



Towards a Sustainable Solution: Factors and Prerequisites of Improving the Kanchan Arsenic Filters Used in the Terai of Nepal: A Review

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Abstract

The issue dealing with the high arsenic (As) concentrations in ground water used as drinking water in Nepal was neglected for a long time. Whereas Bangladesh received much international attention and appropriate filters to remove As were installed, the distribution of the so-called Kanchan filters in Nepal only began in the early nineties. Arsenic itself can be readily released into ground water depending on pH, redox conditions, temperature, and solution composition. However, there is an obvious de-coupling of As and Fe concentrations in ground water. In this regard, the partial low removal efficiency of the installed Kanchan filters can tentatively be explained by the ratio of main and trace elements (particularly Fe and As), pH, flow rates, contact time with the nails, and filter maintenance. This review paper summarizes the identified geological background, origin of the As, the established mitigation option and future improvements of the filters. In that regard, the approach of building the filters with locally available material is still imperative for a country like Nepal where industrial jobs are relatively in scarce.

JEL Classification: I31, O30

Key Words: arsenic, iron, de-coupling, clay minerals, Kanchan filter, removal

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1. Introduction

Nepal's current arsenic issue concerning ground water was recognized much later than in other countries of South Asia (e.g., West Bengal, India; Bangladesh; Cambodia; Vietnam; China). Nepal did not seem to be much affected by arsenic (As) poisoning ground water hosted by the Quaternary alluvial sediments as the landlocked country is dominated by the mountain chain of the high Himalayas and only features a very narrow band of flat land (the so-called Terai, the Indo-Gangetic Plain of southern Nepal) built up by those Quaternary alluvial sediments. The actual drinking water guideline (10 µg/L) for As imposed by the World Health Organization (WHO) is exceeded in several districts (namely Nawalparasi, Bara, Parsa, Rautahat, Rupendehi, and Kapilvastu (Shrestha et al., 2014). As soon as concentration of arsenic exceeds the guideline, detrimental health effects are likely to occur. Characteristic skin lesions including pigmentation changes (melanosis, keratosis), various reproductive, neurological, respiratory, cardiovascular, gastrointestinal and diabetic effects as well as cancers of almost all inner organs are among the most prominent health impacts caused by the long-term intake of As (Smith et al., 2000; Adhikari & Ghimire, 2009; Smith & Steinmaus, 2009; Abdul et al., 2015).

Sharma (1999), in a first report ever published, mentioned arsenic contamination above toxic levels in ground water in the Terai Basin of Nepal. But it was not before 2004 when Shrestha & Shrestha (2004) mentioned that twenty-four percent of ground water samples analyzed (n = 18,635) from the Terai Basin exceeded the WHO limit of 10 µg/L. According to Panthi et al. (2006), 25,058 tube wells in the Terai region had been tested for As, of which 5,686 tube wells (22.7%) exceeded the World Health Organization (WHO) As guideline (As = 0.01 mg/L) and 1,916 tube wells (7.6%) exceeded the Nepal Interim As Standard (As = 0.05 mg/L). In a first report compiled by Nepal Red Cross Society/Environment & Public Health Organization (NRCS–ENPHO) (2002), the prevalence of arsenicosis was indicated between 1.3% and 5.1% among four independent surveys. Approximately 0.5 million people in Terai were at risk of consuming ground water with an arsenic concentration > 50 µg/L, the maximum permissible limit for Nepal (Shrestha et al., 2003). As a consequence of this alerts, in 2003, involving major stakeholders from the drinking water and sanitation sector, the National Arsenic Steering Committee (NASC) was established (Shrestha et al., 2003). Later, Nakano et al. (2014) stated correctly that the populations of the southern Indo-Gangetic plain of Nepal (Terai) struggles with the same arsenic issue as do the inhabitants of Bangladesh.

The arsenic is mainly found in hydrous iron oxides and clay minerals. The toxic agent can easily be solubilized in ground water depending on pH, redox conditions, temperature, solution composition, and climate. Source materials of As contaminating water include organic-rich or black shales, Holocene alluvial sediments with slow flushing rates, minings (mostly often gold deposits), volcanogenic sources, and also thermal springs. Two more environments can lead to high arsenic concentration in potable water: (i) closed basins in arid-to-semi-arid climates (especially in volcanogenic provinces) and (ii) strongly reducing aquifers, often composed of alluvial sediments but with low sulphate concentrations. Especially two triggers can initiate high dissolution of As (> 50 µg/L), an increase in pH above 8.5 or the onset of reductive iron dissolution. High contents of phosphate, bicarbonate,

silicate, and/or organic matter in the ground waters are other prime factors promoting arsenic solubility. The geologic and groundwater conditions triggering high arsenic concentrations are well known and help identify high-risk areas (Nordstrom, 2002; Smedley & Kinniburgh, 2002). The main source minerals of As in Nepal are various clay minerals (Guillot et al., 2015; Mueller & Hug, 2018).

To eliminate arsenic from ground water, so-called Kanchan filters (KAF) were installed in Nepal after 2004. These filters were once developed as a joint venture between Massachusetts Institute of Technology (MIT), Environment & Public Health Organization (ENPHO) and Centre for Affordable Water and Sanitation Technology (CAWST) (Ngai et al., 2005; Ngai et al., 2006; Ngai et al., 2007). However, Singh et al. (2014) reported that, unfortunately, the long-term performance of those filters in Nepal had rarely been tested. The authors from the latter study clearly stated that out of the 41 tube exposing unsafe arsenic levels, KAFs reduced arsenic concentration to the safe level for only 22 tube wells. Therefore, an ongoing project to undoubtedly determine the geological background, effectiveness and improvement of the Kanchan filters used in the Terai was initiated by co-workers from CAWST in cooperation with ENPHO and Eawag, Switzerland in 2015. This review is primarily based on the work of Mueller (2017, 2018, 2019, 2019a, 2020); Mueller & Hug (2018); Mueller et al. (2020a) because after the publications of Ngai and co-workers (Ngai et al., 2005; 2006; 2007) hardly any scientific articles were released internationally. For a detailed description of the arsenic issue in Nepal (Mueller, 2017), this review gives a concise overview of the research done since 2015 in order to elucidate the state and progress towards the much needed improvement of the removal efficiency of the Kanchan filters.

2. Geological Situation concerning Arsenic in Ground Water of the Terai

Landlocked in South Asia, Nepal is principally mountainous, with approximately 6,000 rivers and rivulets, being the resources of extensive ground water reservoirs. The immense topographic variations in Nepal are largely controlled by geology (British Geological Survey, 2001; Thakur et al., 2011). The Himalayas presenting the prominent mountain chain in the country are built up by a wide range of various rocks of metamorphic, sedimentary, and igneous in origin. The heterogeneity concerning arsenic in ground water of the foreland is mainly caused by the huge variety of the source rocks (Mueller, 2017). The Terai Plain itself represents an active foreland basin consisting of Quaternary sediments including molasse units along with gravel, sand, silt, and clay.

These sediments with a high potential for ground water resources are replenished by a high Monsoon precipitation (1,800 - 2,000 mm) and year-round snow-fed river systems. Overall, the geology of the Terai region is basically similar to the Bengal Delta Plain (BDP) and it is the continuation of Indo-Gangetic trough. A detailed description of the geological background of Terai has been described in Mueller (2017).

Districts mostly affected by the arsenic issue in southern Nepal include Nawalparasi (western region), Rautahat and Bara (central region) and Bardia (mid-western region). The fine alluvial aquifers of Nawalparasi district are among the most severely As contaminated in the Terai region. This region is highly contaminated with As, unfortunately, and predominantly determined as As(III), the more toxic form of arsenic. In the sediments of

Nawalparasi district, clays contain high amounts of iron and aluminum. Arsenic is abundantly incorporated in finer particles such as clay minerals as well. Generally, higher concentrations of As were found mostly in the fine-grained clay sediments (black and yellow) and to a lesser extent in coarse-grained sediments (Yadav et al., 2015). Arsenic occurs typically in oxyanionic forms in the aqueous environment. Hydro-geochemical data for ground water of the Terai Alluvial Plain (TAP) aquifers prove a predominantly reducing character, with high HCO_3^- , low SO_4^{2-} , and NO_3^- concentrations.

3. Mechanism of Arsenic Release to Groundwater

The main mechanisms of As release to ground water so far widely discussed and accepted, and was worked out by Nickson et al. (2000) in reference to the conditions in Bangladesh. The authors released the assertion that As in the ground water originates from reductive dissolution of As-rich Fe-oxyhydroxides existing as a dispersed phase (e.g., as surface coatings) on sedimentary grains. Furthermore, this reduction is reinforced by microbial degradation of sedimentary organic matter. There is a widespread consensus that the initial dominant process is the fixation of aqueous As by sorption onto Fe-, Mn-oxide or clay surfaces during high-redox medium-pH conditions (e.g., about 5.5 - 6.5). As a consequence of a rise in pH ($\text{pH} > 6.5$, leading to an increased solubility of arsenic acid) and a negative Eh, the desorptive release of arsenic occurs from sediment into ground water (Stanger, 2005). Arsenic is generally found as the reduced trivalent form [As(III)] in ground water whereas the oxidized pentavalent form [As(V)] is present in surface water. Unfortunately, arsenite [As(III)] as well as inorganic arsenic species are more toxic for living organisms than arsenate [As(V)] or organic forms of arsenic. Regarding ground water conditions in the Terai, Bhattacharya et al. (2003) reported mostly near-neutral to alkaline pH range of 6.1 - 8.0. Redox potential (Eh) levels between -0.20 to -0.11 V advocate fairly reduced condition in the aquifers. The ground water is described as predominantly of Ca-Mg-Na- HCO_3^- -type with HCO_3^- as the principal anion and low levels of Cl^- and SO_4^{2-} . Panthi et al. (2006) and Diwakar et al. (2015) stated that the low redox potential of tube well waters support the theory of reductive desorption, as some particular various redox sensitive elements (e.g., Fe^{2+} , As(III), NH_3) are very abundant in the aquifers of the Terai. The role of clay minerals as carriers of As is occasionally mentioned, as climatic variations in ground water chemistry can serve to distinguish the contributions of the two sources in question (Fe-oxyhydroxides vs. clay minerals), and such variations are particularly pronounced in headwater areas of the Ganges floodplain immediately adjacent to the Himalayan foothills (e.g., the Terai of Nepal) (Brikowski et al., 2014; Guillot et al., 2015). In the district of Nawalparasi most tube wells are drilled 20m below ground level in order to exploit permanently saturated thin sandy layers. At this depth solid phase As(III) and lower valency As-sulphide species are recognized to be the dominant species, while poorly crystalline Fe(III) and Fe oxides are largely absent (Gurung et al., 2005).

In opposition to the above mentioned theory of microbially mediated reductive dissolution of As-rich Fe-oxyhydroxides, the role and influence of As bound and released from phyllosilicates (e.g., micas) was hardly ever discussed (e.g., Stanger, 2005; Chakraborty et al., 2007; Charlet et al., 2005, 2011; Brikowski et al., 2014; Guillot et al., 2015; Yadav et al., 2015; Verma et al., 2016; Uddin, 2017). In this regard, a striking feature of the ground

water exploited in the Terai (as compared to the concentration of Fe and As in ground water from Bangladesh or Vietnam) is the very low average molar ratio Fe/As. In Vietnam the average molar ratio of Fe/As fluctuates between 60 and 68 (Berg et al., 2008) whereas the ratio sums up to 14.88 or 90.49 in Bangladesh (Ahmed et al., 2010; Rahman et al., 2015). In contrast to these numbers, the average molar Fe/As ratio for the 35 samples analyzed adds up to 9.4 (post-Monsoon) and to 6.0 (pre-Monsoon) in Nepal (Mueller & Hug, 2018). Moreover, there is a distinct decoupling of the concentration between Fe and As in the ground water in Nawalparasi district. This decoupling obviously warrants the necessity to rethink the original mineralogical hosts of arsenic and to include the absorption, incorporation and release of As from clay minerals. Diagrams presented by Mueller & Hug (2018) and Mueller (2019) unambiguously depict the correlation between As and the lithophile elements: Na, K, Ca, Mn, Li, B, Sr and Mo. Na, Mg, K and Sr (a replacement of Na and K) can easily be dissociated from interlayers of phyllosilicates [general formula: $X_2 Y_{4-6} Z_8 O_{20} (OH,F)_4$, whereas X: cation in 12-fold coordination (K, Na, Ca); Y: cation in octahedral coordination (Mg, Fe, Al); Z: cation in tetrahedral coordination (Si, Al)]. Na, K and Sr can be derived as well from alkali feldspars. Li, B and Mo are typical trace elements found in various forms of mica. Li and B represent major components of tourmaline. Stueben et al. (2003) reported about tourmaline-containing aquifers enriched in As in West Bengal, India. Mueller (2018) related the typical trace element composition of ground water in the Terai to the origin of the soil minerals being transported from Tertiary leucogranites (rich in B) in the High Himalayas, exhibiting a very low Fe-concentration. To convincingly describe the overall situation in the Terai regarding the mineralogical host of As, Zweifel (2018) hardly detected any Fe(III)hydr(oxides) by X-ray analysis in a drill core from the Nawalparasi district, as opposed to abundant clay minerals in the same drill core. The overall Fe concentration in the ground water as reported by Mueller & Hug (2018) is low and precludes the formation of a considerable amount of Fe(III)hydr(oxides).

In addition to the considerations above, Guillot et al. (2015) reported about an apparent correlation concerning late Quaternary (0.5 - 1.0 million years before present) climate conditions and the concentration of arsenic in the alluvial sediments with As to be dominantly accumulated in sediments deposited during more arid periods. In humid period the lower arsenic contents in sediments can be interpreted by leaching from sandy and silty sediments due to intensive Monsoon rainfall. But according to Mueller & Hug (2018), Mueller (2019), and Mueller (2019a), it was not possible to find a prominent difference concerning As in ground water between climatic seasons. In contrast, the concentration of As in post-Monsoon seems to be potentially higher than in pre-Monsoon season. Yet the huge variation of As concentrations in ground water mirror the heterogeneous sediment composition in the district of Nawalparasi on a municipality-scale as well as the changes over time in redox conditions, pH and temperature. Guillot et al. (2015) already noted that As concentrates explicitly in the clay-dominated sediments in the Terai and is usually correlated with specific elements (Al, K and C) and also mentioned a rather good correlation between K_2O and arsenic content in finer clay fractions, suggesting that biotite contributed to the fixation of this element. The reductive dissolution of As from micas is, therefore, the main mechanism of the elevated contents of As in the ground water in Nawalparasi district.

4. Kanchan Filters and their Constraints

The aforementioned low concentration of Fe, the low molar ratio of Fe/As as well a general short residence time of the ground water within the nail bed of the filters, high pH and high concentrations of As, Na, B, Mo and other trace elements (Mueller & Hug, 2018) clearly limit the performance of the filters. A dry nail bed instead of a permanent immersion of the nails in ground water is another adverse effect concerning the removal efficiency of the filters used in Nawalparasi district. A wet nail bed secures the formation of Fe(III)hydr(oxides), black mixed Fe(II,III)-phase solids and finally to conversion of the latter to magnetite (Fe_3O_4) with integration of As(V) exhibiting a stronger adsorption affinity for As(V) than for As(III) (Wenk et al., 2014). X-ray examinations of nails from some of the monitored filters exhibit siderite (FeCO_3) on their surface (Zweifel, 2018). As this mineral is precipitated in reducing (oxygen-free) environments and, therefore, contains Fe(II), the indication is twofold: (i) the oxidation process in order to enhance rusting of the nails is incomplete and (ii) siderite inhibits the adsorption of As on the nail surface or co-precipitating with Fe(III)hydr(oxides) (Wenk et al., 2014; Guo et al., 2007). Analyses of the X-ray data undoubtedly reveal that siderite is dominantly formed on nails in contact with ground water with a high iron concentration. Therefore, it is vital to enhance contacting time within the nail bed in order to prevent the precipitation of siderite (Mueller, 2019).

5. Factors Influencing the Removal Efficiency of the Kanchan Filters

Since the first field campaign in autumn 2015 at least 30 filters were inspected regularly and ground water samples were collected in pre-Monsoon and post-Monsoon seasons. The removal efficiency from those filters vary in a wide range from 5.81 % to 97.1 % (for details see Mueller, 2020; Mueller et al., 2020a).

There are several reasons as outlined below found to influence the removal efficiency as well as to explain the discrepancy concerning the removal efficiency varying from 5.81 % to 97.1 %:

- (i) Partially complete and dry nail bed or an irregular surface of the bed (promoting channels were ground water which can easily flow without contact to the nails).
- (ii) Due to incomplete oxidation of the ground water flowing through the nail bed, siderite (FeCO_3) could be formed on the nail surfaces leading to a diminished adsorption of As. Unfortunately, siderite is mainly formed on nails when the iron concentration of the ground water is higher than usual (Fe up to 5.8 mg/l, the majority of Fe concentration in ground water fluctuating between 2 and 3 mg/l).
- (iii) Prolonged contact time (up to 30 min) between the nails and the ground water ensured sufficient removal of high concentrations of As despite low concentrations of Fe.
- (iv) Long-term (year-long) use of the lower sand layer leading to a reduced capacity of the fine grained sand (grain size < 2 mm) to remove exfoliated particles with adsorbed As from the nails above.

6. Results of the First Step of Improvements

As a consequence of the above mentioned effect in 2018 and 2019, a first sand layer just above the nail bed and below the brick layer was set for 30 filters. This sand bed should lower the flow through rate as well as to increase the contact time between nails and ground water. The sand itself was separated from the nails by a cloth (cotton-polyester blend) in

order to facilitate maintenance. This sand bed should prevent the nails from drying as well while keeping the nails in place impeding the formation of irregularities within the nail bed. Sampling of all the adapted filters in spring 2019 clearly indicated that most of the filters equipped with an upper sand layer exhibited an improved performance. Best removal rates were achieved when the nail bed was kept wet permanently but not immersed in water. This way, the nails can be seen to be oxygenated best. Generally, the lower sand bed is hardly ever replaced as per testimonies by queried residents. As a consequence, the performance is still not as high as expected.

7. Summary and Future Perspective

A low performance of some of the filters is usually caused by negligence (e.g., displacement of the nails in the nail bed and pouring water into the filter too quickly causing an uneven nail bed) and poor maintenance (e.g., omitting to change the nails and the fine sand in the lower sand layer regularly). This sand will be returned to local concrete manufacturers it was originally bought from in order to recycle it properly and not to bury it as it contains an elevated concentration of As. Arsenic will be immobilized through this way. Uncontrolled deposition of this As bearing sand has to be strictly avoided as surface water could dissolve the toxic elements again whereas once the sand is buried and covered with soil, As could be released to ground water resurgently.

In order to extend the contact time between nails and ground water, placing a tap at the outlet or raising the outlet of the plastic bucket above the level of the nail bed will be installed in order to regulate the outflow. Through this way, the nails should be kept wet and oxide conditions will prevail in the nail bed. Nails and sand of both sand layers have to be replaced on a regular basis.

Lastly, as the most important action, regular and repeated proper instruction courses for the users of the Kanchan filters have to be established. So far, instructions for the users have been somewhat neglected, leading to a poor understanding of the procedure of concerned residents in the Terai. According to resident accounts, often women are responsible for installation and maintenance of the filters. Proper instructions leading to a deeper understanding concerning the use of the filters and adverse health effects of a prolonged intake of As from drinking water supports the empowerment of women in a third world country as well.

After this in-depth examination of a representative number of Kanchan filters in Nawalparasi district, the change in design of these filters seems obvious. With a tap installed at the outlet of the filter the nail bed will be kept wet at all times. Both sand layers will have to be changed at regular intervals as well.

The approach of building the filters with locally available material is still imperative to improve the socio-economic condition of many families in Nepal. This, however, hampers the purchase of industrially-produced filters mainly based on cationic exchanger. Furthermore, such filters cannot be manufactured in Nepal and therefore the issues arising in terms of waste disposal cannot be solved in Nepal actually. Sand and nails acquired locally can be used in concrete production though by regional manufacturers which has economic implication particularly in terms of employment generation in a country where the size of unemployed population is very high.

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