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The Provenance of Arsenic in Southeast Asia Discovered by Trace Elements in Groundwater from the Lowlands of Nepal

Barbara Mueller

Abstract

Arsenic concentrations in groundwater extracted from quaternary alluvial sediments pose a serious health issue for inhabitants living in several countries in Southeast Asia. A widely approved hypothesis states that reductive dissolution of Fe-bearing minerals releases As oxyanions to ground water and the original source of As has to be located in mafic rocks occurring across the entire Himalayan belt. Yet, recent trace element analyses of ground water from the lowlands (Terai) of Nepal show a clear decoupling of As and Fe. The positive correlation of K, Na, and trace elements like Li, B, and Mo with arsenic points out to clay minerals hosting the toxic element. This pattern of trace elements found in the ground water of the Terai also advocates against an original source of As in mafic rocks. The lithophile elements like Li, B, P, Br, Sr, and U reflect trace element composition typical for felsic rocks as an origin of As. All the mentioned elements are components of clay minerals found ubiquitously in some of the most characteristic felsic rocks of the Nepal Himalaya: metapelites and leucogranites—all these rocks exhibiting a high abundance of especially B, P, and As besides Cd and Pb.

Keywords: arsenic, groundwater, trace elements, felsic, Himalaya

1. Introduction

Arsenic concentrations found in the groundwater in quaternary alluvial sediments in the lowland Terai region of Nepal and other countries of South Asia (Bangladesh, India, Nepal, Myanmar, China, Vietnam, Cambodia, and China) often exceed the World Health Organization (WHO) drinking water guideline (10 $\mu\text{g/L}$). The oral intake of arsenic causes various detrimental health issues: skin lesions including pigmentation changes, mainly on the upper chest, arms, and legs, keratosis of the palms of the hands and soles of the feet, and as the most severe effect, cancer of the skin and internal organs [1–3]. The origin of the arsenic contamination is clearly geogenic, and its elevated concentrations in natural ground waters are considered to be due to natural weathering of the Himalayan belt [4–9]. These quaternary alluvial sediments are carried by the Ganga-Brahmaputra river system and build up the Himalayan foreland basin and the Bengal fan—one of the largest modern fluvial deltas of the world [10, 11]. Arsenic is not known as an

element with a high abundance in the Earth's continental crust. Sulfide-bearing mineral deposits are the most common sources of As which also has a strong affinity for pyrite. Weathering of pyrite leads to formation of hydrous iron oxides containing As, but clay minerals are important hosts of As as well. Sediments usually containing 1–20 mg/kg (near crustal abundance) of arsenic can already be a cause of high dissolved arsenic ($>50 \mu\text{g/L}$) in groundwater if one or both of two potential “triggers”—an increase in pH above 8.5 or the begin of reductive iron dissolution—are initiated [12]. Pyrite represents the largest reservoir of As. Besides this iron sulfide, As is mainly concentrated in hydrous iron oxides and clay minerals. Arsenic can be easily solubilized in groundwaters depending on pH, redox conditions, temperature, and solution composition. The sediments of the Terai plain in Nepal are commonly reducing with a pH between 7.0 and 7.8. Beyond that, As in groundwater is only weakly negatively or not correlated with Fe (decoupling of As) but positively correlated with lithophile elements like Na and K. Decoupling between aqueous As and Fe has also been described by [9, 13–15]. The mentioned elements are specifically derived from aluminosilicates such as clay minerals during weathering. More immobile elements such as Fe and Al will therefore be concentrated in the remnants. Clay minerals that are extremely fine-grained are noted to be compatible of adsorbing arsenic to a high degree [16–18]. So far, there is basically a small number of source materials recognized as significant contributors to arsenic in the aquifer: for example, organic-rich or black shales, Holocene alluvial sediments with slow flushing rates, mineralized and mined areas (most often gold deposits), volcanogenic sources, and thermal springs. As there is no correlation found between As and Fe concentrations in groundwater in the Terai of Nepal, an obvious correlation between lithophile element concentration and As the initial source rocks is of felsic and not mafic composition. These trace elements in groundwater reflect the origin of the arsenic in the high Himalayas of Nepal.

2. Geology

All of the four major Himalayan tectonic units are exposed in the Terai (lowland of Nepal): (1) the Tethys Himalaya, delimited at the base by the South Tibetan Detachment system (STDS), (2) the Higher Himalayan Crystallines (HHC) delimited at the base by the Main Central Thrust I (MCT I), (3) the Lesser Himalaya (LH) divided into upper and lower Lesser Himalaya is delimited at the base by the Main Boundary Thrust (MBT), and (4) the Siwaliks, delimited at its base by the Main Frontal Thrust (MFT) as much as the quaternary foreland basin. In the Terai once, east-west flowing rivers built up the Archean crystalline formations deep beneath the Alluvium of the Terai as well as the marine sedimentary deposits forming the high Himalayas and the Siwalik formation. The debris of these formations can be found within confined space [19].

These four units include a wide range of diverse rocks of metamorphic, sedimentary, and igneous in origin. Their differential erosion is accounting for some of the groundwater arsenic heterogeneity seen in the foreland and delta (e.g., [5, 7, 20, 21]). In the realm of provenance of the Terai sediments, the Tethys Himalaya is made up of 10 km of various metasedimentary rocks (limestones, calc-schists, shales, and quartzites) ranging from Cambrian to Jurassic. Leucogranites like the Manaslu leucogranite are also found emplaced within the Tethyan rocks (e.g., [22]).

The Terai region of Nepal itself is the direct prolongation of the Bengal Delta Plain (BDP), and it is the elongation of Indo-Gangetic trough. The Terai plain is composed of quaternary sediments including molasse units consisting of gravel,

sand, silt, and clay and represents an active foreland basin. The rivers in the Terai generally flow from north to south. All major rivers have their origin in the high Himalayas, while minor rivers also emerge from the proximate Siwalik Hills, and hence sediments are deposited in the form of a fan along the flanks of the Terai basin. Organic material is widespread in the fine sediments which were once deposited in inter-fan lowlands, in wetlands, and in swamps [23–25].

The district of Nawalparasi is the most acute studied Terai province related to arsenic-contaminated groundwater in Nepal. The lithology of the Nawalparasi province sedimentary basin belongs to Holocene alluvium including the present-day alluvial deposits, channel sand, and gravel deposits as well as outwash deposits [26]. The major river, the Narayani/Gandaki, which has its source in the Higher Himalaya, flows along the eastern boundary of the Nawalparasi district and exerted a major influence on the underlying unconsolidated Holocene fluvial deposits that include the floodplain aquifer system. Atypical for the Terai, where finer sediments typically increase toward the south, in Nawalparasi, fines predominate in the north, and sand and gravels are found near the Nepal-India border [27]. In the areas with fine-grained sediments, elevated concentrations of As are typically recorded [15, 28, 29].

3. Material and methods

3.1 Sample collection

Around 20 years ago, co-workers from CAWST (Centre for Affordable Water Sanitation Technology) Calgary, Canada, in cooperation with ENPHO (Environment and Public Health Organization) Kathmandu, Nepal, began to install iron-assisted bio-sand filters built on the basis of arsenic removal from water using zero-valent iron (ZVI) media. The modified model now used in Nepal is known as Kanchan filter [30, 31]. Due to growing concerns about the malperformance of some of these filters, a groundwater sampling campaign was initiated by CAWST together with Eawag (Swiss Federal Institute for Environmental Science and Technology), Dübendorf, Switzerland. As reported in [32] and CAWST, the Kanchan filter efficiency under field conditions operating for a long period has scarcely been observed. Since a part of the Kanchan filters still had effluent arsenic concentrations exceeding the Nepal drinking water quality standard value (50 µg/l), groundwater, intermediately filtered, and effluent water for trace element analyses were sampled in October 2015 (post-monsoon). A second field campaign was arranged in pre-monsoon time (April 2017) in order to detect for differences in arsenic concentration in groundwater between the two seasons. Filters were also inspected at household levels. Measurements to improve the efficiency of the filters are under progress.

Thirty-five water samples from around Ramgram, the capital of the district Nawalparasi, were collected from hand pumps in October 2015 and again in April 2017. All pumps were thoroughly flushed before sample collection. Household for sample collections was selected referring to a register established by ENPHO including all groundwater samples exceeding the Nepal drinking water quality standard value (50 µg/L). Sampling sites consisted of groundwater from private tube wells located within the municipalities of Ramgram (former name, Parasi, the capital of the district Nawalparasi), Manari, Panchanagar, Sukauli, and Tilakpur (within proximity of Ramgram). Water samples were acidified with HNO₃ and sent to the laboratory in Switzerland for further examination.

3.2 Trace element analysis

All trace elements in the groundwater samples were determined by ICP-MS (Agilent Technology, 7500 Series, Agilent Technologies, Waldbronn, Germany) at Eawag, Dübendorf, Switzerland, after 1:2 dilution with 0.5 M HNO₃. Each measurement was conducted in triplicate. All ICP-MS determinations agreed to within 3–5% standard deviation [33].

4. Results and discussion

Figure 1a shows the correlation between As and Fe concentrations in the 35 groundwater samples taken in spring 2017. Results from analyses of samples collected autumn 2015 exhibit very similar trends and are therefore not included in this section. There is no visible correlation between As and Fe, and the correlation coefficient is only given for illustration. In contrast to the obvious decoupling between As and Fe, the concentration of As markedly and positively depends on the concentration of various lithophile elements (K shown as an example in **Figure 1b**). As is positively correlated with Na, Li, B, and Mo and negatively correlated with Ca, Mn, and As. Evidently, the reason for such a paradigm is mainly the derivation of As from silicates like clay minerals and feldspars and much less from Fe hydroxides and pyrite. Even though in [34] it is reported about a positive dependence of As on Fe in West Bengal and in [35] a positive correlation between As and Fe ($r = 0.77$) in the aquifer of the Nawalparasi district is also mentioned, a decoupling between aqueous As and Fe has also been observed in [13–15].

In [15] it is stated correctly that decoupling between Fe and As may result from sorption of Fe to other surfaces (i.e., clays) or precipitation of Fe(II) minerals, such as siderite. Anyways, the diagram in **Figure 1b** explicitly shows the correlation between As and the lithophile element K as an example. K as well as Na, Mg, and Sr (replacement of Na and K) can easily be dissolved from interlayers of clay minerals, Na, K, and Sr as well from alkali feldspars or the borosilicate tourmaline. Li, B, and Mo represent common trace elements found in micas; Li and B moreover are main components of tourmaline (general formula, (Ca,K,Na)

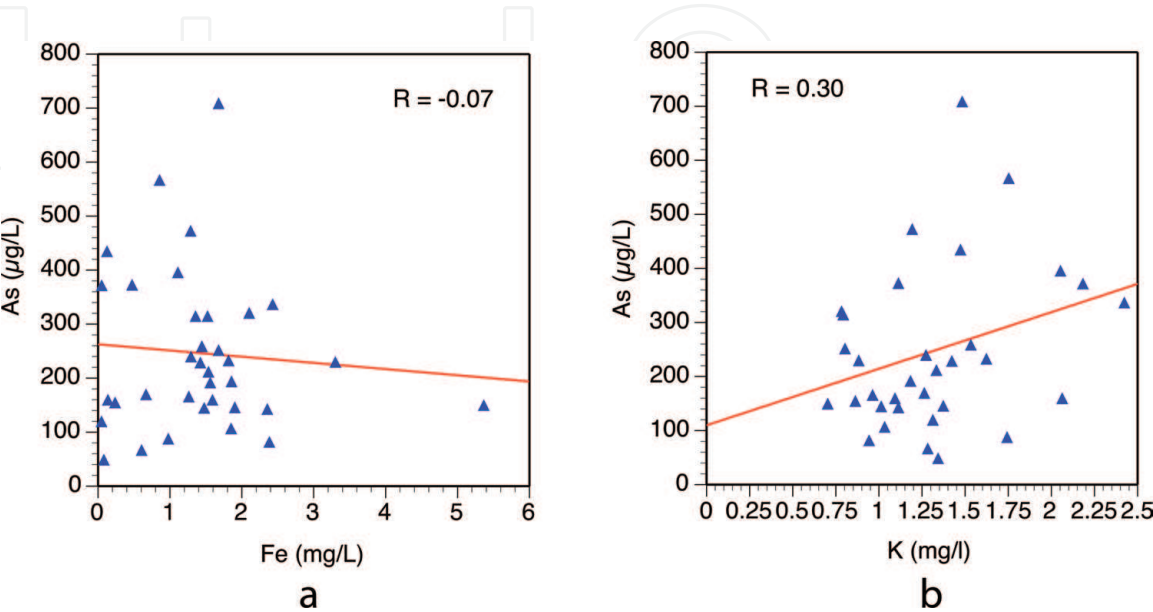


Figure 1. (a) Apparent decoupling between Fe (mg/l) and As (µg/l). The correlation coefficient (r) is included in the diagram for illustration though it is not significant. (b) Correlation between K (mg/l) and As (µg/l).

$(\text{Al, Fe, Li, Mg, Mn})_3(\text{Al, Cr, Fe, V})_6(\text{BO}_3)_3(\text{Si, Al, B})_6\text{O}_{18}(\text{OH, F})_4$. Besides this said, tourmaline is one of the very rare minerals fitting a significant amount of boron in its structure. As reported in [36], authors found tourmaline-containing aquifers enriched in As in West Bengal, India; in [37] it is clearly described how trace elements of groundwater in the Terai of Nepal refer to the origin of the mentioned soil minerals being decomposed from tertiary leucogranites (rich in B) in the High Himalayas. As Fe is predominantly contained in the octahedral layer of clay minerals such as biotite (general formula, $\text{K}(\text{Mg, Fe}^{2+}, \text{Mn}^{2+})_3[(\text{OH, F})_2](\text{Al, Fe}^{3+}, \text{Ti}^{3+})\text{Si}_3\text{O}_{10}]$) and fits hardly in the interlayer, so it is not possible to exchange Fe against K or Na. However, the likely source of As due to the positive correlation between Na, K, and As points out to silicate minerals as As can be readily adsorbed on the surface and edges of these minerals. Muscovite $\text{KAl}_2[(\text{OH, F})_2]\text{AlSi}_3\text{O}_{10}]$ is another frequent clay mineral found in the sediments of Nawalparasi and is well known as a carrier of K and Li, B, and Mo as trace elements.

This obvious decoupling of As from Fe in the groundwater excludes Fe minerals like Fe hydroxides and pyrite being the source of arsenic in the alluvial sediments.

Above all, several trace elements (Li, B, P, V, Cr, Mn, Cu, Zn, Se, Br, Sr, Mo, Cd, P, and U) analyzed in the groundwater samples of Nawalparasi boreholes are among the most prominent besides arsenic which could be found in relevant concentrations. Particularly the presence of the lithophile Li, B, P, Mn, Br, Sr, and U in the groundwater is a striking feature. Siderophile elements like Cu or Zn could be found in minor concentrations or in a few samples only. The immediate attention was attracted by boron as this element is rarely found in significant amounts in common minerals like silicates. Tourmaline is one of the very rare minerals incorporating a significant portion of boron in its structure. As already mentioned in [38], boron in spring waters in the Peshawar basin and surroundings in the Himalayan foreland of Pakistan is closely associated with igneous complexes (most probably with the tourmaline-rich tertiary leucogranites). Even boron is widely known to be present in salt water; such an influence can be neglected in landlocked Nepal having no link to the ocean. Thermal springs can be located in various areas of the Nepal Himalaya, but their influence is considered to be marginal taking the widespread occurrence of tertiary leucogranites besides metapelites and black shales into account. Above all, in [39], elevated boron in metasedimentary rocks of the lesser Himalaya (up to 322 ppm) as well as in the Manaslu leucogranite (up to 950 ppm) where tourmaline represents the boron-containing mineral is reported. Tourmaline-containing aquifers enriched in As in West Bengal, India were detected in [36]. These authors conclude therefore that the heavy mineral assemblage of these aquifers (opaque minerals, garnet, tourmaline, kyanite, rutile, and zircon) prove a mixed metamorphic and igneous provenance for the eroded and deposited materials.

As the anion S was hardly detectable, the oxidation of pyrite in the sediments (suggested, e.g., by [40]) which would lead to an increased concentration of SO_4^{2-} in the groundwater can be clearly ruled out as a mechanism of arsenic release into groundwater. Despite these convincing facts, ophiolites were seen as the initial source of arsenic contained in arsenopyrite (e.g., [6, 16]). But ophiolites do not exist in the Nepalese Himalaya.

In conclusion, the diagram in **Figure 2** presents the most prominent trace elements in groundwater from Nawalparasi district in comparison with the few available data of the Macusani obsidian glass (peraluminous in composition, enriched in As-B-F-P). The data for comparison are taken from [41]. In [42], it was already declared that these volcanic rocks are compositionally and mineralogically equivalent to the Manaslu leucogranite. In article [41], it is reported for the first time that a significant accumulation of arsenic in a peraluminous glass from Macusani (SE Peru) which is representative of anatectic melts is derived from metasedimentary crustal

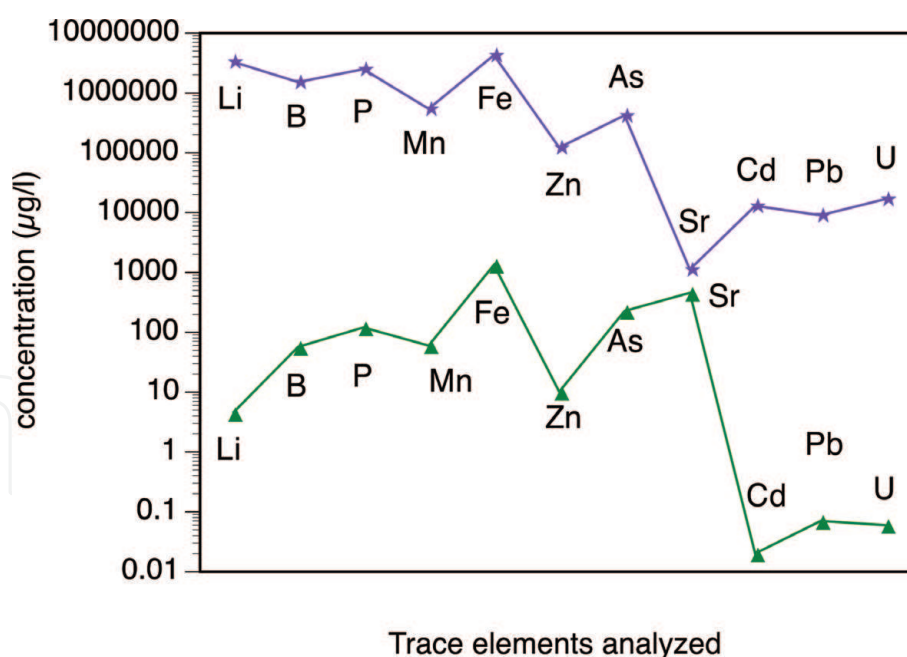


Figure 2.

The most noticeable trace elements in groundwater from Nawalparasi district (green triangles) compared with the few available data of the Macusani obsidian glass (peraluminous in composition, enriched in As-B-F-P). The data for comparison (purple stars) are taken from [41]. Note the logarithmic scale for comparison of concentrations.

protoliths. In this glass, the authors of [41] found accumulations by factors of 10–100 for As, compared with the mean continental crust values and other incompatible trace elements (e.g., Be, B, Rb, Sn, Sb, and Ta), and by factors of 100–200 for Li, Cd, and Cs. In a second article [43], the authors of reference [41] state that remarkable concentrations of Cd (up to ~300 ppm) could be found in quartz-hosted fluid and melt inclusions in hydrous peraluminous systems (pegmatites and leucogranites) for the first time ever. A distinct number of the groundwater samples used for this study show detectable concentrations of Cd. Significant amounts of Li, B, Zn, As, and Pb were also found in quartz-hosted fluid inclusions from the Huanuni tin deposit in Bolivia (hosted in peraluminous granites with $ASI \geq 1.1$) [44]. The indicative trace elements of leucogranites (Li, B, P, Mn, Zn, As, Sr, Pb, and U) are similarly detected in the ground water in Nawalparasi. The high concentration of Sr in groundwater can be based by the frequent occurrence of calcium carbonates in the soil hosting the groundwater. In study [45], the authors mentioned lithologies like rhyolite and shale (greywackes) representing a greater risk of elevated As in groundwaters.

As frequently described in the literature, low-grade metapelites are often considered as protoliths of peraluminous granites (see, e.g., [22, 46, 47]) where concentrations of As, Sb, Be, B, Ba, and Rb by a factor of 5–10 higher than their average crustal abundances (2–5 ppm) [48–50] were not unusual. The leucogranites (two-mica or muscovite-tourmaline mica) found in the Himalayas of Nepal are undoubtedly peraluminous in composition (see, e.g., [22, 51, 52]) and coherently a comparison with the findings from [41] is warranted. Most of the leucogranites analyzed in [51] are peraluminous ($ASI > 1.1$) to strongly peraluminous ($ASI \geq 1.1$). According to Ref. [53], lead represents one of the rare elements usually behaving incompatible during crustal melting. Pb can be significantly enriched in low T S-type granite melts, especially if the proportion of partial melting remains low. Moreover, muscovite, known as being a major mineral of metapelitic sources, can exhibit relatively high Pb contents. The leucogranites in the Nepal Himalayas are widely described to be of crustal origin and are derived from vapor-absent muscovite-dehydration melting of pelitic and psammitic protoliths during the Late Miocene (see, e.g., [47, 54–59]).

As described above, arsenic is primarily dissolved from micas as a major constituent of sediment hosting the groundwater in Nawalparasi. Keeping in mind that the Manaslu area is the watershed of the Nawalparasi water system, assuming the leucogranites as the original source of the arsenic is evidently warranted.

5. Conclusions

So far it was considered that As is mainly released from iron (hydr)oxides, but a major host for sorption and release of As are aluminosilicates such as clay minerals (including micas) into groundwater. The apparent decoupling of the concentrations of Fe and As and the positive correlation between concentrations of Na, K, and As in the groundwater are a significant evidence that As cannot be released from Fe minerals predominantly—so silicates represent the fundamental source of As in sediments. Clay minerals preferentially lose Na and K from their interlayers during chemical weathering and hence become enriched in immobile elements such as Fe and Al. Moreover, the trace elements detected in the groundwater samples of Nawalparasi are well known to be contained in peraluminous obsidian glasses from Peru enriched in As-B-F-P. These glasses equal the leucogranites in the Manaslu area North of Nawalparasi in their geochemistry and represent the original host rocks of arsenic.

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Conflict of interest

No conflict of interest.

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