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THEMATIC ISSUE

Investigating arsenic (As) occurrence and sources in ground, surface, waste and drinking water in northern Mongolia

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Abstract Elevated levels of arsenic in drinking water are found in several parts of Asia. Prolonged intakes of even low concentrations typically have serious health effects. This research paper integrates results of various studies on arsenic contamination of ground, surface, waste and drinking water in north-central Mongolia. Samples were analyzed with the ARSOlux biosensor and the Arsenator field test kit as well as different spectrometric methods (ICP-MS, ICP-OES). Altogether 309 samples were tested for their arsenic concentration, 44 of them with more than one technique. In the study region, the enrichment of heavy metals in surface waters is often linked to mining and coal combustion. The highest concentration of arsenic (As) was detected in the effluent of a gold mine (up to 2,820 μg L^{-1}) and in the ash basin of a thermal power plant (up to 1,170 μg L^{-1}). Five of 54 drinking water samples and 16 of 184 river samples were found to contain As levels above the World Health Organization (WHO) maximum permissible limit (10 μg L^{-1}), with a maximum of 300 μg L^{-1} As. In addition, elevated levels of uranium were detected. The degree and extent of As concentrations exceeding WHO standards were previously unknown and demonstrate the necessity for a more intensive screening as well as possible interventions concerning the intake of arsenic-contaminated drinking water. Preliminary results indicate that the ARSOlux biosensor technology is well suited for a precise quantification of arsenic content at low detection limits in regions where access to central laboratories is difficult.

Keywords Arsenic pollution · Drinking water · Gold mining · Mongolia · Upper Selenga River Basin

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Introduction

Mongolia is a landlocked country located in the heart of Asia between China and Russian Siberia. There are about 210 rivers flowing through Mongolia into Russia and China. Large rivers originate in the country's mountainous northern and western area while very few surface streams are found in the south. The upper basin of the Selenga River, which is the main artery feeding Lake Baikal in Russia, forms the study region of this investigation. Located in Mongolia, it encompasses several major rivers including the Orkhon, which has the Tuul and Kharaa as important tributaries. Since more than half of Mongolia's population and a considerable part of the country's mining and industrial activities are concentrated in the Tuul and Kharaa River Basins, they are of particular relevance in the context of

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water resources management in the transboundary Selenga River Basin (Karthe et al. 2013; Chalov et al. 2013).

While a comprehensive quality monitoring for ground, surface and drinking water in Mongolia is still in its infancy, elevated levels of arsenic have recently been documented in surface water, ground water, soils/sediments and urban vegetation for several locations in northern Mongolia. They appear to be mostly related to mining activities and the combustion of coal containing traces of arsenic (Hofmann et al. 2010; Kasimov et al. 2011a, b; Inam et al. 2011; Murao et al. 2011, Thorslund et al. 2012; Batbayar 2012). The Public Health Institute in Ulaanbaatar conducted extensive well water surveys and clinical examinations (Mongolian Ministry of Health 2004). In 7 of 21 aimags the mean arsenic concentration of the water samples exceeded the maximum tolerable level for drinking water of 10 μg L^{-1} (WHO 2011) and altogether 100,000 people are probably exposed to arsenic contamination in drinking water. Analyses of urine, hair and nails in a study group of 91 persons found evidence of arsenicosis in 16.5 % of the study group (Mongolian Ministry of Health 2004) and further studies from northern Mongolia also demonstrated high arsenic content up to 11 mg kg^{-1} in human hairs ($n = 21$) (Murao et al. 2004, 2011).

Three recent studies (Unurtsetseg et al. 2012; Olkhanud 2012; Nriagu et al. 2013) point to the fact that drinking water in many parts of the Mongolian Gobi provinces is contaminated with arsenic from natural and industrial origin. The Mongolian Ministry of Health selected 62 sums from five Gobi provinces as research sites. Elevated arsenic concentrations were present in 106 of 142 samples, existing arsenic concentrations in 15.4 % of the samples were 1–6 times higher than the drinking water standard of Mongolia (MNS 2005) and the WHO (2011) guideline for drinking water of 10 μg L^{-1} (Unurtsetseg et al. 2012). In Dornod Gobi Aimag 202 water samples were taken by an American-Mongolian research team. These samples ranged in arsenic content from 0.075 to 154 μg L^{-1}, with 20 % of wells exceeding the WHO guideline for arsenic in drinking water (Nriagu et al. 2013). In Southern Gobi region 237 water samples were taken to explore water resources near Oyu Tolgoi mine, where 33 % (78) of all samples showed concentrations higher than 10 μg L^{-1} arsenic and 3 % of wells had concentration higher than 50 μg L^{-1}, ranging up to a maximum of 159 μg L^{-1} (Olkhanud 2012).

For the Kharaa River Basin in northern Mongolia, where a comprehensive survey on the state of water resources was carried out (MoMo Consortium 2009; Karthe et al. 2014; Hofmann et al. 2014), heavy-metal concentrations showed an enrichment as compared to natural background levels although they were usually below or near maximum permissible limits. A first survey including arsenic (Hofmann et al. 2010) found surface water concentrations mostly between 1 and 10 μg L^{-1} while reaching up to 31 μg L^{-1}. A highly elevated level (up to 1,170 μg L^{-1} As) was detected in the ash basins of the thermal power plant in Darkhan. The concentration in nearby drainage trenches was about 78 μg L^{-1} As. This suggests that the combustion of coal is one localized source of arsenic in water bodies (Hofmann et al. 2010) as well as in soils (Kasimov et al. 2011a). In central Mongolia including the capital Ulaanbaatar, the main sources of coal are the deposits in Baganuur, Nalaikh and Chulut, all of which have elevated arsenic contents (Kasimov et al. 2011b). Arsenic concentrations in coal from Baganuur and Nalaikh typically exceed 100 mg kg^{-1}, a level at which toxicity of combustion byproducts is considered to be of serious environmental and human health concern (Mongolian Ministry of Health 2004). Moreover, elevated levels of arsenic have been detected in plant material from Ulaanbaatar, possibly derived from air pollution (Kasimov et al. 2011b).

Gold mining and processing are also known to enhance the release of arsenic and its uptake by humans and livestock (Keshavarzi et al. 2012). Recently, gold mining has emerged as one of the most dynamic sectors of the Mongolian economy. Most gold mines are concentrated in northern Mongolia with a high environmental impact on local rivers, which all drain into the Selenga River: the Zaamar goldfield is located in the Tuul River Basin and two large open pit gold mines are situated at Boroo and Gatsuurt Rivers, respectively. The placer gold mining at the Zaamar site has been estimated to increase total arsenic load of Tuul River by 30 tons $year^{-1}$ (Thorslund et al. 2012). An arsenic content of 46,986 mg kg^{-1} was determined in rocks collected from Gatsuurt gold mine (Tsetsegmaa et al. 2009) and arsenic concentrations in artificial ponds of that mine have been measured to be 121 μg L^{-1} (Gandoljin et al. 2010). The average arsenic content in the tailing dam sediment of Boroo gold mine was determined at 4,419 mg kg^{-1}, thus posing a potential source for future environmental contamination (Inam et al. 2011).

This paper summarizes the results of extensive testing for arsenic, which has been conducted in northern Mongolia with different methods and by various teams between May 2007 and 2013 and included ground, surface, waste and drinking water sources. We assess the existing contamination and identify potential sources of arsenic contamination which may have negative impact on the water quality of this area in the future.

Materials and methods

Study region

The study region comprises the Mongolian part of the Selenga River Basin with a particular focus on the Kharaa,

Tuul and Orkhon subbasins, which are comparable with regard to the physical environment and socio-economic development (Karthe et al. 2013). A highly continental climate with very cold winters and short but warm summers is characteristic of this landlocked Central Asian region. Water availability is naturally limited due to low precipitation and high evaporation rates. Even though only 20 % of the annual precipitation falls during the winter months, and sublimation losses are above 80 %, the melting of snow and river icings produce a first considerable peak in river discharge around May (Minderlein and Menzel 2014). Because of a concentration of rainfall during the summer months, more than half of the annual runoff occurs during the months of July, August and September, albeit with a large interannual variability (Batimaa et al. 2005; Berezhnykh et al. 2012; Hülsmann et al. 2014). While open grasslands dominate low-lying regions, mountainous regions (particularly in the rivers' headwater areas) are typically forested and play a key role in runoff formation (Menzel et al. 2011). The Tuul, Kharaa and Orkhon River Basins are home to Mongolia's three largest cities (Ulaanbaatar, Darkhan and Erdenet, respectively) and to more than half of the country's population. Moreover, the three river basins constitute important centers of agriculture, industry and mining (in particular for gold and copper). This does not only lead to a concentration of consumption but also to contamination risks. At the same time, water pollution in this region may harm a relatively large exposed population (Chalov et al. 2013).

Sampling procedure

Shallow and deep groundwater wells, lakes, rivers and artificial ponds, as well as wastewaters from mining and industry, were sampled in the northern part of Mongolia between May 2007 and May 2013 in the context of different monitoring projects and expeditions. A total of 309 water samples were collected for chemical analysis in Mongolia and Germany.

Routinely, water samples were taken with a 10 L bucket from water sources. At wells water was pumped for 2 min and discarded before collecting a sample. On-site determinations of water quality included measurements of temperature and pH, total dissolved solids (TDS), electrical conductivity (EC), and dissolved oxygen (DO) by a multi parameter tester (WTW, Multi 3430 SET G, Weilheim, Germany). Water samples for chemical analyses were collected in 50 mL vials and were acidified in the field with 10 mM H_3PO_4 for stabilization (Daus et al. 2006). A part of these samples were filtered with a 0.45 µm cellulose acetate filter (see Table 1 for different methods). Whenever arsenic was to be determined using the Arsenator field test kit, samples were not filtered. In the field samples were stored in a cooler box at about 10 °C before laboratory analysis at the Central Geologic Laboratory of Mongolia or shipment to Germany.

Table 1 Description of testing methods

Method	Description
A	Unfiltered samples, stabilized by 10 mM H_3PO_4, tested by ICP-MS at UFZ Magdeburg
B	Unfiltered samples, stabilized by 1 % HNO_3, tested by ICP-MS at FUGRO CONSULT GmbH Berlin
C	Unfiltered samples, stabilized by 1 % HNO_3, tested by ICP-MS at KIWA Control GmbH Berlin
D	Unfiltered samples, stabilized by 10 mM H_3PO_4, tested by ICP-MS at Central Geological Laboratory, Ulaanbaatar
E	Filtered samples, stabilized by 10 mM H_3PO_4, tested by HPLC, ICP-MS and ICP-OES, at UFZ Leipzig
F	Filtered samples, stabilized by 10 mM H_3PO_4, tested by ICP-MS at Central Geological Laboratory, Ulaanbaatar
G	Filtered samples, tested by the ARSOlux biosensor test kit at the National University of Mongolia
H	Unfiltered samples, tested by the Arsenator test kit on site

Chemical analyses

This paper integrates the findings of several studies which were carried out independently from each other and, therefore, used different methods for assessing arsenic concentrations. A short description of the testing methods is found in Table 1. Even though different testing methods imply some limitations in comparability, the added value of this data compilation lies in providing the currently most comprehensive picture of arsenic occurrence in water for north-central Mongolia. Besides different certified laboratory methods such as ICP-MS (method A–F, Table 1), we also used the two field test kits, namely the ARSOlux biosensor (UFZ, Leipzig, Germany) and Arsenator (Wagtech, Palintest, London, UK).

The ARSOlux biosensor field kit (method G, Table 1) contained lyophilized (freeze–dried) bioreporter bacteria (Siegfried et al. 2012). Before the measurement of a sample lot the Junior 9509 luminometer (Berthold Technologies, Bad Wildbad, Germany) device of the kit was calibrated individually with standards of known concentrations of arsenite prepared by dilution of a 1,000 µg L^{-1} $NaAsO_2$ stock solution in demineralized water. Calibration series included four concentrations ranging from 5 to 200 µg L^{-1} arsenite as $NaAsO_2$ (Fig. 7a). A 1-mL portion of arsenite standard or water sample was filled into a plastic syringe and injected into a bioreporter vial by penetrating the stopper. Three replicate vials were filled this way. The vials were shaken five times by hand and kept at a temperature of 30 °C in an incubator. Water samples were occasionally

tenfold diluted prior to incubation to identify arsenite toxicity on the bioreporter cells, which would result in false-negative low bioluminescence. After exactly 2 h incubation, the vials were inserted into the battery-driven luminometer to measure integrated bioluminescence over a 10 s interval. Arsenic concentrations in groundwater were inferred by comparison of luminescence values with those in the calibration series using an automated logarithmic regression, and are thus expressed as arsenite equivalent concentration. Bioreporter bacteria were killed by application of a 6 % H_2O_2 solution followed by autoclaving of used vials and syringes.

The Arsenator test kit (method H, Table 1, Wagtech, Palintest, London, UK) detects total arsenic concentration in water samples by the well-established Gutzeit method. The speciations As (III) and As (V) are both chemically transformed into arsine gas. Upon contact of the gas with a reagent on a test stripe, colored mixed arsenic/mercury halide compounds are formed. The intensity of a yellow to brown colored spot on the stripe is compared to a semi-quantitative color scale. For a more accurate differentiation of very light yellowish signals induced by arsenic concentration lower than 100 μg L^{-1}, the Arsenator test kit also includes a small portable photometer. The test was conducted according to the instruction manual. The reliability of the field kit results of total arsenic was tested by comparison with spectrometric methods. Inductively coupled plasma optical emission spectrometry (ICP-OES; ARCOS, Spectro A.I.), and inductively coupled plasma quadrupole mass spectrometry (ICPqMS; ELAN DRC-e, Perkin-Elmer) were applied for concentrations of arsenic above and below 100 μg L^{-1}, respectively. Total concentrations of chromium, copper, iron, manganese, antimony and uranium were measured with ICPqMS, while sodium, potassium and chloride concentrations were detected semi-quantitatively with ICP-OES. By coupling high-performance liquid chromatography (HPLC) online with ICPqMS, the arsenic species As (III) and As (V) could be differentiated (Mattusch et al. 2000).

Data analysis

Data were compiled in an Excel data sheet and analyzed with STATISTICA 7.1. Nonparametric Whitney–Mann *U* tests and Spearman Rank Correlations were performed on the data. Cross-comparison of different analytical methods for arsenic detection was performed by linear regression using the software Microsoft Excel 2010. Results were compared with the Mongolian National Standard (MNS 2005) and the WHO (2011) guidelines for drinking water quality.

Results

Overview

From 2008 to 2013, a total of 309 water samples were collected from 130 sample points in northern Mongolia during routine monitoring and specific expeditions (Fig. 1, ESM Appendix 1). The results could be categorized into 14 sample types, including different kind of surface and ground waters, drinking water for humans and livestock, various types of waste water and other sorts of technically used water. Eight major types of samples that comprised 297 water tests are shortly described as follows (Fig. 2): rivers ($n = 184$) were most often sampled and showed relatively low median concentration of arsenic of 2.5 μg L^{-1} (range 0.7–190 μg L^{-1}), salt lakes were the second natural water source investigated, but only sampled twice with median arsenic concentrations of 19 μg L^{-1} (range 18.7–19.0 μg L^{-1}). Drinking water wells ($n = 54$) had a median concentration of 2.5 μg L^{-1} (range 0.9–300.0 μg L^{-1}), while herders' wells ($n = 6$) showed a median concentration of 2.7 μg L^{-1} (range 0.05–330.0 μg L^{-1}) of the metalloid (Fig. 2). Effluents from waste water treatment plants ($n = 21$) had even lower arsenic concentrations averaging 2 μg L^{-1} (0.9–3.2 μgL^{-1}). The highest concentration of arsenic was measured in two types of artificial ponds: settling ponds of coal power plants, which serve as deposit for ashes (median 372 μg L^{-1}, range 1.9–1,170.0 μg L^{-1}, $n = 19$), and mine waste water ponds (median 105 μg L^{-1}, range 5.4–221.0 μg L^{-1}, $n = 7$). However, while settling ponds of coal power plants work with a closed water circulation, ponds for mining operation are often drained into natural river systems. Particularly, gold mining effluents in some cases carry high loads of arsenic in median 288 μg L^{-1} (49.0–2,820.0 μg L^{-1}, $n = 4$).

Rivers

Although the arsenic concentration in northern Mongolian rivers was mainly low, it exceeded the 10 μg L^{-1} WHO threshold for drinking water in 16 cases, which made 8.7 % of all measurements, in Bayangol, Boroo, Gatsuurt, Kharaa and Orkhon River (Figs. 3, 4). The maximum concentration of arsenic was measured in an Orkhon River sample with 190 μg L^{-1} (Fig. 4a). In Gatsuurt River 9 of 10 measurements were above this threshold (Figs. 2, 4d). A detailed presentation of data from sites along the rivers clearly demonstrates a fluctuation of arsenic content downstream (Fig. 4), which may be caused by spatial–temporal variation of arsenic input and/or sedimentation and binding and subsequent leaching of arsenic in river

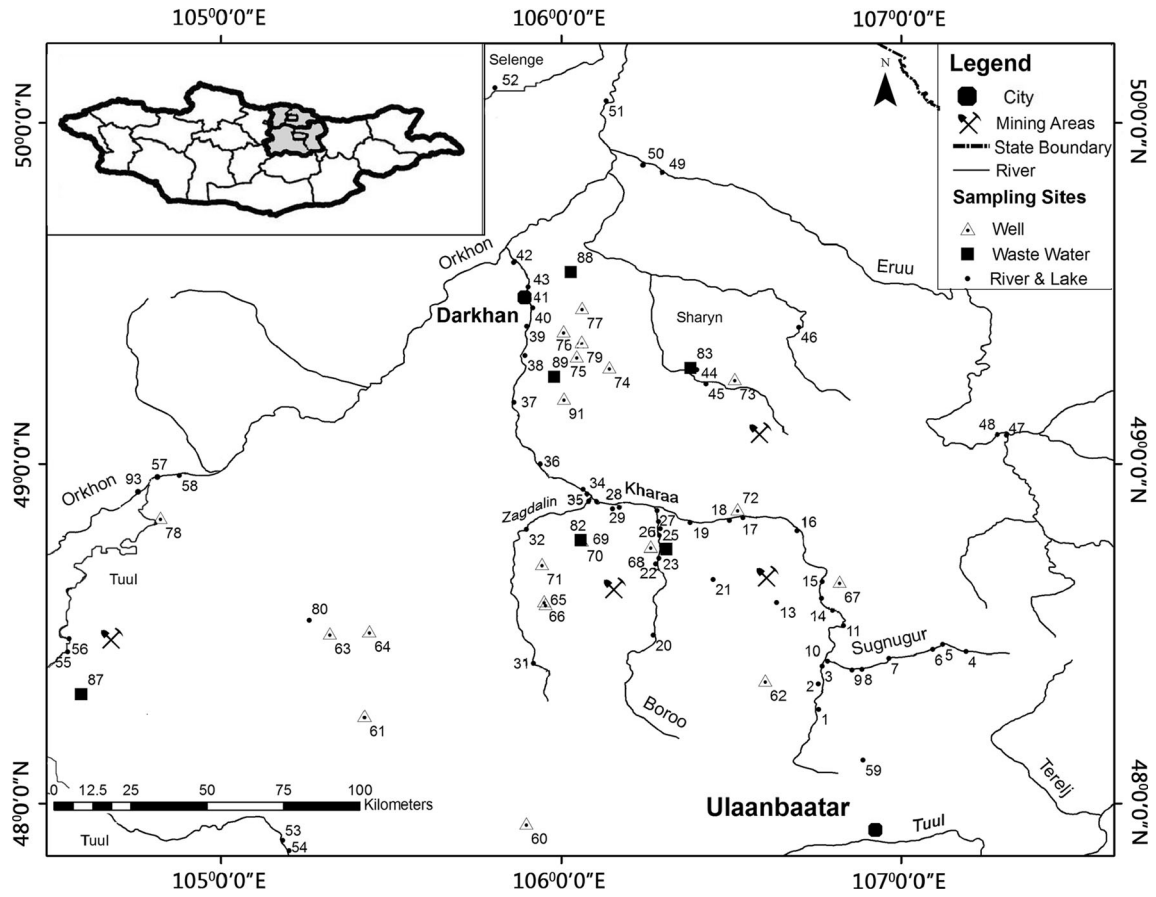

Fig. 1 Map showing the geographic location of the sample sites of this study in northern Mongolia. Sample sites comprise ground, surface and drinking water resources in the Selenge, Tuv, Darkhan Uul and Bulgan Aimags (provinces) that are *shaded* in the *inserted map* of whole Mongolia

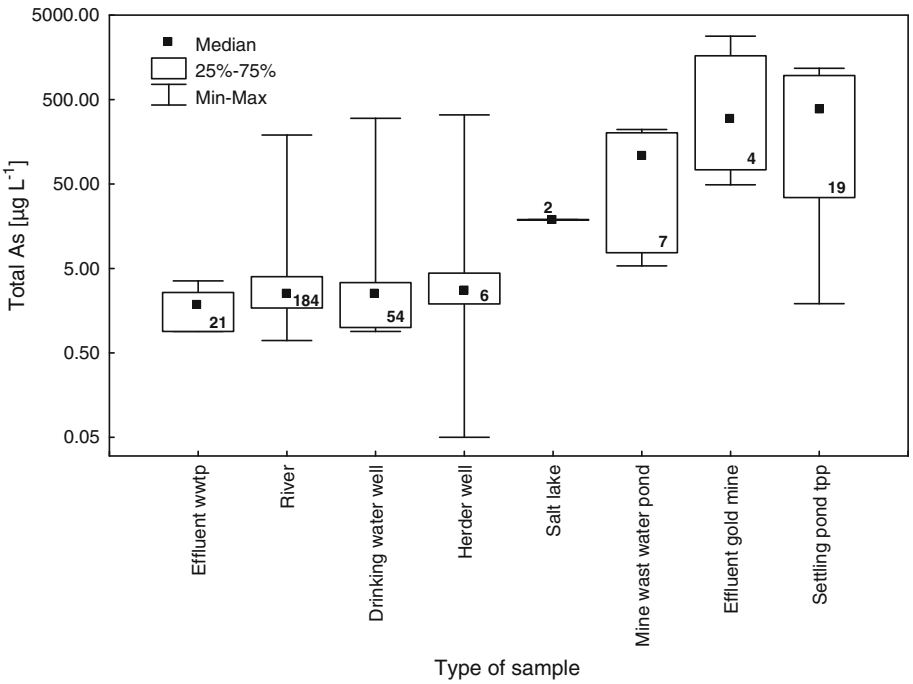

Fig. 2 Sample types for 297 water samples taken in the course of this study. The figure shows the eight major groups of samples. *Numbers* refer to the *n* of the sample type. Mind the logarithmic scaling of the *Y*-axis. *wwtp* wastewater treatment plant, *tpp* thermal power plant

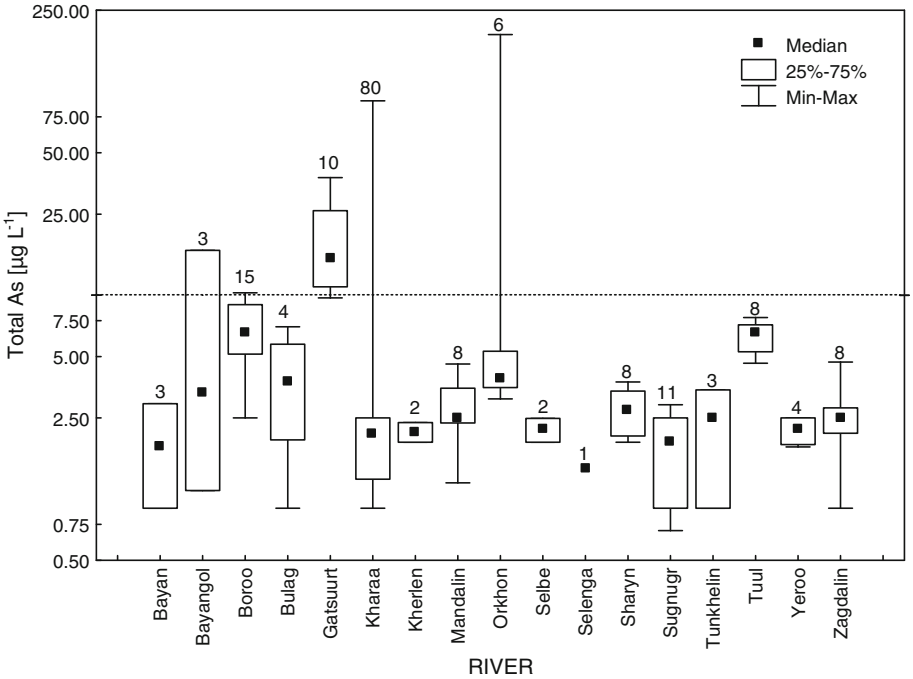

Fig. 3 Arsenic content of rivers in northern Mongolia during our study from May 2007 to May 2013. Mind the logarithmic scaling of the *Y*-axis. The *dotted line* gives the WHO guideline value for drinking water of 10 µg L^{-1}. *Numbers* show the number of samples (*n*) per river. Maximum *values above* the *threshold* were measured for Bayangol (16.6 µg L^{-1}), Boroo (10.2 µg L^{-1}), Gatsuurt (37.8 µg L^{-1}), Kharaa (90 µg L^{-1}) and Orkhon River (190 µg L^{-1})

sediments. Moreover, our data exhibit discharge of arsenic at certain river sections, viz. peaks of arsenic concentration were found downstream river junctions of Orkhon–Tuul (Fig. 4a) and Boroo–Kharaa (Fig. 4b). For Boroo River, our data shows fluctuations of arsenic up to 400 % of the measurement upstream the river, with an increase in arsenic concentration downstream (Fig. 4c), while in Orkhon (Fig. 4a) and in Gatsuurt River (Fig. 4d), a continuous dilution of arsenic concentration downstream mining area was observed.

Wells

Arsenic concentration higher than 10 µg L^{-1} was measured in 9.3 % of all 54 samples, so in five cases altogether (Fig. 5). These extreme values showed concentrations up to 94 and 300 µg L^{-1} for Zuunkhaara deep well and Borewell 3, respectively. In six samples from herder wells, we found one outlier of 330 µg L^{-1} arsenic for a well, otherwise measurements were below 5 µg L^{-1}.

Artificial ponds for waste and processing water

The highest concentrations of arsenic were found in artificial ponds for waste and processing water of gold mining operations and coal-fired power plants (Fig. 2). Arsenic concentration ranked up to 221 µg L^{-1} in mining waste water ponds and was especially high in mining effluents with a maximum of 2,820 µg L^{-1} arsenic found in the mining effluent of Bor Tolgoi gold mine, exceeding the Mongolian standard for waste water (MNS 2011) 282 times. These high concentrations demonstrated that gold mining may dramatically influence arsenic concentration in surface waters. In Kharaa River, arsenic concentration peaked at Kharaa–Boroo junction (Fig. 4b), presumably influenced by mining effluence from gold mining operations in the upstream catchment area of the Boroo River. At the Boroo River we found effluent of a small-scale mining operation where water from a waste water pond drained directly into a small creek and subsequently into the Boroo River with gradually diminished arsenic concentrations (mean As concentration of 73 µg L^{-1}) (Fig. 6).

A maximum arsenic concentration of 1,170 µg L^{-1} was found in the settlement pond of the Darkhan coal power plant. A high mean content of arsenic of about 450 µg L^{-1} suggest a considerable impact on the environment, although measurements were highly variable over time. While samples from autumn ($n = 7$) showed a very high arsenic concentration (median 1,050 µg L^{-1}), samples that were taken in spring ($n = 9$) had a significantly lower arsenic concentration (median 59.37 µg L^{-1}, Mann–Whitney *U* test $U = 5.00$ $Z = 2.81$, $p = 0.005$).

Correlation of the arsenic concentration and environmental data

Arsenic content of water samples was highly significantly ($p < 0.001$) positively correlated (Spearman rank correlation, SRC) with water temperature ($R = 0.32$), pH value ($R = 0.25$) and total dissolved solids ($R = 0.26$) (Table 2). Moreover, arsenic concentration in water samples was found to be highly significantly correlated (SRC,

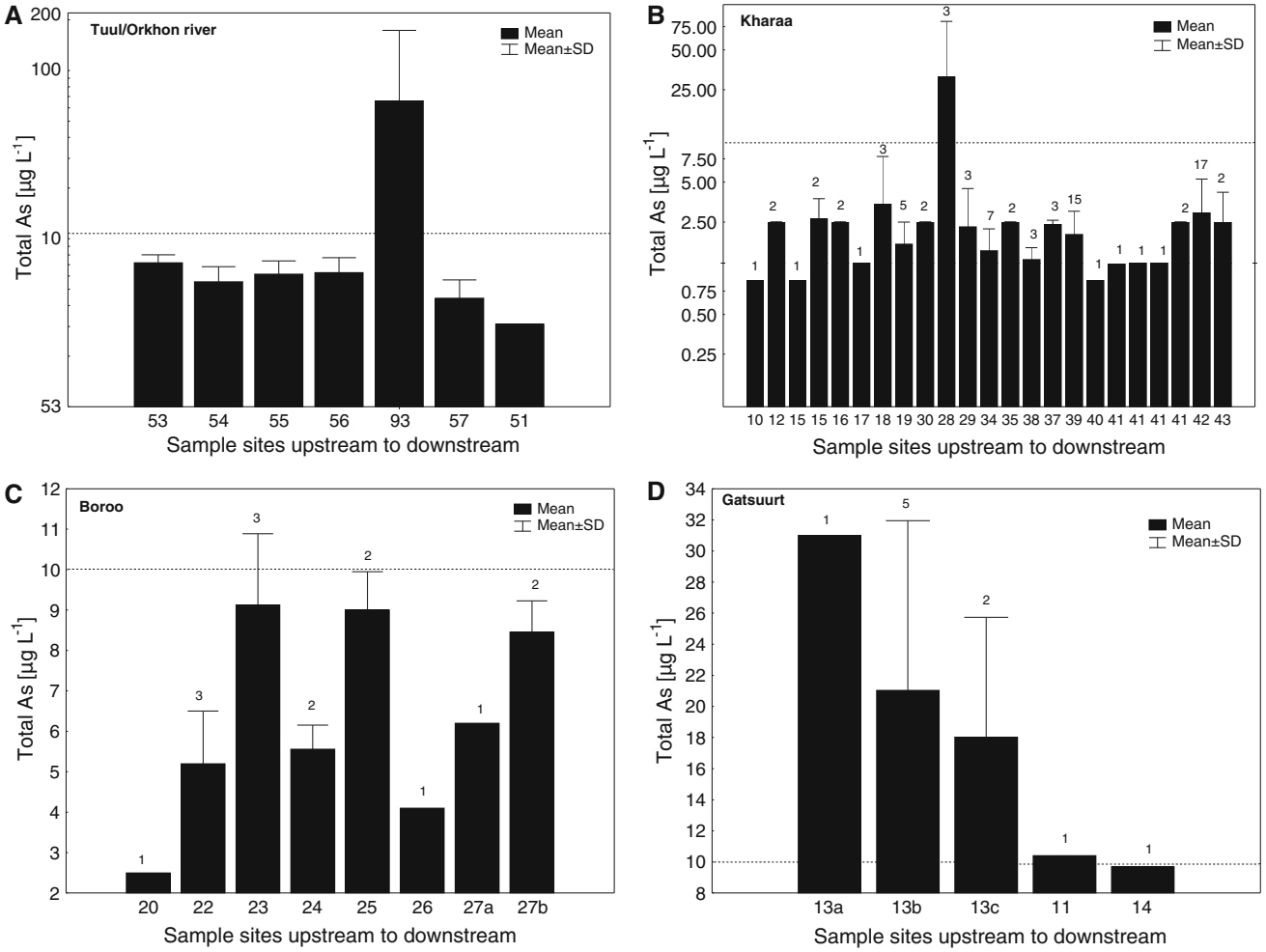

Fig. 4 Changes in arsenic concentration along rivers. Shown is the distribution of arsenic concentration at samples size ordered from upstream (*left*) to downstream (*right*). Mind the different scaling and *start* value of the *Y*-axis (**a**, **b** with logarithmic scaling). The *broken lines* mark the WHO guideline value for drinking water of 10 µg L^{-1}. Sample sites are given according to Fig. 1, with each column representing a distinct samples site, while numbers are adjusted to optimal presentation in the map. **a** Tuul/Orkhon River. Site 53 to 56 Tuul River (Lun to downstream Zamar mines), Site 93 Orkhon River downstream Erdenet, Site 57 downstream Tool–Orkhon junction. **b** Kharaa River. This sample site was investigated most thoroughly. Site 28 is downstream the Boroo–Kharaa River junction, with higher arsenic load coming from the mining areas at Boroo River. **c** Boroo River. Samples were taken upstream, midstream and downstream of the Boroo Gold mining area, with site 25 downstream an influx channel for mining effluents. **d** Arsenic content in different distance from Gatsuurt Gold Mine. Site 13a is taken immediately near the mining site; downstream sample sites are further remote from that area

$p < 0.001$) with concentration of sodium ($R = 0.57$), chloride ($R = 0.49$) and uranium ($R = 0.41$). More results are given in Table 3.

Laboratory data from ICP-MS and ICP-OES

To check the accuracy of arsenic detection methods, concentration data in field samples collected in spring 2013 ($n = 44$) were determined with three different spectrometric methods (A, E, F) in the Central Geological Laboratory in Ulaanbaatar, Mongolia (ICP-MS) and the laboratory of the Department Analytical Chemistry of the UFZ in Leipzig, Germany (HPLC-ICPqMS, ICP-OES). The results were in good agreement, but slightly lower arsenic concentration levels were detected for some specific samples by HPLC-ICPqMS and ICP-OES in Germany (Fig. 7b) compared to the data determined in Mongolia.

Performance of the ARSOlux Biosensor and Arsenator field test kits

The results for arsenic concentration detected by the ARSOlux biosensor and Arsenator field test kits were in good quantitative agreement with the results of laboratory measurements (Fig. 7c; Table 4). The concentrations

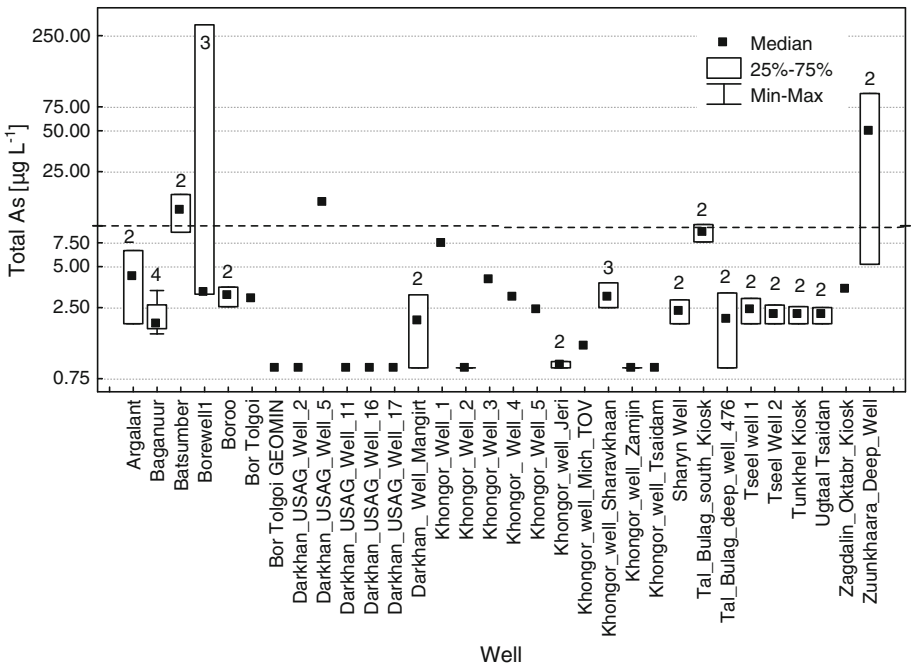

Fig. 5 Arsenic concentration in 32 drinking water wells in our survey area. The numbers refer to the *number of measurements* (*n*). Maximum *values above the threshold* were measured for Batsumber (17.0 µg L^{-1}), Borwell 3 (300.0 µg L^{-1}); Darkhan USAG Well 5 (14.8 µg L^{-1}), Tal Bulag South Kiosk (10.2 µg L^{-1}) and Zuunkhaara deep well (93.9 µg L^{-1}). The *broken line* gives the WHO guideline value for drinking water of 10 µg L^{-1}. Mind the logarithmic scaling of the *Y*-axis

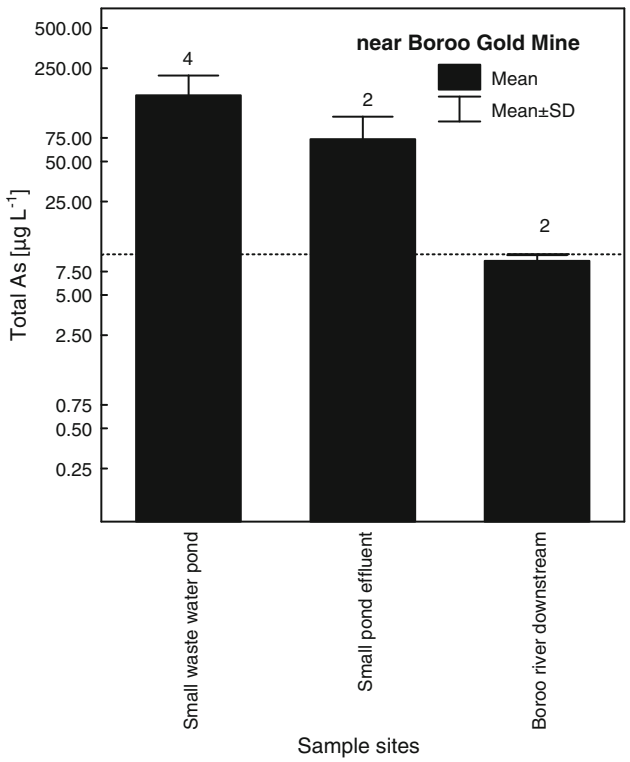

Fig. 6 Arsenic concentration of samples collected at surface water ponds ca. 1 km beneath the tailing dam of Boroo gold mine in comparison with river water of Boroo Gol

detected in unfiltered samples by the Arsenator were slightly higher than the data measured by ARSOlux in filtered samples.

Discussion

Our research comprises samples that were taken over a time span of 5 years in northern Mongolia. While most other comprehensive studies from Mongolia focused on monitoring of ground and drinking water (Mongolian Ministry of Health 2004; Hofmann et al. 2010; Batbayar 2012; Olkhanud 2012; Unurtsetseg et al. 2012; Nriagu et al. 2013) our investigation also includes an assessment of river and process water from industrial and mining activities to check for the reasons of arsenic pollution. The aggregated results draw the most comprehensive picture of arsenic contamination in north-central Mongolia available to this date.

Impact of gold mining on arsenic load of rivers

Gold mining has been spotted as a frequent source for high arsenic loads in many regions of the World (Ravenscroft et al. 2009; Keshavarzi et al. 2012), and our results corroborate these findings. Besides gold the bedrock contains high concentrations of arsenic (Tsetsegmaa et al. 2009), which is solved during the gold washing and extracting process. At Gatsuurt gold mine Gandoljin et al. (2010) measured mining effluents with a concentration of 121 µg L^{-1} arsenic, while Enkhdul et al. (2010) reported 136 mg kg^{-1} arsenic for the sediment of Gatsuurt mid river. The present study revealed high arsenic concentrations in artificial ponds for mining processing water and in mining effluent, e.g., in the case of the artisanal small-scale mining site Bor Tolgoi (ca. 9 km WNW of Boroo gold

Table 2 Spearman rank-order correlation of environmental descriptors with arsenic content of water samples ($n = 309$, ICP-MS methods A–E)

	Water temp. (°C)	pH	EC (μs cm^{-1})/TDS (mg L^{-1})	Oxygen (μg L^{-1})	Depth of well (m)	Total As (μg L^{-1})
Water temp. (246)	1.00	**0.46*****	**−0.14**	−0.12	0.31	**0.32*****
pH (207)	**0.46*****	1.00	0.13	**0.25*****	**0.61****	**0.25*****
EC/TDS (282)	**−0.13**	0.13	1.00	−0.04	**−0.50**	**0.26*****
Oxygen (229)	−0.12	**0.25*****	−0.04	1.00	0.36	0.11
Depth of well (24)	0.31	**0.61****	**−0.50**	0.36	1.00	−0.23

Given are coefficients of Spearman rank-order correlations for the factors water temperature, pH, electric conductivity/total dissolved solids, oxygen and the depth of the well in case of ground water samples

Total n for descriptors are given in brackets in the first column, units are given in the first row and for calculation missing data were pairwise deleted. see ESM Appendix 1 for the data

Bold correlations are significant at $p < 0.05$

Higher significances are marked as ** $p < 0.01$, *** $p < 0.001$

Table 3 Spearman rank-order correlation of the concentration of different elements with the arsenic content of water samples ($n = 43$)

	pH	EC (μs cm^{-1})/TDS (mg L^{-1})	As (μg L^{-1})	As (III) (μg L^{-1})	As (V) (μg L^{-1})	U (μg L^{-1})
Cr (μg L^{-1})	0.25	0.02	0.12	−0.18	0.29	**0.49*****
Cu (μg L^{-1})	0.01	0.06	0.24	−0.06	0.24	0.24
Fe (μg L^{-1})	−0.08	−0.20	0.01	−0.04	−0.14	−0.08
Mn (μg L^{-1})	−0.07	0.23	**0.32***	0.29	0.07	−0.02
Sb (μg L^{-1})	0.03	0.13	**0.38***	0.14	**0.35***	0.14
U (μg L^{-1})	**0.37***	**0.57*****	**0.41*****	0.22	**0.51*****	1.00
K (mg L^{-1})	**0.36***	**0.57*****	**0.57*****	**0.44****	**0.58*****	**0.63*****
Na (mg L^{-1})	0.08	**0.92*****	**0.38***	0.26	**0.39***	**0.57*****
Cl (mg L^{-1})	0.24	**0.79*****	**0.49*****	**0.35***	**0.53*****	**0.68*****

Given are coefficients of Spearman rank-order correlations for the concentrations of chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), antimony (Sb), uranium (U), sodium (K), potassium (Na) and chloride (Cl) with pH and electric conductivity/total dissolved solids of the samples, as well as with concentrations of total arsenic (As), the arsenic species As (III) and As (V) and—for comparison—with uranium (U). All samples were measured by method E only (see Table 1)

Bold correlations are significant at $p < 0.05$

Higher significances are marked as ** $p < 0.01$, *** $p < 0.001$

Data are given in ESM Appendix 2

mine) direct measurement of the arsenic concentrations in mining waste water ponds and the effluent discharge revealed values of up to 2,820 μg L^{-1} that will contaminate the upper groundwater layer.

In contrast to small-scale mining the big facilities have different operation procedures. Thus, the Boroo gold mine waste water pond has been designed as a zero discharge facility; that is, supernatant water from the dam is stored in a tailing reservoir and will not be discharged to the environment. However, high concentrations of arsenic in the tailing dam sediment of Boroo gold mine were already measured by Inam et al. (2011), who also tracked underground flows of heavy metals (including As) from the dam to monitoring wells situated downhill. Mining waste water in that study showed arsenic concentrations of 1,746 μg L^{-1}, while a maximum of 46 μg L^{-1} was measured at the monitoring wells that all were above the Mongolia water quality standard for As. The highly fluctuating arsenic concentrations in Boroo River water presented here (Fig. 4c) may have been influenced by underground flows from mining areas. The enrichment of arsenic in tailing deposits, contaminated leachate and its accessibility by livestock, birds and other wildlife is a major point of concern. Moreover, in case of dam destruction and release of tailing deposits and water of the tailing reservoir itself a serious impact to the environment may occur.

As a consequence of the distribution of pollution sources arsenic concentration varied strongly along rivers (Fig. 4) with an overall trend of reduced concentration downstream, where larger water volumes led to dilution of the arsenic concentration, which is a common effect in large river

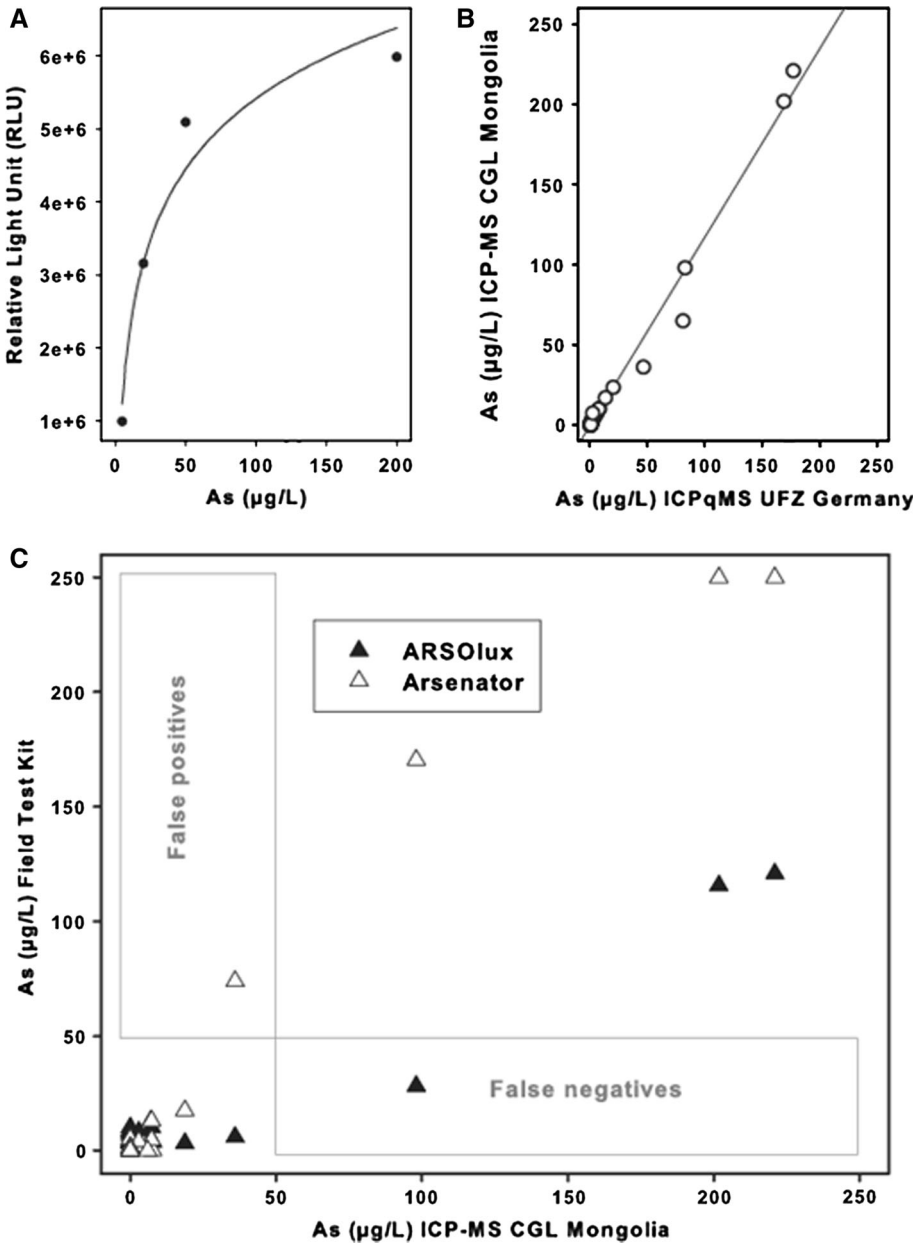

Fig. 7 Calibration curve of the ARSOlux biosensor (**a**), cross-analysis of ICPqMS data from UFZ Germany and ICP-MS results of the Central Geological Laboratory in Ulaanbaatar, Mongolia ($n = 44$, **b**). Cross-comparison of arsenic results measured by ICP-MS in the Central Geological Laboratory in Ulaanbaatar, Mongolia and data measured with the arsenic field test kits ARSOlux biosensor and Arsenator ($n = 22$, **c**)

Table 4 Arsenic concentrations detected with different analytical methods (total $n = 42$)

Method	ARSOlux [G]	ICP-MS (Mongolia) [F]	ICPqMS (Germany) [E]	HPLC-ICPqMS (Germany) [E]	
Unit	Total As, µg L^{-1}	Total As, µg L^{-1}	Total As, µg L^{-1}	As (III), µg L^{-1}	As (V), µg L^{-1}
Rivers ($n = 21$)	6 (3–12)	4 (0–23)	4 (1–21)	0	5 (2–13)
Drinking water wells ($n = 12$)	7 (3–26)	2 (0–17)	2 (0–14)	0 (0–5)	3 (2–7)
Mining effluent ($n = 4$)	67 (4–121)	132 (8–221)	108 (5–177)	0	64 (5–104)
Industrial and municipal waste ($n = 5$)	16 (6–10)	23 (3–65)	21 (2–81)	0	14 (2–59)
All ($n = 42$)	13 (3–121)	18 (0–221)	16 (0–177)	0 (0–5)	12 (2–104)

Given are averages with minimum and maximum values. Letters in square brackets refer to methods described in Table 1

systems (Mueller et al. 2008). In terms of mass flow contributions the river loads of arsenic have been investigated by Thorslund et al. (2012) for the Tuul River as the most polluted river in Mongolia. During the period 2005–2008, the average net increase for dissolved mass flows downstream of Zamaar gold field was 9 tons year^{-1} arsenic and reached about 30 tons year^{-1} in 2008. Therefore, this mining area has been identified as a major contributor of heavy-metal and arsenic influxes into the Selenga River system (Chalov et al. 2012; Thorslund et al. 2012). Interestingly, perhaps due to seasonal effects, we measured only low arsenic concentration in Tuul River downstream the Zaamar goldfield in May 2013. Recent investigations by Hofmann et al. (2013) also state increasing arsenic river loads at the outlet of Kharaa River basin, reaching 1.7 tons year^{-1} arsenic in 2011 and 3.3 tons year^{-1} in 2012. Arsenic concentration in Tuul River near Ulaanbaatar originates from natural and anthropogenic resources and was found strong enough to cause adverse aquatic biological effects (Dalai and Ishiga 2013). In contrast our measurements in Tuul River at Lun Bridge more than 200 km downstream revealed low arsenic concentrations, thus pointing towards spatial variation of arsenic load, possibly caused by binding of arsenic to the river sediment. These contrasting results demonstrate a lack of knowledge regarding the seasonal impact of mining areas on arsenic loads of rivers in steppe regions and identify some issues for further research in Mongolia: (1) identification of major dischargers and of probable occurrence areas of heavy metals (see also Rodríguez-Lado et al. 2013), (2) monitoring seasonal and spatial effects on arsenic load in rivers and sediments, (3) modeling discharge and sedimentation of arsenic along the rivers and within the course of the years (see also Brumbaugh et al. 2013; Chalov et al. 2014). Since groundwater recharge is mainly fed by bank infiltration from rivers and most of the drinking water extraction sites of the cities of Darkhan and Ulaanbaatar are situated in the river floodplains, the groundwater quality is already affected by increasing levels of arsenic and other heavy metals (see Dalai and Ishiga 2013).

Threats for the ground water

Still samples of well water in northern Mongolia ranked mostly well below the WHO maximum permissible limit for arsenic of 10 μg L^{-1}; however, 10 % of the wells exceeded that limit with a maximum load of arsenic of 330 μg L^{-1} in herder wells and 300 μg L^{-1} for drinking water wells, five of which surpassed WHO level. It can be summarized that drinking water seems less affected by arsenic pollution than in other parts of the country, viz. in Dornogobi Aimag, where 20 % of the wells were contaminated (Nriagu et al. 2013), or in Southern Gobi with 16.4 % of the boreholes above the recommendation threshold (Olkhanud 2012). In 2004 nationwide 10.3 % of 867 water samples contained arsenic with an average arsenic content of 14 ± 3 μg L^{-1} (Mongolian Ministry of Health 2004); however, due to intensified mining activities all over Mongolia, arsenic pollution may have increased since that time.

While the Mongolian Ministry of Health' survey accounts for coal and mineral deposits in the vicinity of the wells as a latent cause of danger (Mongolian Ministry of Health 2004), our study points towards gold mining sludge, ash deposits and settling ponds of power plants as further potential pollution sources. In fact up to 1,170 μg L^{-1} of arsenic was detected in the settling pond of Darkhan coal power station, and the total of 19 samples for all settling ponds showed a median arsenic concentration of 372 μg L^{-1}. These high loads of arsenic in the processing water pose a serious threat to the surrounding ground water and the population that uses it. Darkhan USAG Well 5 is in only 3.8 km distance from the power station and had an elevated arsenic concentration (14.8 μg L^{-1}), indicating a potential pollution by underground flow of contaminated process water (see also Inam et al. 2011, as discussed above). Seasonal changes of weather conditions may impact the arsenic concentration of the source pool, e.g., by higher dilution of processing water because of pronounced precipitation in spring and early summer, or increasing concentration in autumn after summer drought periods, as indicated by our results and in the literature (Bhattacharya et al. 2011; Aguilar-Muniz et al. 2013).

Interestingly, high arsenic concentration was positively correlated with high concentration of uranium in 43 of our samples. This points towards another element that is frequently detected in Mongolian water samples and poses an additional danger to human health: 11 of 33 samples taken from rivers or drinking water wells had uranium concentrations above the provisional WHO drinking water guideline of 15 μg L^{-1}, with Ugtaal tsaidan and Tseel No. 2 drinking water wells both at levels ≥60 μg L^{-1}. These results corroborate findings from Ulaanbaatar (Nriagu et al. 2012), Dornogobi Aimag (Nriagu et al. 2013) and eastern Mongolia (Linhoff et al. 2011, lake water) where even higher values have been documented.

Arsenic pollution as a general threat in Mongolia

It can be summarized that arsenic pollution is a serious and increasing threat for water quality in northern Mongolia and several research results confirm enhanced uptake of arsenic by Mongolian villagers (Murao et al. 2004, 2011). Uptake of arsenic by humans may not only occur with drinking water; as Mongolian coal has high arsenic content (Mongolian Ministry of Health 2004) and coal firing during

the harsh winters produces high levels of air pollution, arsenic concentration in soil and plant material from Ulaanbaatar (UB) is unusually high (Batjargal et al. 2010; Kasimov et al. 2011a, b) and similarly livestock from Tuv Aimag (UB region) showed higher arsenic contents (0.06 mg kg^{-1}) in tissue of liver and parenchyma than livestock from other Mongolian regions (Šimoník 2012). Serious human health problems may result from long time uptake of arsenic as it is known from Inner Mongolia (Guo et al. 2007; Lamm et al. 2006; Wade et al. 2009; Xia et al. 2009). For the future, the natural geogenic arsenic background has to be considered in risk assessments of (anthropogenic) water pollution for whole Mongolia. For Kharaa River basin a first estimate for natural geogenic background conditions of groundwater is given by Hofmann et al. (2014). A better knowledge of the geological background is needed to improve decisions of water engineers and mining operators. A promising approach has been recently conducted in China with a statistical risk model to classify safe and unsafe areas with respect to geogenic arsenic contamination and the related probability of arsenic concentrations exceeding the 10 mg L^{-1} threshold in ground waters (Rodríguez-Lado et al. 2013).

Bacterial biosensors: a promising option for arsenic screening in Mongolia

Major obstacles for extensive arsenic monitoring are the limited reliability or practicality, and/or the relatively high costs of existing analytical methods. Microbial reporter technologies (bacterial biosensors) have been proposed as an alternative, rapid, and cost-effective method to detect chemical species in aquatic samples (Harms et al. 2005; Siegfried et al. 2012). The bioreporter bacteria or biosensors in some cases consist of genetically modified bacteria that produce a reporter protein in response to the presence of a target chemical. Luminescent bacterial biosensors responding to arsenite and arsenate (Stocker et al. 2003; Trang et al. 2005) have been applied in the present study. The genetically modified (GMO) bioreporter bacteria included in the ARSOlux test kit remain in sealed vials throughout shipping, storage, application, disinfection and autoclaving. The Central Commission on Biologic Safety of the German Federal Office of Consumer Protection and Food Safety has stated in a risk assessment report (ZKBS 2013) that the application of the present ARSOlux biosensor field kit does not present a potential hazard to humans, animals and the environment. The risk assessment was prepared after a request of the Biosafety Committee of the Mongolian Ministry of Environment and Green Development. The Biosafety Committee of Mongolia permitted the import of the GMO ARSOlux biosensor for contained use. Biosensors such as ARSOlux could offer a cost-effective and environmentally friendly alternative to the cumbersome and expensive methods currently used for detection of arsenic and other contaminants.

Conclusions

To avoid the further contamination of groundwater and surface water resources in Mongolia with heavy metals the implementation of a set of measures is necessary. These include mitigation procedures in mining areas, containment of existing dump sites and processing water ponds, and search for save drinking water wells (see Zhang 2013), as well as capacity development of Mongolian institutions and the implementation of a monitoring system combined with effective analytical tools (Hofmann et al. 2010).

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