

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 concerning the high arsenic concentrations in ground water used as drinking water in the lowland of Nepal was neglected for a long time. Whereas mainly Bangladesh received much international attention and support to develop appropriate filters to remove As, in Nepal the installation of the so called Kanchan filters only begun in the early nineties when adverse health effects were already observed. Arsenic itself can be readily released into ground water depending on pH, redox conditions, temperature, and solution composition. Moreover, the widely spread hypothesis asserts that reductive dissolution of Fe-bearing minerals releases As-oxyanions. However, there is an obvious de-coupling of As and Fe concentrations in ground water resulting in loss of correlation concerning these two elements. Beyond that, As is positively correlated with Na and K, the molar ration of Fe/As in the ground water is very low and in conclusion therefore clay minerals (containing a minor amount of Fe in their structure) have to be regarded as a substantial host of As. 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 summarizes the identified geological background, origin of the As, the established mitigation option and future improvements of the filters.

Keywords: Arsenic, iron, de-coupling, clay minerals, Kanchan filter, removal

1. INTRODUCTION

Unfortunately, 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 As poisoning ground water hosted by 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, Rupandehi, and Kapilivastu (Shrestha et al., 2014)). As soon 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 and Ghimire, 2009; Smith and 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. Later, Nakano et al. (2014) stated correctly that the population of the southern Indo-Gangetic Plain of Nepal (Terai) struggle with the same arsenic issue as do the inhabitants of Bangladesh, for example. But it was not before 2004 when Shrestha and 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 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 WHO (World Health Organization) 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 NRCS–

ENPHO (Nepal Red Cross Society/Environment & Public Health Organization), 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 $> 50 \mu\text{g/L}$, 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).

The element arsenic is mainly found in hydrous iron oxides and clay minerals. The toxic agent can easily be solubilized in groundwater depending on pH, redox conditions, temperature, solution composition and climate. Source material of As contaminating water include: organic-rich or black shales, Holocene alluvial sediments with slow flushing rates, mining (most 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 ($> 50 \mu\text{g/L}$): 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 and Kinniburgh, 2002).

To eliminate arsenic from ground water so called Kanchan filters (KAF) were installed in Nepal. 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) (see Ngai et al. 2005, Ngai et al. 2006; Ngai et al. 2007). However, Sing 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, 2015 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. This review is primarily based on the work of Mueller (2017); Mueller, 2018; Mueller and Hug, 2018; Mueller (2019); Mueller (2019a); Mueller (2020); Mueller et al. (2020a) as since the publications of Ngai and co-workers (see Ngai et al. 2005, Ngai et al. 2006; Ngai et al. 2007) hardly any scientific articles were released internationally. For a detailed description of the arsenic issue in Nepal see Mueller (2017).

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 (BGS, 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 (see 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. Over all, the geology of the Terai region is basically similar to the Bengal Delta Plain (BDP) and it is the continuation of Indo-Gangetic trough. For a detailed description of the geological background see (Mueller 2017).

Districts mostly affected by the arsenic issue in Nepal include Nawalparasi (Western Region), Rautahat and Bara (Central Region) and Bardia (Midwestern Region). The fine alluvial aquifers of Nawalparasi are among of 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 as well abundantly incorporated in finer particles like clay minerals. Generally, higher concentrations of As were found mostly in the fine-grained clay sediments (black and yellow)

and to a lesser extent the in coarse-grained sediments (Yadav et al., 2015). Arsenic occurs typically in oxyanionic forms in the aqueous environment. Hydrogeochemical data for groundwater of the TAP (Terai Alluvial Plain) 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 so far widely discussed and accepted was worked out by Nickson et al. (2000) in reference to the conditions in Bangladesh. The authors released the assertion that As in the groundwater originates from reductive dissolution of As-rich Fe-oxyhydroxides existing as a dispersed phase (e.g. as a coating) 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 (i.e. about 5.5-6.5). As a consequence of a rise in pH ($\text{pH} > 6.5$) 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 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) report about a mostly near-neutral to alkaline pH range of 6.1-8. Redox potential (Eh) levels between -0.20 to -0.11 V advocate fairly reduced condition in the aquifers. The groundwater 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) as well as Diwakar et al. (2015) state that the low redox potential of tube well waters support the theory of reductive desorption, as some particular various redox sensitive elements (i.e. 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 groundwater 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) (see e. g. Brikowski et al. 2014; Guillot et al., 2015). In the district of Nawalparasi most tube wells are drilled 20 m below ground level in order to exploit permanently saturated thin sandy layers. At this depths 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 (see e. g. Stanger, 2005; Chakraborty et al., 2007; Charlet et al., 2005; Charlet et al., 2011; Brikowski et al., 2014; Guillot et al., 2015; Yadav et al., 2015; Uddin, 2017; Verma et al., 2016). 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 ration 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 this 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 and Hug, 2018). Moreover, there is a distinct decoupling (lack of correlation) of the concentration between Fe and As in the ground water in Nawalparasi. This decoupling obviously warrants the necessity to rethink the original mineralic hosts of arsenic and to include the absorption, incorporation and release of As from clay minerals. Diagrams presented in Mueller and Hug (2018) and Mueller (2019) unambiguously depict the correlation between 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 dissociates from interlayers of phyllosilicates, Na, K and Sr as well from alkali feldspars. Li, B and Mo are typical trace elements found in various forms of micas, Li and B represent major components of tourmaline (general formula: $(\text{Ca}, \text{K}, \text{Na})(\text{Al}, \text{Fe}, \text{Li}, \text{Mg}, \text{Mn})_3(\text{Al}, \text{Cr}, \text{Fe}, \text{V})_6(\text{BO}_3)_3(\text{Si}, \text{Al}, \text{B})_3\text{O}_{18}(\text{OH}, \text{F})_3$). Stueben et al. (2003) report about tourmaline-

containing aquifers enriched in As in West Bengal, India. Mueller (2018) relates 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. To convincingly complete the overall situation in the Terai regarding the mineralic host of As Zweifel (2018) hardly detected any Fe(III)hydr(oxides) by X-ray analysis in a drill core from the district Nawalparasi - as opposed to abundant clay minerals in the same drill core.

In addition to the considerations above Guillot et al. (2015) reported about an apparent correlation concerning late Quaternary (0.5 - 1.0 million years b.p.) 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 periods 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 and Hug, (2018); Mueller (2019); 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 K₂O and arsenic content in finer clay fractions, suggesting that biotite contributed to the fixation of this element.

4. KANCHAN FILTERS AND THEIR CONSTRAINTS

The aforementioned low concentration of Fe, the low molar ratio of Fe/As as well as 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 (see 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. 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 (Fe₃O₄) 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 (FeCO₃) on their surface. As this mineral is precipitated in reducing environments and therefore contains Fe(II) the indication is two fold: (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). Analysis 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 contact time within the nail bed in order to prevent the precipitation of siderite (see also 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 found to influence the removal efficiency:

- Partially complete and dry nail bed or an irregular surface of the nail bed (promoting channels where ground water can easily flow without contact to the nails).
- Due to incomplete oxidation of the ground water flowing through the nail bed, siderite (FeCO₃) 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 high.
- 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.

- 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 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 seen to be oxygenated best. As usually the lower sand bed is hardly ever replaced according testimonies by queried residents the performance is still not as high as expected.

7. SUMMARY AND FUTURE PERSPECTIVE

A low performance of some of the filters are usually caused by neglectation (e. g. displacement of the nails in the nail bed after time, pouring water into the filter to speedy 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).

In order to extend the contact time between nails and ground water, a tap at the outlet or raising the the outlet of the plastic bucket to above the level of the nail bed will be installed in order to regulate the outflow. This way, the nails should be kept wet and oxic 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 more or less neglected, leading to a poor understanding of the concerned residents in the Terai. According statements of residents often women are responsible for installation and maintenance of the filters. Proper instructions leading to a deeper understanding concerning 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.

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