

## Non-carcinogenic risk assessment of groundwater in South Gobi, Mongolia

Koyomi Nakazawa, Osamu Nagafuchi, Kanji Okano, Ken'ichi Osaka, Etsuji Hamabata, Jamsran Tsogtbaatar and Javzan Chojjil

### ABSTRACT

Thirty-nine groundwater samples were collected from wells near the Tavan Tolgoi and Oyu Tolgoi mines in Mongolia and at a relatively pristine site in northern Mongolia during August to September 2013, and analyzed for the concentrations of  $F^-$ ,  $NO_3^-$ , Hg, As, Al, V, Mn, Co, Ni, Cu, Zn, Se, Mo, Cd, Sb, and Pb. A probabilistic risk assessment found that >95% of the population in the areas was at risk from drinking well water. The hazard index (HI) was >1, indicating a non-carcinogenic risk to human health. At Oyu Tolgoi, the hazard quotient (HQ) of the As concentration (mean 6.63  $\mu\text{g/L}$ ) was >1. At the northern site, the 95th percentile HQ was <1 but the 95th percentile HI was >1. The ratios of nitrogen and oxygen stable isotopes indicated that  $NO_3^-$  contamination of groundwater at Oyu Tolgoi and Tavan Tolgoi was caused by livestock waste. Mercury accumulation in livestock was examined from concentrations in livestock forage and in hair and wool samples from livestock in the South Gobi region. Sheep wool had the same level of mercury as in Japan, but camel, horse, and goat hair had high levels.

**Key words** | groundwater, hazard quotient, mining site, Mongolia, probabilistic risk assessment

**Koyomi Nakazawa**  
**Osamu Nagafuchi** (corresponding author)  
 The Kyushu University Museum,  
 6-10-1, Hakozaki, Higashi-ku,  
 Fukuoka 812-8581,  
 Japan;  
 Comprehensive Research Organizations of  
 Fukuoka Institute of Technology,  
 3-30-1 Wajiro-higashi Higashiku,  
 Fukuoka 811-0295,  
 Japan;  
 and  
 The Institute of Statistical Mathematics,  
 10-3 Midori-cho, Tachikawa,  
 Tokyo 190-8562,  
 Japan  
 E-mail: [nagafuchi@bene.fit.ac.jp](mailto:nagafuchi@bene.fit.ac.jp)

**Kanji Okano**  
**Ken'ichi Osaka**  
**Etsuji Hamabata**  
 Division of Environmental Sciences,  
 University of Shiga Prefecture,  
 Hassaka cho 2500,  
 Hikone,  
 Shiga prefecture 522-8533,  
 Japan

**Jamsran Tsogtbaatar**  
**Javzan Chojjil**  
 Mongolian Academy of Sciences,  
 Institute of Geography,  
 Baruun Selbe-15,  
 Ulaanbaatar 15170,  
 Mongolia

### INTRODUCTION

Groundwater is an essential freshwater resource for many people in dry, remote, and sparsely populated regions. Groundwater pollution caused by contaminants arising from human activities is a serious problem in such areas. Because groundwater moves through rocks and regolith, it is prone to dissolving chemicals as it moves. The quality of groundwater varies depending on the properties of the water-bearing geological strata.

Large amounts of groundwater are used in the mining industry, and the depletion and pollution of groundwater are serious environmental concerns associated with the industry. The contamination of groundwater causes various environmental problems, particularly health risk to

people, livestock, and wildlife (Sun *et al.* 2014; Giri & Singh 2015).

Mongolia is a mineral-rich country that in 2000 earned approximately equal amounts of export revenue from mining and livestock (Fujita *et al.* 2013). Subsequent rapid growth in mineral exploitation meant that by 2010, mining products were responsible for about 90% of exports (Fujita *et al.* 2013). As of 2002, some 800 deposits with many kinds of ores had been discovered, and over 600 sites were being mined (UNEP 2002). Rare earths and rare metals are abundant at many of these sites, and global capital for their development has flowed into the country (Oyu Tolgoi LCC 2014). With the widespread construction of electric

power stations, the mining of copper, zinc, uranium, gold, iron ore, coal, fluorine, and silver (Fujita *et al.* 2013) has caused the Mongolian economy to grow rapidly. However, this surge in mineral extraction has also been causing serious concerns about regional environmental degradation. Mining activities can harm air, water, soil, fauna, and flora at regional scales, and traditional nomadic lifestyles (Byambaragchaa 2015; Shimamura 2015). To achieve sustainable development, the mining industry must address such critical environmental problems. One of the more pressing environmental issues is groundwater contamination.

Environmental contamination in Mongolia has been actively studied in recent years (Badrakh *et al.* 2008; Sikder *et al.* 2013; Nagafuchi *et al.* 2014; Nadmitov *et al.* 2015). We have shown that levels of chemical species in South Mongolia's groundwater pose a potential risk to livestock and human health (Nagafuchi *et al.* 2014). We measured a few groundwater samples, and in a pasture area,  $\text{NO}_3^-$  and As values exceeded WHO drinking guidelines. Nomads are concerned that the incidence of disease among livestock is increasing (Shimamura 2015). Therefore, groundwater used as drinking water may pose a risk to human health.

To date, no assessments of risks to human health from groundwater used as drinking water are known to have been performed in this region. Because large-scale mining activity has been under way in the area for several decades, linking groundwater contamination to past mining activity was not possible in the current study. However, any health risk could be assessed, and any progression in contamination could be identified in the future using this study as a benchmark. This paper presents the results of a 2013 survey of groundwater in the vicinity of Oyu Tolgoi and Tavan Tolgoi, two large mining areas in the South Gobi Desert. For comparison, we also surveyed a relatively distant area in northern Mongolia.

To characterize the level of environmental pollution in the South Gobi, we used the results of the 2013 chemical survey to perform a preliminary non-carcinogenic risk assessment of  $\text{F}^-$ ,  $\text{NO}_3^-$ , Hg, As, Al, V, Mn, Co, Ni, Cu, Zn, Se, Mo, Cd, Sb, and Pb in groundwater. This is the first study to assess the risks to human health caused by  $\text{NO}_3^-$  and metals in the groundwater of the South Gobi Desert and northern Mongolia.

## MATERIALS AND METHODS

### Description of survey sites

In the South Gobi Desert, where precipitation is a mere 50–250 mm per year (Fujita *et al.* 2013), potable water is obtained from groundwater. The study involved collection and analysis of 39 groundwater samples for chemical analysis from two areas in the South Gobi (Oyu Tolgoi and Tavan Tolgoi) and one in northern Mongolia (Figure 1). Oyu Tolgoi, one of the world's largest new copper and gold mines, is located approximately 550 km south of Ulaanbaatar and 80 km north of the Mongolia–China border. Tavan Tolgoi, located 150 km northwest of Oyu Tolgoi and about 240 km north of the Mongolia–China border, has one of the largest untapped coking and thermal coal deposits in the world.

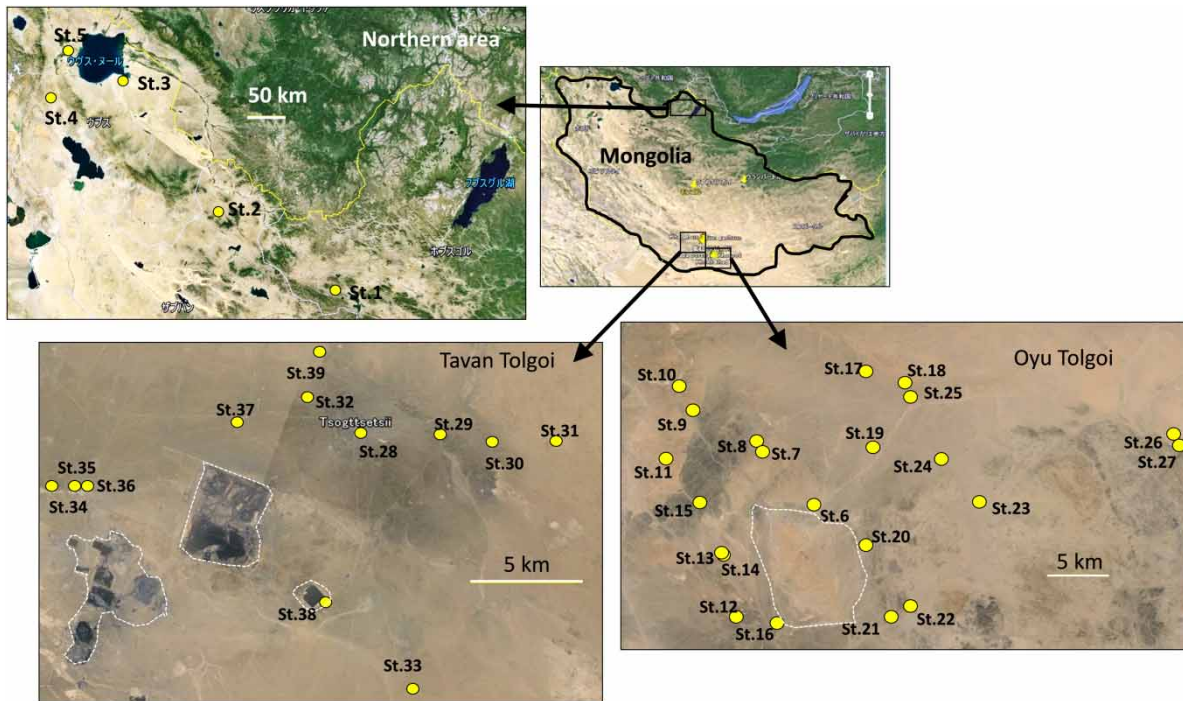
### Groundwater sampling and analysis

The sampling survey was conducted from August to September 2013. In the South Gobi, the sampling sites were wells located within 30 km of the two mining sites. pH, electrical conductivity, and temperature were measured on-site. Samples were collected from wells by bucket and were stored and transported in 250 mL polyethylene bottles without prior filtration.

After filtration (Dismic CS-25, Advantec, Tokyo), the concentrations of  $\text{F}^-$  and  $\text{NO}_3^-$  were measured by ion chromatography (761 Compact IC, Metrohm, Herisau, Switzerland).

The Hg concentration was measured by an RA-3320FG+ mercury analyzer (Nippon Instruments Co. Ltd, Osaka, Japan). Samples were then acidified with 0.4 N  $\text{HNO}_3$ . The concentrations of the other trace elements (Al, V, Mn, Co, Ni, Cu, Zn, Se, Mo, Cd, Sb, and Pb) were determined by inductively coupled plasma mass spectrometry (ICP-MS; NexION 300, Perkin Elmer, Japan). Triplicate analysis of samples yielded relative percentage differences of <5%. The As concentration was measured by atomic absorption spectrometry (AAS; AAS800, Perkin Elmer, Japan). We used the ICP-MS results only for the elements that are assigned RfD (ingestion reference dose) values by the US EPA (2014).

The nitrogen isotope ratio ( $\delta^{15}\text{N}_{\text{NO}_3^-}$ ) and oxygen isotope ratio ( $\delta^{18}\text{O}_{\text{NO}_3^-}$ ) were measured by the denitrifier method (Sigman *et al.* 2001; Casciotti *et al.* 2002) at the International



**Figure 1** | Locations of sampling sites (St. 1–St. 39) in a survey of groundwater quality in three areas of Mongolia, one in the northern area free of mining activity and two in the vicinity of large mines, Tavan Tolgoi and Oyu Tolgoi. Broken lines show the mining area.

Research Center for River Basin Environment, University of Yamanashi, Japan.

### Livestock hair and feed sampling and analysis

In general, human hair and blood are used as biological indicators for estimating exposure to methyl mercury (Me-Hg) (US EPA 1997a). Mercury ingested or absorbed by animals tends to concentrate in their hair also. Therefore, the Hg concentration in hair is used as a bio-indicator of Hg exposure (NIMD 2013). During August and September 2013, hair or wool samples were obtained from the livestock of Mongolian nomads at South Gobi: sheep ( $n = 11$ ), camels (calves:  $n = 2$ , adults:  $n = 1$ ), horses ( $n = 1$ ), and goats ( $n = 3$ ). Pasture grass (aboveground biomass) samples ( $n = 5$ ) were also obtained from five nomad families at South Gobi. For comparison, samples were also obtained from sheep ( $n = 5$ ) and an exotic pasture grass ( $n = 1$ ) from an uncontaminated experimental field of the University of Shiga Prefecture, Japan. Each sample was washed twice by hand with commercial neutral detergent, rinsed three times in

tap water, air-dried, and placed into a small polyethylene bag for storage before measurement. Hg in samples was analyzed by the thermal decomposition and gold amalgamation AAS method (model MA-2, Nippon Instrument Co. Ltd, Japan).

### Risk assessment

Non-carcinogenic risk was assessed by the US EPA method (US EPA 2010, 2011). Metals in water are considered to pose a risk to human health mainly through direct consumption of water.

The exposure doses through ingestion of water were calculated according to US EPA (2010):

$$D_i = \frac{C_{wi} \times IR \times EF \times ED}{BW \times AT} \quad (1)$$

where  $D_i$  is the exposure dose through ingestion of water ( $\mu\text{g}/\text{kg}/\text{day}$ ),  $C_{wi}$  is the concentration of metals in drinking water ( $\mu\text{g}/\text{L}$ ),  $IR$  is the ingestion rate (L/day),  $EF$  is the

exposure frequency (days/year), *ED* is the exposure duration (years), *BW* is average body weight (kg), and *AT* is the pathway-specific period of exposure for non-carcinogenic effects (days). If we could not obtain a suitable value, we based values except  $C_{wi}$  and *EF* on US EPA (1989):  $IR = 2$  L/day; *ED* = the national upper 90th percentile value, i.e., 30 years;  $BW = 70$  kg;  $AT = ED \times 365$  days/year = 10 950. *EF* was set at 90 days/year because the typical usage pattern of the nomad people is to use well water only in the summer months.

The hazard quotient (HQ) was calculated from  $HQ = D/RfD$ , where *D* is the exposure dose through ingestion of water defined in Equation (1), and *RfD* is the value of the ingestion reference dose obtained from US EPA (2014).

To assess the overall potential or non-carcinogenic effects posed by more than one chemical, a hazard index (HI) approach was based on the US EPA guidelines for health assessment of chemical mixtures (WHO 2006; US EPA 2014). This approach assumes that simultaneous subthreshold exposure to several chemicals could result in an adverse health effect (US EPA 1986). The HI was calculated from the sum of the HQs of *n* hazardous species:

$$HI = \sum_{i=1}^n HQ_i \quad (2)$$

### Boundaries for assessment of risk to human health

We considered three exposure pathways: dermal, oral, and inhalation. We focused on the risk to the resident nomads of chronic metal exposure via drinking water. We used the *RfD* value of the total concentration of each element (in all oxidation states) for the risk assessment. We assumed that the nomads use the well water only in summer, because in other seasons they rely on snowmelt, so we collected samples only during summer, and therefore set *EF* to 90 days (June to August). We also assumed that each well is used for drinking at the same frequency. To conduct a probabilistic risk assessment, we assumed a log-normal concentration distribution for each element in the groundwater in each area: for some variables, this distribution may provide a better fit than a normal distribution, and environmental data generally do not take negative values.

Therefore the log-normal distribution is empirically useful because it cannot take negative values and should fit the data well (Kammen & Hassenzahl 1999; Nakazawa *et al.* 2016). Random Monte Carlo simulations were performed with Crystal Ball software (Oracle Co., KKE Inc., Japan). The Latin hypercube sampling method was used with 10,000 iterations to define the probabilistic distribution (US EPA 1997c).

## RESULTS AND DISCUSSION

### Physicochemical characteristics of groundwater samples

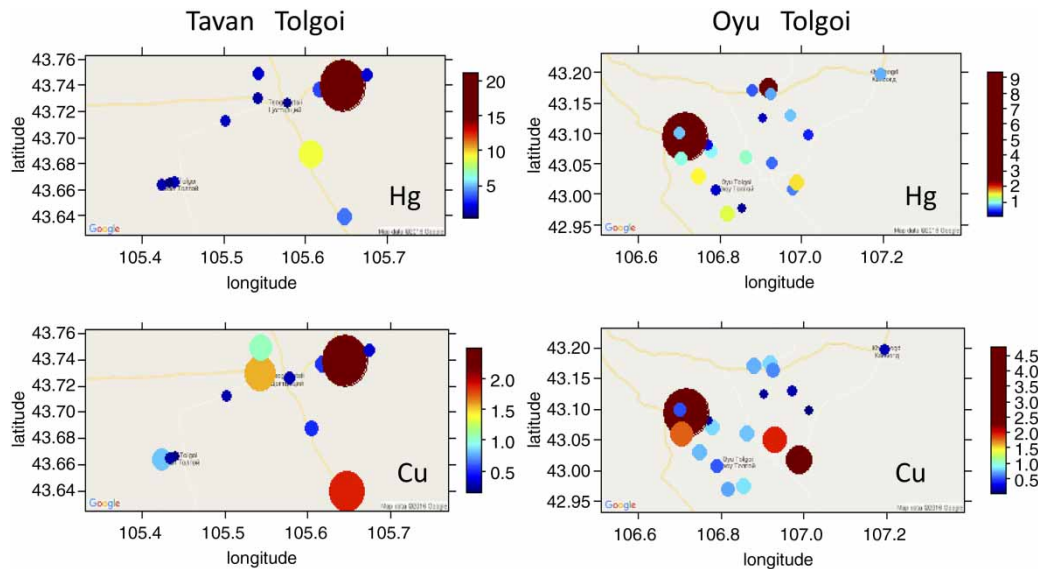
Generally, Hg indicates contamination from coal mines, and Cu indicates contamination from copper mines. The concentration of Hg was higher at Tavan Tolgoi than at Oyu Tolgoi (Figure 2). That of Cu was high at Oyu Tolgoi (Figure 2). The concentrations of both indicate that the groundwaters are influenced by mining.

The results of the analyses are summarized in Table 1, along with World Health Organization guidelines for the maximum permissible levels of some contaminants in drinking water (WHO 2006). Concentrations were below the maximum levels for the most part, but those of  $F^-$ ,  $NO_3^-$ , and As in some samples exceeded the levels at Oyu Tolgoi and Tavan Tolgoi, as in our previous study (Nagafuchi *et al.* 2014).

### Estimation of risk to human health of drinking water

Few previous studies have reported the non-carcinogenic risk of  $NO_3^-$ ,  $F^-$ , and metals in groundwater in Mongolia, and in general only limited information about the toxicity effects of these species is available. Using the results of the groundwater analyses, we performed a non-carcinogenic probabilistic risk assessment of  $NO_3^-$ ,  $F^-$ , and 13 metals in the samples obtained from the three areas.

The US EPA recommends that the HQs of individual chemical species and the total non-carcinogenic HI should be <1 (US EPA 1989). In the samples from the northern area, no values exceeded 1 (Figure 3). However, among the 15 species analyzed,  $F^-$  contributed the most to the



**Figure 2** | Distribution of Hg and Cu concentration in groundwater.

95th percentile value of HI, followed by  $\text{NO}_3^-$ , As, and Mo. As a result, 4.3% of the population was considered to be at risk by ingesting drinking water ( $\text{HI} > 1$ ) (Figure 4).

In Oyu Tolgoi, the 95th percentile HQ of As (1.63) and  $\text{F}^-$  (1.10) exceeded 1. After As and  $\text{F}^-$ , Mo and  $\text{NO}_3^-$  contributed the most to the 95th percentile value of HI of the 15 species, which totaled 3.85. In Tavan Tolgoi, only the HQ of  $\text{F}^-$  exceed 1 (1.12). After  $\text{F}^-$ , the greatest contributions to the 95th percentile value of HI came from  $\text{NO}_3^-$ , As, Mn, and Mo. As a result, the 95th percentile HI exceeded 1 (3.18). As a result, 95% of the population exceeded the no-risk HQ of 1 at the 95th percentile in both mining areas (Figure 4). The estimated human health risks of drinking water are clearly higher in Oyu Tolgoi and Tavan Tolgoi than in the northern area.

Our results suggest that  $\text{NO}_3^-$ ,  $\text{F}^-$ , As, Mn, and Mo pose the most serious health risks for the use of groundwater for human consumption in Mongolia. Future research efforts should focus on the origin of these elements. We have taken some steps in that direction by examining the origin of  $\text{NO}_3^-$  (below).

It should be noted that the assessment methods used in this study involved possible sources of variability and uncertainty. Our sampling was conducted in August to September 2013; therefore, we did not examine the seasonal variation in water quality, which could reveal different levels of

risks. However, the nomads' reliance on snowmelt outside the summer months means that seasonal variation in groundwater quality is of less relevance. Nevertheless, further investigation of possible contamination of snowmelt is also needed. Differences in consumer age and exposure conditions could also result in different risks. In particular, we could not take into consideration the peculiarities of the nomad lifestyle with respect to the frequency of groundwater consumption and the periods of time spent in different grazing areas. Furthermore, the possibility of underestimating risk should be considered: we considered only the risk of drinking water and omitted the risk associated with food intake.

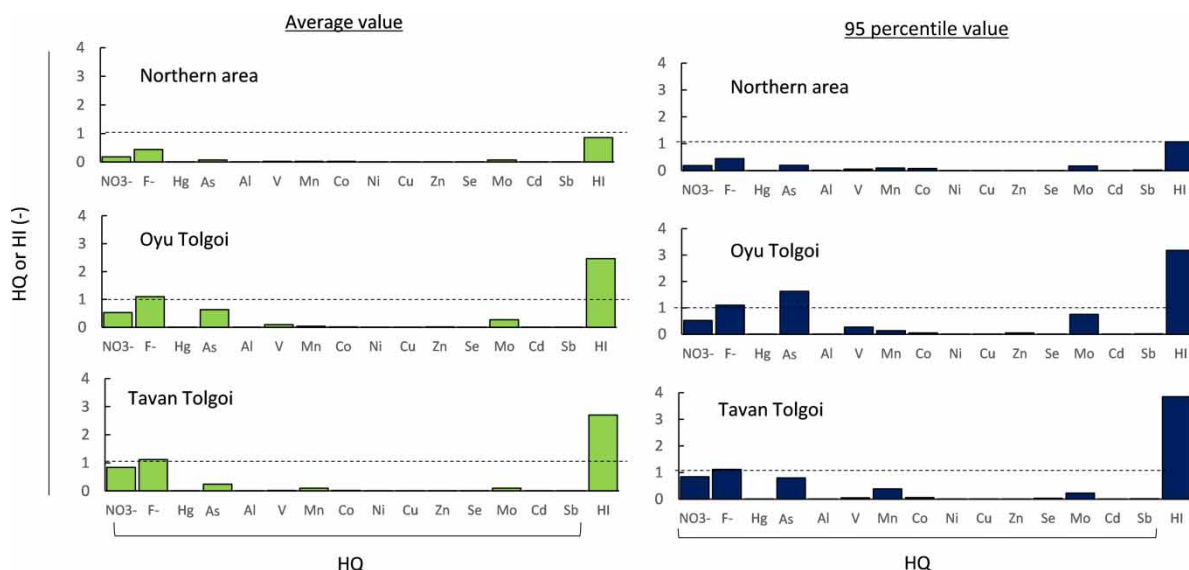
### Dietary intake of elements

Exposure to potential contaminants in the diet should also be considered. Since reliable data on food intake by nomads were not available, we did not directly evaluate this pathway. But since the diet of the nomadic grazers includes regular intake of meat and fermented milk derived from their own livestock, we could indirectly evaluate the potential for dietary intake of pollutants by measuring pollutant levels in the livestock. We analyzed exposure of livestock to mercury by sampling hair or wool from goats, sheep, horses, and camels and their food sources (Table 2).

**Table 1** | Summary statistics for the chemical analysis of groundwater samples from three areas of Mongolia

Parameter	Unit	Northern area (n = 5)				Oyu Tolgoi (n = 22)				Tavan Tolgoi (n = 12)				WHO guideline
		Ave.	SD	Min.	Max.	Ave.	SD	Min	Max.	Ave.	SD	Min.	Max.	
F <sup>-</sup>	mg/L	0.62	0.24	0.37	0.90	1.54	1.40	ND <sup>†</sup>	5.46	1.56	0.93	0.38	3.78	1.5
NO <sub>3</sub> <sup>-</sup>	mg/L	10.3	8.40	2.68	24.5	29.6	20.1	3.76	80.3	47.1	36.2	0.25	110	50
Hg	ng/L	9.01	4.61	4.72	16.5	1.15	1.92	0.05	9.37	3.80	5.90	0.47	21.0	6,000
As	μg/L	0.75	0.73	0.05	1.95	6.63	5.69	0.19	25.8	2.57	3.72	0.23	12.2	10
Al	μg/L	131	277	1.23	626	90.0	350	0.47	1,650	46.1	88.8	1.36	247	–
V	μg/L	3.91	3.13	0.33	8.22	16.0	18.2	0.70	61.5	3.78	2.15	0.07	6.58	–
Mn	μg/L	23.0	45.9	0.01	105	30.7	100	0.09	475	84.7	275	0.55	957	–
Co	μg/L	0.23	0.41	0.01	0.96	0.15	0.27	0.01	1.31	0.17	0.32	0.04	1.16	–
Ni	μg/L	1.75	2.67	0.01	6.21	0.50	0.62	0.03	2.16	0.57	0.64	0.06	1.98	70
Cu	μg/L	1.55	1.86	0.15	4.48	0.82	1.08	0.01	4.77	0.81	0.76	0.14	2.49	–
Zn	μg/L	22	46	0.30	105	136	394	2.46	1,900	46.3	18.1	8.07	70.2	–
Se	μg/L	0.42	0.44	0.08	1.12	0.55	0.43	0.04	1.53	1.93	1.77	0.21	5.32	40
Mo	μg/L	12.1	10.0	1.14	24.3	48.1	47.4	4.15	177	17.8	11.1	4.98	47.6	–
Cd	μg/L	0.01	0.01	ND	0.03	0.03	0.03	0.01	0.10	0.01	0.01	ND	0.03	3
In	μg/L	0.01	ND	ND	0.01	0.01	ND	ND	0.02	ND	ND	ND	0.01	–
Sb	μg/L	0.09	0.09	0.02	0.22	0.09	0.07	0.01	0.26	0.09	0.08	0.02	0.30	20

ND = below the detection limit.



**Figure 3** | Average value and 95th percentile value of HQ or HI in each element estimated in Northern area, Oyu Tolgoi, and Tavan Tolgoi.

In the samples obtained from the South Gobi, the concentration of Hg in horse hair was  $10.0 \mu\text{g}/\text{kg}$  ( $n = 1$ ) and the average concentration in goat hair was  $8.31 \mu\text{g}/\text{kg}$  ( $n = 3$ ). Both were higher than the concentrations in sheep's wool,  $4.8 \mu\text{g}/\text{kg}$  ( $n = 11$ ), from the same area. In general, the fleece is removed from sheep every year, but the coats of horses and goats (surface hair) are not, which might explain the difference.

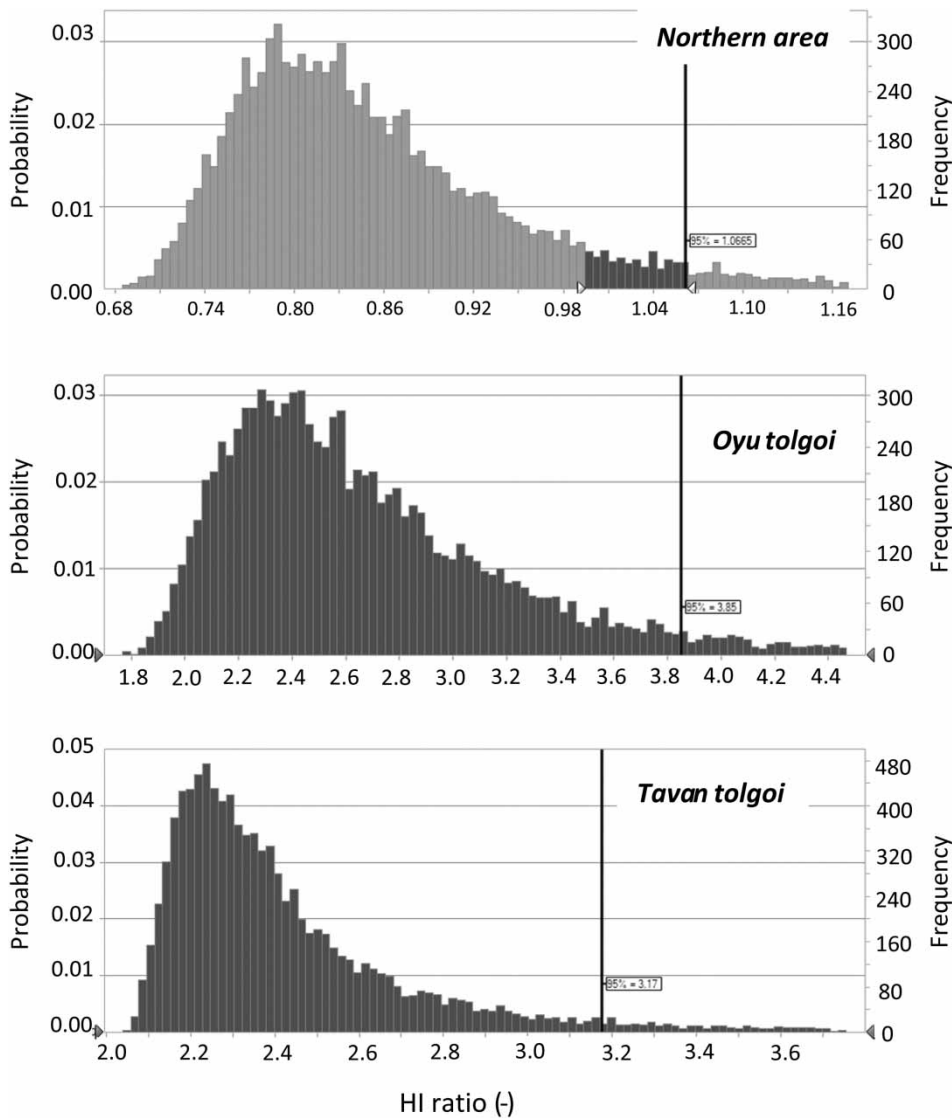
The concentration of Hg was high also in the hair of camel calves, at  $11.63 \mu\text{g}/\text{kg}$  ( $n = 2$ ), but that in the sample of adult camel hair was only  $0.74 \mu\text{g}/\text{kg}$ . In general, mercury is excreted in breast milk (US EPA 1997b), so milk is the most likely source of mercury exposure in camel calves. Because of inter-species variability, we could not use the concentration of Hg as an indicator of pollutants in livestock, so further study is needed.

The concentration of Hg in wool samples from sheep of the South Gobi was higher than that from sheep in Japan, but not significantly ( $t$ -test,  $P > 0.05$ ). Thus, to what level of mercury contamination are sheep exposed? The concentrations in pasture grass were  $3.35 \mu\text{g}/\text{kg}$  in the South Gobi and  $3.09 \mu\text{g}/\text{kg}$  in Japan, and therefore comparable. The criterion set by the Feed Safety Law in Japan for mercury in pasture grass is  $<400 \mu\text{g}/\text{kg}$  (FAMIC 2014), about 100 times the concentration in the pasture grass in the South Gobi. We calculated the total daily mercury intake by

sheep by assuming a dietary dry matter consumption of  $1.0 \text{ kg}/\text{d}$ , and a total ingestion of 22.6% soil and 77.4% pasture grass (Smith *et al.* 2009). The mercury intake from pasture grass in the South Gobi and Japan were almost equal at  $2.59 \text{ mg}/\text{d}$  and  $2.39 \text{ mg}/\text{d}$ , respectively. Thus, sheep are not presently accumulating mercury. However, high concentrations were observed in the hair of the sampled camel calves, goats, and horse. Additional work on the metabolism of mercury in livestock or estimation of the exposure of livestock to mercury would help to better understand its accumulation in livestock in Mongolia. In addition, Smith *et al.* (2009) found that Pb concentrations exceeded the safety threshold for sheep in winter (seasonal variation was recorded) in a mine-tailing-contaminated area in Wales. Therefore, further study is needed to evaluate metal accumulation in livestock in Mongolia and the risk to human health it causes by the consumption of livestock products.

### Origin of $\text{NO}_3^-$ in groundwater

In our previous study (Nagafuchi *et al.* 2014), the  $\text{NO}_3^-$  concentration in groundwater exceeded the WHO guideline value. In this study, the HQ value of  $\text{NO}_3^-$  in South Gobi tended to be high compared with the other species. We measured the nitrogen isotope ( $\delta^{15}\text{N}_{\text{NO}_3^-}$ ) and oxygen



**Figure 4** | Probabilistic distribution of HI estimated in Northern area, Oyu Tolgoi and Tavan Tolgoi. Log-normal distributions were assumed for each element concentration in these three areas and random Monte Carlo simulations were performed.

isotope ( $\delta^{18}\text{O}_{\text{NO}_3}$ ) ratios (Figure 5) to examine the possible origin of the  $\text{NO}_3^-$ . The ratios of  $\delta^{15}\text{N}_{\text{NO}_3}$  in Oyu Tolgoi and Tavan Tolgoi ranged from 0‰ to 10‰ and those of  $\delta^{18}\text{O}_{\text{NO}_3}$  varied widely from 5‰ to 25‰. There was almost no difference in the  $\delta^{15}\text{N}_{\text{NO}_3}$  and the  $\delta^{18}\text{O}_{\text{NO}_3}$  values between Oyu Tolgoi and Tavan Tolgoi, which are consistent with human or livestock waste sources (Kendall *et al.* 2007). During our survey, we saw nomads visiting the wells in these areas and drawing groundwater for their livestock. This activity, and the elevated  $\text{NO}_3^-$  concentrations, suggest that the groundwater has been contaminated by

livestock waste. From our risk analysis, the HQ value of  $\text{NO}_3^-$  did not exceed 1; however, continuous monitoring would be needed in the possible event of any intensification of well water usage.

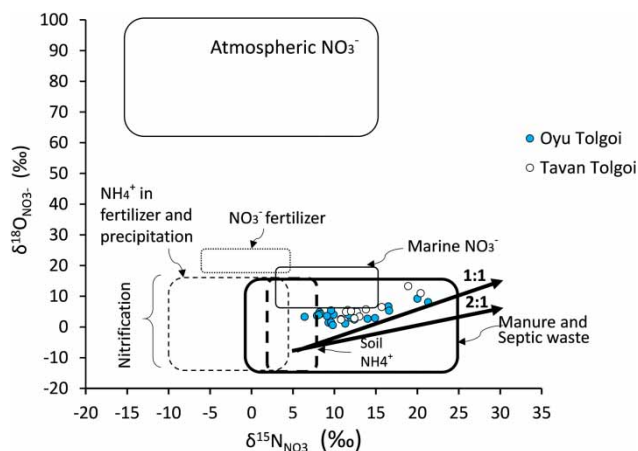
## CONCLUSIONS

We estimated the potential risk to human health from drinking groundwater near the world's largest coal mine (Tavan Tolgoi), a copper and gold mine (Oyu Tolgoi), and a



**Table 2** | Mercury concentration in wool or hair from sheep, camel, horse and goat and in forage grass in Mongolia and from sheep wool in Japan

Source of sample	Site	n	Average concentration ( $\mu\text{g}/\text{kg}$ )	Standard deviation (-)
Sheep	Adult Mongolia	11	4.80	4.50
	Adult Japan	5	2.50	1.20
Camel	Calf Mongolia	2	11.60	4.77
	Adult Mongolia	1	0.74	-
Horse	Adult Mongolia	1	10.00	-
Goat	Adult Mongolia	3	8.31	4.25
Forage grass	- Mongolia	6	3.41	0.57
	- Japan	1	3.09	-

**Figure 5** | Ratios of stable isotopes of nitrogen and oxygen from groundwater samples near Oyu Tolgoi and Tavan Tolgoi. Colored rectilinear regions of the coordinate space and their annotations indicate the ranges of values consistent with the origin of nitrogen and oxygen isotopes as reported by Kendall et al. (2007).

relatively pristine area (in northern Mongolia). High concentrations of each chemical species were associated with the Tavan Tolgoi and Oyu Tolgoi areas, and these may pose a non-carcinogenic risk to human health. Isotope ratio analysis indicated that high levels of  $\text{NO}_3^-$  originated from livestock waste. High mercury concentrations were not observed in wool or sheep forage, but were observed in the hair of camels, goats, and a horse. These were only preliminary exploratory samples, so more accurate estimates of the contribution of food intake to the HQ and HI are needed.

This research indicates groundwater pollution in the South Gobi Desert, which poses a risk to human health. Continued work is needed to determine trends in this

pollution, and all stakeholders need to be involved in discussing ways to achieve sustainable development of mineral resources and to establish a clearer framework for the accurate evaluation of human health risk in this rapidly developing region.

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