the pressure on efficiency levels of economic and social systems. In turn, rising energy and raw material demands have driven further expansion of mining activities that influence and even disturb natural surfaces and related subsystems (Farrington 2000; Bolormaa et al. 2012; Kim et al. 2014a; Pfeiffer et al. 2014). Assessing these impacts is a fundamental step in order to keep a sustainable balance between desired economic benefit and deterioration in health and environmental concerns.

One specific feature of Mongolia's booming mining sector is the coexistence of major mining operations and artisanal and small-scale mining (ASM). ASM activity occurred during the decades since 1990 and confronted Mongolia's regulatory authorities with this informal sector (Grayson et al. 2004). Assessing environmental impacts in ASM areas is a key prerequisite for regional land and water management (Cane et al. 2015; Suvd et al. 2015). Publications focusing on ASMrelated livelihoods in Mongolia further support this observation (Navch et al. 2006; IRIM 2011; Purevjav 2011; Smith et al. 2016).

Mining areas in Mongolia are source areas of environmental pollution by heavy metals and metalloids including As (Pfeiffer et al. 2014; Batbayar et al. 2018). Coal ASM activities are generally not associated with severe emissions of chemical pollutants. However, extraction procedures lead to significant dust mobilization (Bian et al. 2010; Pandey et al. 2016) including remobilization of dust by driving vehicles (Goossens and Buck 2009). Furthermore, combustion in power plants or settlements deteriorates air quality, including the emission of environmentally effective substances generated during the combustion process, e.g., SO_2 and NO_x but also As-laden ash (Matschullat 2000).

Arsenic holds a serious hazardous potential under reductive conditions in Quaternary alluvial sediments and low pH values, which typically occur under humid climates (e.g., Bangladesh, India; Chatterjee et al. 1995; Herath et al. 2016). Besides, regions under arid climate and alkaline soil pH conditions may experience As mobilization, including the potential to enter regional food chains (Southern Mongolia: Olkhanud 2012; Nriagu et al. 2013; Inner Mongolia: Smedley et al. 2003; Neidhardt et al. 2012). Arsenic, especially arsenite and arsenate, are carcinogenic, if regularly consumed. However, since As geochemistry is complex (Ballantyne and Moore 1988; Welch and Stollenwerk 2003; Ravenscroft et al. 2009; Kim et al. 2014a), a variety of environmental parameters need to be investigated in this concern.

In Mongolia, elevated As concentrations in surface waters have been reported from river catchments exposed to metal mining activities (Inam et al. 2011; Bolormaa et al. 2012; Purevdorj 2012; Pfeiffer et al. 2014; Hofmann et al. 2015, Batbayar et al. 2017, Batbayar et al. 2018, Yondonjamts et al. 2019). Furthermore, As bioaccumulation was identified in river fish, including some species targeted by recreational and small-scale commercial fishery (Kaus et al. 2016). Although As often acts as a substitute for S in minerals that are frequently present in coal deposits (e.g., arsenopyrite, pyrite; Ballantyne and Moore 1988), only limited information on As enrichment in the environment is available for Mongolian coal mining areas. High As concentrations in coal deposits and surrounding compartments of the biosphere are reported from China (Smedley et al. 2003; Wang et al. 2006; Tang et al. 2018; Zhang et al. 2018). Moreover, in urban centers of Mongolia, the combustion of coal has been linked to As enrichment in soils and the biosphere (Kasimov et al. 2011; Sorokina et al. 2013).

A pilot study in summer 2015, screening the geoecological situation around Nalaikh, a peri-urban district of Mongolia's capital Ulaanbaatar, showed slightly elevated As contents in urban soils and alluvial sediments. A strong informal coal ASM sector has developed in Nalaikh since the political shift in the early 1990s. Coal is the major energy resource of Mongolia and not only used in power stations but also in gers (Mongolian yurts) and ger settlements. Nalaikh's surroundings are extensively used for pasture (mostly cattle and sheep). Thus, animals are exposed to potentially harmful As concentrations in their environment. Based on this background, this study has the following objectives: (a) assessment of As contents and concentrations in the environmental subsystems, such as sediments and water; (b) evaluation whether As enrichment in Nalaikh's environmental system might be related to (ASM) coal mining activities; (c) identification of potential As distribution pathways in this semiarid, coalmining-affected area and how these compare to other regions; and (d) assessment of potential risks, risk areas, and prevention measures against As exposure of people and livestock on these pathways, if necessary.

This study is related to a project developing a rehabilitation concept for the Nalaikh mining area (MA). Within this project, besides the environmental compartments soil, water, and air, socioeconomic conditions were also taken into account to derive recommendations for a better understanding and management of Nalaikh's environment. The developed rehabilitation concept already proved its transferability to other mining products than coal or to various ecozones (Knippertz 2005). For geochemical and hydrochemical analyses, in total, 43 soil sites and 14 water sites were sampled in October 2016 in order to assess the state of the terrestrial and aquatic environment of Nalaikh (Walk 2017).

Study area

Figure 1 shows the topography of the study area, ca. 35 km southeast of Ulaanbaatar, in a depression of approximately 25×20 km within the Tuul River catchment. The geology of the Nalaikh Depression is represented by Devonian and



Fig. 1 Topography of the Nalaikh Depression and its surrounding

Carboniferous sandstone with thicknesses of up to 3000 m (MNEC 2010), which had been breached by granite plutons during the Triassic and Jurassic (Fig. 2; Dagwa-Ochir et al. 2005). Synchronously with magmatic activity, an extensive stress field formed the Nalaikh Depression, including coal formation during the Lower Cretaceous (MNEC 2010). During the Neogene and the Quaternary, the Nalaikh Depression experienced the accumulation of clastic sediments originating from the surrounding Mesozoic and Paleozoic mountain ridges. Except for some outcropping bedrock peaks, presumably late Quaternary unconsolidated eolian mantles of 0.5 to 3 m thickness and composed of sandy silt and local debris cover the area. They are the dominant substrate for steppe soil formations such as widespread kastanozems. The immediate surrounding of stream channels shows a rather alluvial sedimentology comprising reworked and layered silts and sands.

Mongolia is characterized by continental climate conditions exhibiting high annual air temperature amplitude and low precipitation (Angerer et al. 2008; Karthe 2018). Nalaikh's climate conditions are well represented by climate data from Ulaanbaatar (ca. 100 m lower and ca. 35 km to the northwest). Annually, precipitation sums up to about 250 mm, while almost 70% fall during the summer months. The mean annual temperature is just below 0 °C. In January and July, mean air temperatures are at -21 °C and 16 °C, respectively. Annual potential evapotranspiration is about thrice the precipitation causing arid conditions throughout the year (1982–2012; Climate-data.org). The Nalaikh Depression exhibits discontinuous permafrost of up to 3 m thickness (Wu et al. 2009a).

The Nalaikh Depression is drained by the ephemeral Nalaikh River, flowing north as a tributary of the Tuul River (Selenge-Lake Baikal catchment) (Fig. 1). Additionally, a minor ephemeral channel drains the Nalaikh residential area about 4 km NW of the Nalaikh River. The groundwater table varies between 2 and 45 m below the surface in Quaternary alluvial sediments (Online Resource 2). *Bus Nuur* (Fig. 3f) is a shallow waterfilled depression, presumably originating from the collapse of a *pingo* (a depression formed by the melting of permafrost). Under the current hydrographic conditions, it is not a part of the Nalaikh River catchment. Bus Nuur is shallower than 3 m and the water surface occupies ca. 43,500 m² (April 2016). However, locals report a continuously decreasing size of Bus Nuur during the previous decade (see also Online Resource 6). It represents an important drinking water source for the local livestock.



Fig. 2 Geological map of the Nalaikh Depression; the dashed line indicates the course of a cross section shown in Online Resource 1

Land use in the study area can be separated into residential areas, military, an industrial area that contains brickyards, a chicken farm, a slaughterhouse and cement factory, a now defunct glass factory (Perlinger and Urban 2018), and severely disturbed surfaces due to mining activities (see Online Resource 3). Furthermore, single features are the Nalaikh coal power plant and some isolated industrial edifices. Moreover, all areas around Nalaikh are used as pastures. The residential area is located in the northwestern part of Nalaikh. It comprises a small urban part (ca. 0.8 km²) and a larger area occupied by ger settlements (ca. 5.6 km²). Beside the coal power plant near the urban center, the area south and southeast is dominated by coal mining and industrial use. Decades of (uncontrolled) coal extraction led to a severe disturbance of natural surfaces in an area of about 15 km². The area is characterized by numerous overburden and coal heaps, numerous ASM shaft entrances (Fig. 3b), sagging phenomena, and expansion cracks around the active open cast coal mine. Additionally, parts of the area are used as a communal waste dump site.

Methodology

Hypotheses: mobilization and distribution scenarios

Controlling factors for As (im-)mobilization in the environment are pH conditions and the redox potential. Generally, the As adsorption potential increases with positive surface charge of the sorbent (Ferguson and Gavis 1972). There are four major mobilization processes for As in the environment as presented in Ravenscroft et al. (2009) and Herath et al. (2016): (a) reductive dissolution in pH near-neutral and reducing waters, dominantly mobilizing As(III); (b) alkali desorption in waters containing dissolved oxygen, nitrate, or sulfate, dominantly mobilizing As(V) under rather high pH conditions; (c) sulfite oxidation characterized by low pH conditions, high sulfate, and, often, high iron concentrations; and (d) geothermal mobilization, characterized by high water temperatures. Regarding further As redistribution, for example, groundwater transport was identified as a potential process between sediments and water (Keimowitz et al. 2005).



Fig. 3 Photos of the study area. (a) the coal mining area east of Nalaikh; note the entrance to an ASM shaft in the approximate center of the image; (b) the natural surfaces in the area are severely disturbed by ASM coal mining and brick industry; (c) Bus Nuur (Nuur = lake; < 3 m deep; ca. 43,500 m2 in April 2016) represents an important surface water resource for livestock in the area

Moreover, Kim et al. (2014b) revealed eolian redistribution of As-laden particles in a semiarid mining region.

Similar to Keimowitz et al. (2005), we set up three mobilization and distribution scenarios (MDS; A, B, C) according to As geochemistry and prevailing environmental conditions potentially relevant for the study area. Against this background, we developed the sampling strategy and further conducted data interpretation.

MDS-A represents As mobilization and distribution by groundwater. In case of As-containing lithologies acting as aquifers, As might be mobilized in dependence of prevailing pH and redox conditions, both via reductive dissolution and alkali desorption. Reducing conditions may also develop in the presence of solid organic matter (e.g., coal deposits), facilitating As release from iron oxides and sulfides (Herath et al. 2016). Thus, dissolved As may become available in groundwater wells in the Nalaikh Depression. Additionally, As-laden waters might be contributed to intermediate and surface runoff. However, since As is adsorbed to mineral surfaces over a wide pH range, a long lateral transport is not likely (Stollenwerk 2003).

MDS-B considers As being exposed during ASM activities and dissolved in water. Under aerobic conditions large amounts of As may be released via sulfide oxidation progressively leading to a lowering of pH conditions. Accordingly, acid mine drainage (AMD) in the surrounding of coal mining activity may deliver strongly As-enriched waters due to sulfide oxidation (Langmuir 1997; Goldhaber et al. 2003). Particularly, tailing deposits exposed to precipitation are likely affected by this process (Ravenscroft et al. 2009). AMD is acknowledged as a serious threat in other regions of Mongolia (e.g., Shariin Gol coal deposits, World Bank 2011). From a geological and public health perspective, it would be problematic, if such anthropogenically Ascontaminated waters infiltrate and reach groundwater aquifers.

MDS-C outlines atmospheric emission, transport, and deposition of As on soil surfaces and water bodies. Arsenic, as mineral compounds or adsorbed to coal or dust, may be emitted via surface deflation processes (Kim et al. 2014b), volatized from soils at low temperatures, or be emitted by industrial and residential facilities and fossil fuel driven traffic (Matschullat 2000; Smedley and Kinniburgh 2002). Regarding Nalaikh, this scenario comprises the deflation of As from the ASM area or emission from the power plant and ger settlements. In both cases, As distribution would result in a dominant NW-SE-oriented distribution of As in soils and water, according to the dominant wind direction. It is likely that local emissions from coal combustion in Nalaikh also lead to the deposition of dust particles enriched with heavy metals (Pötter 2017). Moreover, the eolian transport of dust particles originating from Ulaanbaatar, which suffers from some of the world's most polluted air during the winter months (Cavanaugh 2017; Davy et al. 2011; Ganbat and Baik 2016; Hasenkopf et al. 2016; Nishikawa et al. 2011), cannot be excluded and has been instrumentally observed (S. Lodoysamba 2016, pers. comm.). Investigations on pollution gradients

between Ulaanbaatar and Nalaikh are still needed and would allow for the distinction between regional and local distribution patterns.

Although the scenarios channelize the discussion about As distribution in the study area, a potential overlap of scenarios needs to be considered in the discussion.

Sampling methods and strategy

In order to investigate the distribution pathways of As in the surroundings of Nalaikh and its mining area, surface sediments, soils, and water (standing and running; surface water and groundwater wells) were sampled. Sites were carefully selected according to potential topographically and meteorologically controlled distribution pathways of As-laden water and/or particles. For example, a NW-SE and SSW-NNE soil catena was sampled, hypothesizing an eolian distribution of As according to the dominant wind direction. The S-Nflowing drainages and adjacent sediments (e.g., Nalaikh River east of the ASM area) were investigated in order to analyze the As distribution in a leeward location of a potential source (Fig. 4a). We inspected anthropogenic sites, such as the ash basin of the Nalaikh coal power plant and the severely disturbed surfaces of the ASM area. Figure 5 indicates the sites of, in total, four categories of soil and sediment samples: soil A-horizons (A), fluvial-alluvial sediments (FL), geologic substrate (G) representing background samples for the interpretation, and anthropogenic substrate (Y), referring to heaps, tailings, and ash depositions.

Besides sampling the different substrates, soil A-horizons were sampled in two depths (0-5 cm and 5-15 cm) in order to

investigate potential surface enrichment of As, indicating an eolian input using the ratio between surface and depth As contents (modified from Kim et al. 2014b). The sampling intervals were selected based on the evaluation of the sampling pits to represent undisturbed surfaces. Furthermore, we considered net dust accumulation being 5 cm at maximum during the industrial period of Nalaikh and Ulaanbaatar, i.e., since the beginning of the twentieth century. Thus, this interval is representative for a potential anthropogenic As dispersal, while, in contrast, the 5–15 cm interval represents pre-industrial time.

Water sampling took place in October 2016 and sampling sites (Fig. 4b; Table 1) were selected according to hypothetical As distribution pathways (eolian distribution after storage or combustion of coal in the mining area versus hydrologic distribution by surface or groundwater). In addition to the two streams draining the Nalaikh Depression, groundwater wells (tapping the Quaternary aquifer) and Bus Nuur were sampled. One sample was taken from fresh snow to evaluate "blank" precipitation hydrochemistry. Unfortunately, no surface water sample could be obtained from the mining area. Lacking surface water confirms a deep groundwater table, maybe also influenced by the severe mining activity in the area, inhibiting the water accumulation on the surface. Furthermore, no pumps are required among the mine sites to avoid groundwater intrusion, what confirms a groundwater table lower than the mine shafts' depth (up to ca. 50 m; Online Resource 2). Weather conditions during sampling were dry and stable. Snow melt of an average ~2 cm thick snow cover occurred during field work, yet no significant increase in discharge could be identified. Samples are categorized into surface water



Fig. 4 Sampling site distribution of (a) soil and sediment samples and (b) water samples



Fig. 5 (a) I_{geo} values of soils and sediments in the Nalaikh Depression; (b) surface enrichment of As in soils calculated from two depths (0–5 cm; 5–15 cm). Dashed lines indicate transects presented in Fig. 6a and b (for geology legend see Fig. 2; for land use legend, see Fig. 4)

(SW), groundwater (GW), industrial waste water (YW), and precipitation (P). Three aliquots of ca. 50 mL were taken per sample. Two aliquots were filtered through a 0.45 μ m mesh to separate suspended particles. The third aliquot was kept unfiltered. The latter and one filtered aliquot were acidized to pH < 2 with concentrated HNO₃ to avoid precipitation of heavy metal complexes. The other filtered sample was kept at original pH and used for major ion analyzes. All samples were kept airtight and at 7 °C until analyzed. During sampling,

water samples were analyzed for dissolved oxygen, pH, electric conductivity, and temperature using a WTW Multi3430 device.

Laboratory

Soil pH was determined on 5 g air-dried sample material stirred in 15 ml 0.01 M CaCl₂ solution and measured with a

Table 1 Water samples and measurement procedures; T temperature; κ specific electric conductivity; * dilution factors for AAS measurements of Na⁺ and K⁺ are given in parentheses.

Sample	N [°]	E [°]	Description	On-site meas.*	AAS**
P-01	107.242636	47.770743	Snow	Т, pH, O2, к	Na (100), K
SW-01	107.262066	47.778414	Stream	T, pH, O2, к	Na (100), K
SW-02	107.263714	47.780148	Stream	T, pH, O2, к	Na (100), K
SW-03	107.267169	47.7847	Stream, partly frozen	T, pH, O2, к	Na (100), K
SW-04	107.2717	47.790317	Stream, partly frozen	T, pH, O2, к	Na (100), K
SW-05	107.27175	47.791717	Pond	T, pH, O2, к	Na (100), K
SW-06	107.300975	47.70911	Stream pond	T, pH, O2, к	Na (100), K
SW-07	107.313301	47.726035	Stream pond	Т, рН, О2, к	Na (200), K (10)
SW-08	107.341006	47.778318	Lake (Bus Nuur)	T, pH, O2, к	Na (200), K (10)
GW-01	107.269095	47.791272	Artesian well	T, pH, O2, к	Na (100), K
GW-02	107.256882	47.769692	Well	T, pH, O2, к	Na (100), K
GW-03	107.265481	47.788525	Well	T, pH, O2, к	Na (100), K
GW-04	107.265007	47.686998	Spring	T, pH, O2, к	Na (100), K
YW-01	107.257789	47.769474	Ash basin (power plant)	Т, рН, О2, к	Na (100), K

All filtered and unfiltered aliquots were analyzed for Mg and Ca by titrimetry and for Mn, Fe, and As by ICP-MS

WTW pH-Electrode SenTix 940 after 1 h of resting and calibration to pH 4.01, 7.0, and 10.011.

For element geochemistry of soils and sediments, samples were sieved to the $< 63 \mu m$ fraction and dried at 105 °C. In case of too little material in the desired size fraction, samples were ground in order to achieve sufficient material in the appropriate range for pellet pressing. In further interpretation, only equally processed samples and their data were compared (e.g., calculation of surface enrichment). About 8 g of the samples were mixed with 2 g of binding wax (FLUXANA CEREOX Licowax) and pressed to pellets for the Spectro Xepos X-ray fluorescence (XRF) device. Each sample pellet is measured twice including a 90° rotation after the first measurement to avoid matrix effects of a single measurement. Online Resource 4 provides quality control on each element's error range during XRF measurements. For As, the errors range between 0.9 and 8.8% of the measured values with a mean error of 3.9% (median 3.7%, standard deviation 1.3%). Most of the other elements reveal even narrower error ranges.

In order to evaluate samples for their pollution load in comparison to the surrounding natural substrate, the geoaccumulation index I_{geo} (Müller 1979) was used:

$$I_{geo}(As) = In(c_{As}/1.5 \times b_{As})$$
(F1)

where c_{As} is the As concentration of the respective sample and b_{As} represents the geochemical background value. Since global As concentrations in the upper crust vary strongly (Taylor and McLennon 1995; Ketris and Yudovich 2009), a regional background value should be applied (Rubio et al. 2000). Multiplication by 1.5 is applied to account for lithologic variations of the representative geogenic background. Thus, the As content of the silty-sandy deposits (G-01) typical for the area and without any indication of anthropogenic interference was used as a reference for the geogenic background value ($b_{As} =$ 8.6 mg/kg).

Quantification of As surface enrichment via eolian (re-)distribution of As-laden particles in topsoils was conducted following Kim et al. (2014b) with SE_{As} being the ratio between topsoil (here: 0–5 cm) and subsoil (here: 5–15 cm) As concentrations.

Water samples were analyzed in the Central Geological Laboratory (CGL) in Ulaanbaatar. Filtered aliquots were used for determination of major cations by Atomic Absorption Spectrometry (AAS; Na, K in emission mode) and EDTA titrimetry (Mg, Ca). Table 1 shows the applied methods and respectively needed dilution factors (Skoog and Leary 1996). Among titrimetry, detection limits of Mg and Ca were 0.5 mg/L. Quality control was conducted using sample duplicates and standard solutions, revealing

uncertainties of $\leq 0.3\%$ and $\leq 7\%$ among AAS and titrimetric measurements, respectively (Table 2). Inductively coupled plasma mass spectrometry (ICP-MS; Thermofisher, ICP-MS Xseries II) was applied at CGL for the determination of Mn, Fe, and As in filtered and unfiltered aliquots. The "Trace Metals in Drinking Water" (TMDW; by High-Purity Standards) was used as certified reference material. Two sample duplicates were measured for quality assurance. Relative deviation between the two measurement runs were 0% for Fe, Mn, and As. Analysis of TMDW revealed measurement uncertainties of 7%, 0%, and 3.8% for Fe, Mn, and As, respectively (Table 3). Detection limits for Fe is at 100 µg/L and for Mn and As at 1 µg/L.

For a further geochemical characterization of dissolved As, salinity (total dissolved solids, TDS) was estimated on the basis of measured electrical conductivity κ (*TDS* = $\alpha^*\kappa$), where α represents a coefficient (usually 0.55–0.75; Hem 1985; McNeil and Cox 2000). By linear regression between the sum of cations and electrical conductivity, α could be approximated to 0.715 (data available in Table 2).

Potential redox processes in the hydrosphere compartments were inferred based on the measured concentrations of O_2 , Fe, and Mn using the procedure of McMahon and Chapelle (2008) and Chapelle et al. (2009).

Results

Soils and sediments

Geochemical properties

In total, 28 topsoil sampling sites of kastanozem soils were described and sampled in 2 depths. Upper subsamples (suffix A, depth 0–5 cm) reflect homogenous properties of low CaO (presumably CaCO₃) contents of 15 to 45 g/kg, compared to the reference material (G-01; 159 g/kg) indicating decarbonatization of topsoils (see Online Resource 4). pH values range between 5.42 and 8.67. Lower topsoil samples (suffix B; 5–15 cm) behave similarly; however reaching the highest pH value at 9.19. CaO shows a slightly larger range of 14 to 64 g/kg, indicating a lesser degree of decarbonatization. Arsenic contents in all topsoil samples range between 4.6 ± 0.4 and 16 ± 0.5 mg/kg, indicating no significant enrichment. Surface enrichment is calculated among the pairs of topsoil samples (see 4.1.2).

Seven fluvial-alluvial sediment samples were taken, which exhibit larger heterogeneity among CaO contents than sampled kastanozems (16 to 108 g/kg). pH values are generally basic (7.47 to 9.61), however, with a clear separation between rather neutral samples (FL-01 to FL-

Table 2 Hydrochemical results and respective methods; T temperature, κ specific electric conductivity

	On site measurements			AAS		Titrimetry		ICP-MS						
Sample	T [°C]	pН	O ₂ [mg/L]	κ [μS/cm]	Na ⁺ [mg/L]	K ⁺ [mg/L]	Mg ²⁺ [mg/L]	Ca ²⁺ [mg/L]	Fe _{dis} [µg/L]	Fe _{tot} [µg/L]	Mn _{dis} [µg/L]	Mn _{tot} [µg/L]	As _{dis} [µg/L]	As _{tot} [µg/L]
P-01	5	8.4	8.98	124	292.1	2.7	1.22	24.05	<100	1010	8	131	1	2
SW-01	4.7	7.7	5.75	811	310.6	2.02	41.31	72.14	<100	<100	30	20	3	3
SW-02	4.9	8.6	11.59	982	337.9	1.46	53.46	88.18	<100	<100	19	16	2	1
SW-03	2	8.3	11.63	1300	337.6	3.4	58.32	108.22	<100	140	195	203	2	2
SW-04	1.8	8.6	14.64	1044	342	2.87	60.75	76.15	<100	<100	197	201	3	3
SW-05	5.2	8.4	10.14	1550	495.9	2.98	55.89	40.08	<100	240	40	53	4	4
SW-06	4.8	8.1	10.88	1485	403.6	4.49	75.33	108.22	<100	<100	25	25	4	5
SW-07	6.3	9.6	18.1	2920	828.9	22.14	123.93	40.08	110	330	20	63	11	14
SW-08	5.4	9.1	11.09	4080	960.8	39.02	187.11	84.17	<100	110	20	59	97	101
GW-01	2	8.0	2.36	605	301	1.57	24.3	44.09	260	250	104	103	1	1
GW-02	7.7	7.3	1.5	1368	309.5	1.72	70.47	88.18	260	150	266	269	1	1
GW-03	8.3	7.9	5.85	745	95.6	1.05	34.02	56.11	<100	<100	24	25	1	1
GW-04	2.8	7.9	8.62	465	273	0.6	29.16	60.12	<100	130	3	6	16	17
YW-01	5.2	9.5	10.43	821	339.8	4.04	21.87	120.24	<100	450	25	43	49	49
Quality control SW-05					497.39	2.98	55.89	42.88	< 100		40		4	
duplicate														
rel. Deviation [%]					0.3	0.0	0.0	7.0	0.0		0.0		0.0	
Standard solution					3.00	3.00								
Test CS-2179					3.03	3.03								
rel. Deviation [%]					1.0	1.0								
SW-06_ duplicate									<100		25		4	
rel. Deviation [%]									0.0		0.0		0.0	
Test CS-2177									107		40		77	
CRM-TMDW									100		40		80	
rel. Deviation [%]									7.0		0.0		3.8	

04) and those with significantly higher values (9.21–9.61; FL-05 – FL-07) reflecting the observed salt precipitation on floodplain surfaces. Sediments range from clayey-silty to sand-dominated grain sizes, which account for larger heterogeneity in elemental composition than among the rather homogeneous topsoils. Arsenic contents range between 4.8 ± 0.3 and 11.8 ± 0.4 mg/kg, also negligible in regard of As enrichment.

Six samples from anthropogenic substrates comprise overburden, coal heaps, and ash deposits (Y-02 was sampled in two depths, like kastanozem samples). Accordingly, parameters show the largest range of the dataset: pH and As contents range from pH 5.2 to pH 9.8 and from 7 ± 0.4 (coal flitter; Y-05) to $107.9\pm$ 1 mg/kg in coal ash (Y-06). Higher As contents compared to the reference natural substrates were also measured in overburden (Y-02A and B: 31.6 ± 0.6 mg/kg; $69.4 \pm$

Table 3 Correlation coefficients r between As and pH (based on pH index values), Fe_2O_3 , MnO, and S in upper (A-A) and lower (A-B) topsoil samples.

Sed. class	n	pН	Fe ₂ O ₃	MnO	S
A-A	28	-0.05	0.58	0.17	-0.07
p value; signif. (<0.05)		n.s.	0.001	n.s.	n.s.
A-B	28	-0.58	0.58	0.43	-0.28
p value; signif. (<0.05)		0.001	0.001	0.02	n.s.

Detailed results per sample and over a wide range of elements are presented in Online Resource 4 0.7 mg/kg), coal heaps (Y-03: 28.1 ± 0.6 mg/kg), and ash of the power plants ash basin (Y-01: 24.8 ± 0.4 mg/kg). Online Resource 4 lists all soil samples and respective geochemical results in a spreadsheet.

Spatial distribution patterns of arsenic in soils and sediments

 I_{geo} values among Nalaikh soils and sediments as well as SE_{As} values in topsoil samples and Y-02 are given in Fig. 5. According to the I_{geo} (Müller 1979), uncontaminated to moderately contaminated soils and sediments are mostly represented by samples from the MA and the urban area of Nalaikh (Fig. 5a). Values along the Nalaikh River do not show any downstream trend. While samples outside of occupied areas mostly reflect uncontaminated values, uncontaminated to moderately contaminated topsoils occur near the MA and brickyards. Since only three samples in the MA show $I_{geo} < 0$, a general accumulation of As in soils around ASM activity is inferred.

Fig. 6 Transects depicting spatial variations in soil As concentrations, parameters of surficial samples A-xx-A and pH values; their courses are shown in Fig. 5a. (a) Running NW-SE following the dominant wind direction: highest values reached in the MA; (b) following the Nalaikh River with five surface samples, however, showing slight enrichment when bypassing the MA. Error ranges of the As measurement span between 0.4 and 0.5 mg/kg (< 5.84%, see also Online Resource 4)

Relating the potential As enrichment to a specific MDS requires the identification of a dominant eolian or hydrologic distribution of As in the landscape system. Figure 5b shows SE_{As} to be rather > 1 in anthropogenically occupied or influenced areas, such as the MA (especially A-25, SE_{As} of 2.5), the residential area, and the northwestern drainage. In order to evaluate MDSs, Fig. 6 depicts the results of soil As geochemistry along two transects. Figure 6a envisages MDS-C using a NW-SE transect, following the dominant wind direction. A slight increase in As concentration and respective I_{geo} becomes apparent when the transect crosses the urban area of Nalaikh and the adjacent MA.

Soils along the Nalaikh River do not generally reflect systematic accumulation of As. However, A-18 and A-19 exhibit SE_{As} values of 0.8 and 0.53, respectively, which are the lowest SE_{As} values in this study, indicating higher As concentrations in 5–15 cm depth than on the surface layer. Sampling sites further downstream the Nalaikh River (A-20, A-21) also show $SE_{As} < 1$, which is particularly in contrast to urban areas and



the MA. The trend of higher As concentrations in relation to anthropogenic activity is also apparent in the middle part of Fig. 6b (A-06), where the transect passes by the MA in the SE.

Water

Table 2 provides details on the laboratory results of obtained water samples.

Hydrochemical properties

Waters around Nalaikh are relatively rich in Na and poor in K (except SW-07 and SW-08). Contents of dissolved Ca and Mg depend strongly on the water type. However, they are highest in surface waters. This is furthermore reflected in TDS contents. Surface waters exhibit the highest salinities of 500 to 3000 mg/L, whereas groundwater has TDS contents of < 1000 mg/L. Na⁺ / (Na⁺+Ca²⁺) indicates geochemical influence on hydrochemistry by (oceanic) precipitation (Na) versus rock weathering (Ca) (see Fig. 7a; Gibbs 1970; Appelo and Postma 2005), while an increasing TDS content indicates evapoconcentration. The sample from Bus Nuur reflects the strongest evaporation influence with a TDS content of almost 3000 mg/L, followed by standing surface waters (> 1000 mg/ L). As expected, groundwater samples exhibit a lower evaporation influence. Sample YW-01 from the power plant's ash basin does not strongly differ from the other surface waters in hydrochemical terms.

Characterizing the As variety occurring in water requires consideration of pH conditions and redox potential (here approximated by the pH, O₂, TDS, and Fe and Mn contents). pH values range between 7.3 and 9.5 with groundwater samples having pH values of around 8 and slightly below (Fig. 7b). Surface water samples are alkaline with pH levels ranging between 7.6 and 9.5 and a majority of samples between 8 and 8.6. High pH values of surface waters were found in stagnant waters such as Bus Nuur (SW-08; pH 9.1) and the lower Nalaikh River (SW-07; pH 9.6). At the same time, these two samples have the highest TDS load (Fig. 7a). The YW-01 sample from the ash basin exhibits a pH of 9.5, probably a result of exchange processes with suspended ash particles.

Oxygen content is the determining criterion for assigning redox conditions (McMahon and Chapelle 2008; Chapelle et al. 2009). All samples reveal O_2 concentrations between 5.75 and 18 mg/L, except for GW-01 and GW-02 (2.4 and 1.5 mg/L, respectively; Fig. 7b). A threshold value of 0.5 mg/L classifies all samples of this study as oxic (McMahon and Chapelle 2008).

Arsenic concentrations

In the following As concentrations refer to the measured As_{dis} concentrations. Figure 8 provides the As concentrations measured in water samples from Nalaikh. Precipitation, surface water, and groundwater samples mostly exhibit As concentrations below the WHO guideline for drinking water of 10 µg/L.



Fig. 7 General characterization of water samples from Nalaikh; (a) evaporation and weathering indices; based on Gibbs 1970. (b) O_2 vs. pH and the As concentration shown using the color scheme. *P* precipitation; *GW* groundwater; *SW* surface water; *YW* industrial

waste water; the blue dashed line at an O_2 concentration of 0.5 mg/L represents the transition from suboxic-anoxic conditions to oxic conditions



Fig. 8 The concentrations of dissolved As in water samples. Note the highest value of 97 μ g/L in Bus Nuur (SW-08). Also, the spring sample of the Nalaikh River (GW-04; 16 μ g/L) reveals a value above the WHO drinking water guideline indicating As input by groundwater. For legend on the

underlying geology, refer to Fig. 2. The landuse legend can be obtained from Fig. 5a. Bar diagram shows As concentrations in water samples. The dashed red line indicates the WHO guideline of 10 μ g/L As in drinking water

GW-01, GW-02, and GW-03 represent drinking water wells for Nalaikh's population and do not exhibit any As contamination. However, GW-04 (16 µg/L) and SW-07 (11 µg/L) slightly exceed the guideline value. Particularly SW-08 $(97 \ \mu g/L)$ and YW-01 (49 $\ \mu g/L)$) are ten and five times higher than the WHO guideline, respectively (Fig. 7). The As concentration represented in GW-04 is likely responsible for a relatively increased As input to the catchment of the Nalaikh River. Although the springs are not specific drinking water sources, they are used as such by livestock, similar to the entire Nalaikh River and Bus Nuur. The As concentrations along the Nalaikh River (SW-06; SW-07), however, are lower than in the springs (GW-04). Contrarily to those, As concentrations in Bus Nuur (SW-08) are very high, exceeding the concentration in the power plant's ash basin by almost a factor of 2.

Discussion

Geochemical interpretation

In order to identify the prevalent As species in soils and sediments, linear correlations between As concentrations, major element oxides, pH conditions, and selected trace elements were conducted (Table 4). Values for FL and Y02-Y05 are provided for the sake of completeness since their respective sample sizes are not statistically robust. Strong association of As and Fe is evident by r = 0.58 in upper and lower soils (A-A; A-B). Association with S is, however, rather low and slightly negative.

In the subsurface soils, the moderately strong and negative correlation with the pH coincides with the positive Fe correlation, indicating that As is supposedly adsorbed to pedogenic Fe oxides. This is further supported by an only weak positive correlation between As and Mn (r = 0.43). Similar results are presented in fractionation experiments by Yondonjamts et al. (2019) on topsoils from the Erdenet copper mine in Northern Mongolia. However, under the observed pH conditions, Mn oxides generally do not contribute to As adsorption due to predominantly negative surface charges (Tan et al. 2008). Association with Fe-rich, sulfuric minerals such as pyrite is unlikely, regarding the weak negative and nonsignificant correlation between As and S.

Relating pH and O_2 to As concentrations in water allows evaluation of their influence on the As-enriched waters (see above, Fig. 7b). Surface waters exhibit higher pH values and O_2 concentrations than groundwater. Specifically, SW-07 and SW-08 reveal the highest pH values, beside the industrial waste water sample YW-01 of > 9.1 and have high O_2 concentrations (> 10 mg/L). A lacking relation between As_{dis} with Fe_{dis} and Mn_{dis} and evidently high O_2 concentrations suggests that reductive dissolution of Fe minerals is an unlikely mobilization process. Furthermore, taking into account the alkaline pH conditions of all sampled waters, the influence of sulfide oxidation seems to be negligible, as it would lead to a noticeable pH reduction. Instead, desorption of As from, e.g., Fe (hydr-)oxides or coal ash is favored under the given hydrochemical conditions. Moreover, the Nalaikh River spring (GW-04) is slightly alkaline and oxic pointing to alkali desorption as the dominant mobilization process. However, As mobilization via reductive dissolution or sulfide oxidation cannot be entirely excluded in case of local pH and redox conditions change during the ascend of the groundwater in the southern part of the Nalaikh Depression before reaching the surface.

pH and redox conditions allow inferences about the As species prevalent in the respective samples. Under given pH values between 7.3 and 9.6 and respective oxic conditions, arsenate (As(V), $H_3AsO_4^{2^-}$) is the thermodynamically most stable species (Ferguson and Gavis 1972). Under variable pH conditions, other As species, such as arsenite (As(III)), may occur. However the redox potential in all samples is sufficiently high to prevent reduction to arsenite. Since As(V) compounds are generally less toxic than As(III) compounds (Ferguson and Gavis 1972; Summers and Silver 1978), the human toxicity of As in waters around Nalaikh can be considered low.

Evaluation of mobilization and distribution scenarios

In order to identify distribution mechanisms of As, MDSs were set up for a distinct area under specific framework conditions and influences, such as semiarid climate, coal mining, and urban infrastructure. The evaluation of MDSs required various parameters for verification. Although SE_{As} (Kim et al. 2014b) produced plausible results, its application as conducted here requires critical discussion. The depth intervals have to be carefully selected in order to sample representative units. So far, no reliable measurements of redistributed mining-affected sediment are available for Nalaikh. When investigating eolian (re-)distribution in rather young miningaffected landscapes (<100 years), it is probably most representative to sample only the topmost, initially compacted grains as representing the surface layer in SE calculation. This holds especially true in (semi-)arid regions where redistribution by infiltrating surface runoff can be considered low.

Geogenic As input via groundwater discharge (MDS-A)

Near Nalaikh, single, slightly elevated As concentrations in groundwaters ascending to the spring of the Nalaikh River (GW-04) can be referred to geogenic origins. The Neoproterozoic Unegt group (varieties of granitic lithologies) contains extensive sulfide occurrences (e.g., pyrite) often in association with copper, silver, and gold (Dorjsuren et al. 2006; Dril et al. 2010). It is thrusted onto the Devonian Sergelen formation composed of sandstone and quartzite south of Nalaikh (see Fig. 2; Dagwa-Ochir et al. 2005) where the Nalaikh River originates. The oxidation of pyrite is a likely explanation for elevated As concentrations (Armienta et al. 2001; Smedley and Kinniburgh 2002) in the Unegt group. After ascending along the thrust faults at the Sergelen-Unegt contact, these waters surface in the spring ponds of the Nalaikh River (GW-04). In sulfides, As is present in reduced state and potentially released via oxidation (Schreiber et al. 2003). However, also sandstones and quartzites, representing the Sergelen formation, may serve as As host rocks, potentially releasing As via alkali desorption (Schlottmann et al. 1998; Heinrichs and Udluft 1999).

Thus, the Nalaikh River is fed to As concentration around the WHO guideline (GW-04) which slightly decrease along its course (SW-06, SW-07). Probably due to freshwater input by surface runoff along the Nalaikh River, As does not concentrate or migrate downstream to reach noticeably increasing concentrations. Additionally, rather low sediment As contents around the occurrence of comparably As-rich waters (Nalaikh River, Bus Nuur; FL-04; FL-06; FL-07) do not indicate a significant pathway between hydrologically transported As into surrounding alluvial sediments.

As such, a geogenic As contribution to the surface hydrological system of the Nalaikh Depression is evident. Since the less toxic As(V) is the dominating As species and only one groundwater sample shows a slightly elevated As concentration, no serious threat is attributed to MDS-A.

Anthropogenic origin by mine water-derived mobilization (MDS-B)

Some slightly reduced As and S concentrations in the MA overburden material (Y-02-Y05) in contrast to the unweathered Cretaceous clayey shales (G-02) might indicate some sulfide oxidation, however, not severely influencing the surrounding environment. A significant finding is that groundwater samples taken from the lower Nalaikh Depression (GW-01 through GW-03) do not show any evidence of elevated As concentrations. The first reason is probably low infiltration due to dry climate conditions and, moreover, a low water demand for artisanal coal mining. However, a thick vadose zone composed of quaternary silt and sand in combination with cretaceous clayey shales forms a barrier to potentially As-enriched percolating water, even in case of higher infiltration. MDS-B is therefore considered unlikely for a substantial distribution of As to the environment of the Nalaikh Depression.Of course, higher infiltration, as common in different mines other than coal mines, may lead to increased As leaching via AMD and transport into aquifers (e.g., Cheng et al. 2009; Wu et al. 2009b).

Anthropogenic emission and eolian distribution (MDS-C)

Generally, As enrichment in soils and sediments can be considered low and without serious threats to the environmental compartments. However, geoaccumulation and surface enrichment confirm distribution predominantly to the southeast from Nalaikh, following the main wind direction (Fig. 6). This suggests an eolian distribution pathway between a rich source (city of Nalaikh, power plant, ger settlements, MA) and the deposition areas to the southeast. Since bioaccumulation in topsoils would most likely result in a distribution pattern unlike the wind-related pattern, we conclude that bioaccumulation is not the driving factor for As distribution in topsoils. Although, they might be coincidently similar, we consider the eolian distribution the most likely explanation. Highest surface concentrations in soils occur near the coal power plant, presumably related to ash deposition, in the MA characterized by widespread coal and overburden heaps. Bounds and Johannesson (2007) found As enrichment in soils around a coal shipping terminal, indicating eolian coal dust emissions being responsible for As distribution and an impact on soil geochemistry. In Nalaikh, a supraregional input from coal power plants and ger settlements in Ulaanbaatar has to be considered as an additional source of particulate matter input (Guttikunda et al. 2013). However, since studied transects follow the local characteristics in and around Nalaikh, like the power plant or the MA, this background input is not dominating local As distribution. Consequently, no As enrichment was found in soils of the Nalaikh Depression out of reach of anthropogenic features.

Apart from traffic, particulate urban emissions from Nalaikh are mostly related to coal, which, as shown, bears considerable As concentrations. Similarly to Ulaanbaatar (Guttikunda 2007; Guttikunda et al. 2013), ash from domestically and industrially burnt coal is a likely source for eolian As distribution in the Nalaikh Depression. Arsenic in the Nalaikh coal is likely related to authigenic sulfides generated during coal diagenesis. Nalaikh coal is generally low in sulfur (G-02: 15.62 ± 0.02 mg/kg SO₃; MNEC 2010), which coincides with a low As content of $2.4 \pm$ 0.1 mg/kg (Online Resource 4; Yudovich and Ketris 2005). However, As is strongly concentrated in ash during combustion (Yudovich and Ketris 2005; Ketris and Yudovich 2009), making coal ash the more significant As carrier than unburnt coal (Y-06: $107.9 \pm 1 \text{ mg/kg As}$). Batjargal et al. (2010) related high As contents in soils from Ulaanbaatar to ash emissions from the local coal power plants. For Nalaikh, the large extent of coal combustion in ger settlements and the power plant suggests a significant influence on eolian As distribution related to fly ash (Guttikunda et al. 2013; Sorokina et al. 2013; Pötter 2017). Additionally, different studies indicate that As is especially enriched in the fine ash particles compared to bottom ash (Seames and Wendt 2002; Yudovich and Ketris 2005). This puts anthropogenic ash emissions particularly in focus of As distribution in the Nalaikh Depression. A direct impact of coal extraction procedures on deflation of As-laden particles from the MA could not be separated from the urban impact by combustion. In contrast, combustion is most likely responsible for increasing As concentrations in soils downwind of Nalaikh, due to higher emissions and high As concentrations in fly ash.

The Bus Nuur special case

Bus Nuur exhibited by far the highest As concentrations in water investigated in this study (SW-08, 97 µg/L; Fig. 8). The oxic and strongly alkaline conditions suggest alkaline desorption as the major As mobilization process in Bus Nuur. The original source of such high As concentrations is, however, unclear. Similarly to the Nalaikh River catchment, the Bus Nuur catchment includes the thrust fault contact between the Unegt group and the Devonian lithology, which likely serves as a source for As via groundwater discharge (see Chapter Geogenic As input via groundwater discharge (MDS-A)). However, there are no channels or any signs of current inflow to the lake, unless unconfined surface runoff contributes freshwater during precipitation events or the snow melt period. Additionally, the regional groundwater table is significantly lower than Bus Nuur's water level, prohibiting groundwater contribution. Hence, a constant hydrogeological As input is unlikely. Locals report a Bus Nuur water level decrease of two thirds during the past decade, probably due to water removal and evaporation while the water body lacks freshwater refill. Google Earth imagery reveals a high potential variability of the lake's surface area. Thus, a ca. 24% surface area reduction is recorded between September 2013 and April 2016 (see Online Resource 6). Hence, a negative water balance might lead to an evapoconcentration of As (Drever 1982). For example, Eary (1998) predicted efficient evapoconcentration of As(V) in mine pit lakes. Gao et al. (2007) have shown a similar behavior for agricultural evaporation ponds. This interpretation is supported for Bus Nuur by TDS concentrations of up to 3000 mg/L indicating the strong influence of evaporation on the hydrochemistry of Bus Nuur (Fig. 7a). Furthermore, with a pH around 9, alkaline desorption is the most likely As mobilization process under recent conditions, which are comparable to the sites investigated by Eary (1998). Regarding presented distribution scenarios, neither the hydrologic nor the eolian pathway appears responsible for raising As to such regionally exceptional levels. Bus Nuur is cut off from natural surface and groundwater pathways (MDS-A and -B). Eolian enrichment is not reflected in adjacent sediments (FL-05, FL-06, FL-07, A-21) which would support MDS-C. Locals sporadically report that Bus Nuur was used as a waste dump in the past. Although,

explicitly, no evidence for that was found, mere speculation remains about, e.g., direct anthropogenic ash disposal leading to As enrichment in the stagnant water (e.g., Ruhl et al. 2010) of Bus Nuur. Another explanation may be a long-term concentration of As over the last decades due to a subtle input from both geogenic and anthropogenic sources (MDS-A and MDS-C) and subsequent evapoconcentration in the endorheic sink (Gao et al. 2007). Monitoring seasonal and interannual variability of As concentrations in Bus Nuur would facilitate the understanding of potential causes of As enrichment.

It is noted that one sampling campaign does not grant a representative picture of annual variability. However, occasional sampling from previous and successive years confirm that proxy results range in the same magnitude as in this campaign. Seasonal variations in As concentrations were reported by Batbayar et al. (2017) showing higher concentrations during summer time compared to spring conditions. Similarly, Yan et al. (2016) investigated seasonal variations of As in surface waters of four subregions in Lake Taihu (China). They show, by trend, highest concentrations during summer and autumn compared to winter and spring. These findings suggest a potentially lower annual average of the As concentration in Bus Nuur than measured here, since the sampling took place during autumn. A regular monitoring is a crucial part of any rehabilitation concept, and, in the special case of Bus Nuur's As concentrations, it could help to increase the representativeness of results presented here and also improve the understanding of seasonal variations and the identification of the origin of this local contamination.

Supraregional comparison

Compared to other mining regions in Mongolia, As enrichment in the environment around Nalaikh is generally considered low. Soil samples exhibit fairly low As concentrations, and, due to semiarid conditions and moderate pH values, As mobility is restricted (Ravenscroft et al. 2009). In areas of higher pH values, such as natural salt pans in the lower reach of the Nalaikh River, As(V) may be desorbed and mobilized more easily from pedogenic iron oxides (e.g., goethite, ferrihydrite; Dixit and Hering 2003; Busbee et al. 2009). Around the urban area and the MA, slightly positive variations from the As background occur. Bolormaa et al. (2012) published As concentrations of ca. 550 mg/kg in north Mongolian soils affected by mining activities. However, coal mining activity in and around Nalaikh does apparently not result in a strong As enrichment in adjacent soils, although slight surface enrichment was detected. The highest As concentrations were measured in coal and ash deposits, whereas soils and sediments were close to natural background conditions. However, in vicinity to anthropogenically modified surfaces, such as ash and coal heaps, local As redistribution is indicated by higher concentrations in the immediate surrounding of these features.

The works of Pfeiffer et al. (2014) and Batbayar et al. (2017) provide a comprehensive overview over the situation of As (and other potential pollutants) in Northern Mongolia. Northern Mongolian salt lakes tend to moderately exceed WHO guidelines (Batbayar et al. 2017), while As concentrations in NW Mongolian salt lakes are negligible (Baatar et al. 2017). Waters and ponds affected or even created by mining and power production activity often show a strong As enrichment (Pfeiffer et al. 2014; McIntyre et al. 2016; Batbayar et al. 2017). This general picture is also reflected by the results of this study. Especially, findings about As enrichment in ash basins of coal power plants are confirmed in Nalaikh, even though concentrations were lower than those detected at larger power plants as in Darkhan (N-Mongolia), where As concentrations of up to 1170 µg/L were measured (Pfeiffer et al. 2014). However, in Nalaikh, no indications of groundwater pollution by anthropogenic activity were detected (as compared to Inam et al. 2011; Pfeiffer et al. 2014; Batbayar et al. 2017). Bus Nuur serves as an example for salt ponds without outflow under current water level conditions. Its As concentration (SW-08) represents the highest measured in this study with an almost tenfold excess of the WHO guideline. Because of its accessibility, it constitutes the most significant threat to the health of livestock (and indirectly to meat consumers) in and around Nalaikh. Any regular consumption of this water should be avoided.

In Nalaikh, geogenic As input and urban coal combustion are the most influential factors on As redistribution. This represents a typical setting of Central Asian peri-urban environments (e.g., Shi et al. 2013). However, the Nalaikh case may be considered somewhat special, since widespread ger settlements emit concentrated As bound to fly ash as a diffuse source and at very low altitudes (ca. 2-3 m) to the environment. Arsenic emissions were often studied in association with fly ash emitted from coal power plants (Hart et al. 1995; Rausch et al. 1995; Gong et al. 2018), obviously magnitudes larger as single facilities and emitting at much higher altitudes (several tens of meters) than private stoves used in Nalaikh and all over Mongolia. Here, results indicate the potential influence of these diffuse, low-altitude sources on As emission. While the effect on As contents in soils is considered rather low, stagnant surface water bodies are specifically prone to As enrichment by evapoconcentration (Gao et al. 2007). Semiarid continental areas host numerous endorheic basins facilitating the process of evapoconcentration. Especially those with a comparably high proportion of animal husbandry and nomadism should consider the presented scenarios of As distribution and enrichment when evaluating potential threats to human and livestock health.

Conclusions

Three scenarios (MDS A to C) were developed according to regional framework conditions (climate, geological and soil conditions, anthropogenic influence) which were used as hypotheses for environmental distribution of As in the Nalaikh Depression. Setting up mobilization and distribution scenarios in order to narrow down explanations for As distribution revealed great potential for the analyses of different environmental compartments in this semiarid environment despite a limited number of samples. In the future, studies may further develop these scenarios according to various settings. The findings of this study are summarized as follows:

(a) The hazardous potential of As concentrations in soil and water of the Nalaikh Depression is largely negligible. However, local phenomena indicate As enrichment in sediments and soils as well as water, partly used to feed and water livestock. Soils and sediments show the highest enrichment near the coal power plant and in the immediate surrounding of coal ASM. Water samples mostly revealed uncritical values. However, water from Bus Nuur has the potential to reach > $100 \mu g/L$, exceeding WHO guidelines for drinking water by the tenfold.

(b) An immediate or even dominating impact of ASM coal mining activities on As concentrations in surrounding areas was not uncovered. Nevertheless, As emission into the land-scape is probably directly related to eolian transport of coal ash after combustion in the local power plant and growing *ger* settlement. Since As is specifically enriched in fly ash, this pathway is considered the most effective for As distribution.

(c) Investigating the mobilization and distribution scenarios of As in the Nalaikh Depression reveals a most likely combination of a geogenic input from As-rich lithologies in the source area of groundwater feeding the Nalaikh River, and the anthropogenically induced eolian redistribution, particularly, via coal ash. As such, the results presented here imply a scenario potentially valid in regions under heavy use of coal combustion. Particularly prone, beside Mongolia, are continental areas with long, cold winters and resulting heating demands (Central Asia, China, Siberia, Central US/Canada).

(d) There were no serious risks found on large spatial scales regarding As in the environmental compartments of the Nalaikh Depression. However, Bus Nuur as an important drinking water pond for local livestock experiences extraordinary high As concentrations ($\approx 100 \ \mu g/L$). Neither regular consumption by livestock nor by humans can, thus, be recommended.

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Compliance with ethical standards

Disclosure of potential conflicts of interest The authors declare that they have no conflict of interest.

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