Arsenic in groundwater in the southern lowlands of Nepal and its mitigation options: A review

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<th>Journal:</th>
<th>Environmental Reviews</th>
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<td>Manuscript ID</td>
<td>er-2016-0068.R2</td>
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<tr>
<td>Manuscript Type:</td>
<td>Review</td>
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<tr>
<td>Date Submitted by the Author:</td>
<td>15-Dec-2016</td>
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<tr>
<td>Complete List of Authors:</td>
<td>Mueller, Barbara; University of Basel,</td>
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<td>Keyword:</td>
<td>arsenic, arsenic contamination, release of arsenic to the groundwater, removal of arsenic</td>
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Arsenic in groundwater in the southern lowlands of Nepal and its mitigation options: A review

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Abstract: As in several other countries of south east Asia (namely Bangladesh, India, Myanmar, China, Vietnam, Cambodia) arsenic concentrations in the groundwater of the lowlands of Nepal (the so called Terai) can reach concentrations which are unsafe to humans using the groundwater as drinking water. Whereas Bangladesh has received much international attention concerning the arsenic crisis, Nepal was more or less neglected. The first report about arsenic contamination of the groundwater above toxic levels in Nepal was published in 1999. Twenty-four percent of samples analyzed (n = 18,635) from the Terai Basin exceeded the WHO guideline of 10 µg/L. Since the first overall survey from 2001, only sporadic information on the situation has been published. The geological and geochemical conditions favour the release of the contaminant as arsenic can be easily solubilized in groundwaters depending on pH, redox conditions, temperature, and solution composition. The thin alluvial aquifers of the Terai are some of the most severely As contaminated. These sediments constituting a huge proportion of the Terai aquifers are derived from two main sources, (i) sediments deposited by large rivers that erode the upper Himalayan crystalline rocks, (ii) weathered metasediments carried by smaller rivers originating in the Siwalik forehills. The generally low redox potential, low SO$_4^{2-}$, high DOC, PO$_4^{3-}$ and HCO$_3^-$ concentrations in groundwater signify ongoing microbial mediated redox processes favoring As mobilization in the aquifer. Other geochemical processes, e.g., Fe-oxyhydroxides reduction and carbonate dissolution are also responsible for high As occurrence in groundwaters. Originally, Gagri filters (two filter system with chemical powder) and later iron-assisted bio-sand filters were commonly used to remove arsenic and iron from well water in Nepal – these two options were believed to be the best treatment option at household levels. This review will focus on the description of the overall situation (geogenic issues, occurrence of arsenic in the sediments of the Terai, mechanisms of the release of arsenic to the groundwater, mitigation options).

Keywords: arsenic; arsenic contamination; release of arsenic to the groundwater; removal of arsenic; mitigation
Introduction

In the groundwaters of several countries of south east Asia (namely Bangladesh, India, Nepal, Myanmar, China, Vietnam, Cambodia), arsenic can naturally reach concentrations that are hazardous to human health if geological and geochemical conditions favour the release of this contaminant. The World Health Organisation (WHO) has imposed a drinking water guideline with a value of 10 µg/L for arsenic. When this value is exceeded, health risks are likely to occur. Excess uptake of arsenic causes a range of adverse health effects like characteristic skin lesions including pigmentation changes, mainly on the upper chest, arms and legs, and keratoses of the palms of the hands and soles of the feet and as the most severe effect, cancer (Smith et al., 2000; Adhikari and Ghimire, 2009).

Arsenic itself is not found in high abundance in the Earth's continental crust; it is less abundant than several of the “rare-earth” elements. Unlike the rare-earth elements, however, arsenic is commonly concentrated in sulphide-bearing mineral deposits, and it has a strong affinity for pyrite, one of the more ubiquitous minerals in the Earth's crust. It is also concentrated in hydrous iron oxides and clay minerals. Arsenic can be easily solubilized in groundwaters depending on pH, redox conditions, temperature, and solution composition. Many geothermal waters contain high concentrations of arsenic, too. Natural arsenic in groundwater at concentrations above the drinking water guideline of 10 µg/L is not uncommon. A small number of source materials are now recognized as significant contributors to arsenic in water supplies: 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. Two other environments can lead to high arsenic: (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. Young sediments in low-lying regions of low hydraulic gradient are characteristic of many arsenic-rich aquifers. Ordinary sediments containing 1 to 20 mg/kg (near crustal abundance) of arsenic can give rise to high dissolved arsenic (> 50 µg/L) if initiated by one or both of two possible “triggers” - an increase in pH above 8.5 or the onset of reductive iron dissolution. Other important factors promoting arsenic solubility are high concentrations of phosphate, bicarbonate, silicate, and/or organic matter in the ground waters. These solutes can decrease or prevent the adsorption of arsenate and arsenite ions onto fine-grained clays and especially iron oxides. Arsenite tends to adsorb less strongly than arsenate often causing arsenite to be present at higher concentrations. The geologic and groundwater conditions that promote high arsenic concentrations are now quite well known and help identify high-risk areas (Nordstrom, 2002, Smedley and Kinniburgh, 2002). The water table within the IGB (Indo-Gangetic Basin, including the Terai) alluvial aquifer is typically shallow (<5 m below ground level). Abstraction of groundwater can also influence arsenic flux: It can flush aqueous
arsenic from the aquifer; irrigation pumping protects deeper groundwater in some instances, by
creating a hydraulic barrier, but it seems that high-capacity deep pumping may draw arsenic down to
levels in the Bengal aquifer system which are otherwise of good quality (MacDonald et al., 2016)
Whereas Bangladesh has received much international attention concerning the arsenic crisis (e. g.
Hug et al., 2011 and references therein), Nepal was more or less neglected, though the population of
the southern lowlands of Nepal (the so called Terai, the Indo-Gangetic Plain of southern Nepal) face
the same arsenic contamination of the groundwater (Nakano et al., 2014). The study of arsenic
concentrations in the groundwater in Nepal began only after the severity of the arsenic contamination
problem in the Bengal delta was recognized in 1998. The first report of arsenic contamination in
groundwater above toxic levels in Nepal was made from the Terai Basin (Sharma 1999). Twenty-
four percent of samples analyzed (n = 18,635) from the Terai Basin exceeded the WHO limit of 10
µg/L (Shrestha and Shrestha 2004). Since the first overall survey conducted by WHO (2001), only
sporadic information on the situation has been published. Available documents later indicated that
the region of As contamination extends even into the Terai region and that 24 districts in Nepal,
including all 20 Terai districts and four hill districts, exhibit arsenic contamination (Bhattacharya et
al. 2003; Neku and Tandukar 2003; Shrestha et al. 2003; FAO, 2004; Tandukar et al. 2005; Panthi et
al., 2006; Maharjan et al., 2006; Pokhrel et al. 2009; Emerman et al., 2010; Thakur et al., 2011).
Although the Terai constitutes less than 20 % of the Nepal’s surface, it contains over half of the total
arable land and is home to about 50 % of the total Nepalese population, i.e. 30 millions of
inhabitants. Groundwater is the main source of water for drinking and irrigation in the Terai area.
Over 90 % of the Terai population draws groundwater from tube wells for drinking, household use,
and irrigation (Guillot 2015).
According some publications, 25,058 tubewells in the Terai region have been tested for As, of
which 5,686 tubewells (22.7%) exceed the WHO (World Health Organization) As guideline (As =
0.01 mg/L) and 1,916 tubewells (7.6%) exceed the Nepal Interim As Standard (As = 0.05 mg/L)
(Panthi et al. 2006). It is estimated that there are perhaps 200,000 tubewells in the Terai region and
that 3.5 million Nepalese have no access to drinking water that does not exceed the WHO As
guideline (Mahat and Shrestha 2008; Mahat and Kharel, 2009; Pokhrel et al. 2009). In the most
recent report from NASC-NRCS (National Arsenic Steering Committee/National Red Cross Society,
2011) the total database covers 1.1 million wells tested between the years 2003 to 2008.
Approximately 1.73% showed values above the Nepal drinking water standard of 50 ppb, while
approximately 5.37% of tube wells contain 11-50 ppb of arsenic concentration. The percentage of all
tube wells exceeding 50 ppb varies from 0.05% of the wells in the district of Jhapa to 11.69% in the
district of Nawalparasi.
The most severe arsenic contamination is prevalent in several districts of the Terai namely
Nawalparasi, Bara, Parsa, Rautahat, Rupandehi, and Kapalivastu (Shrestha et al., 2014). Maharjan et al. (2005) reported that 29% of more than 20,000 tube wells had arsenic concentrations exceeding the World Health Organization (WHO) guideline (10 µg/L), that the prevalence of arsenicosis varied between 1.3% and 5.1% (average of 2.6 %; see NRCS–ENPHO (Nepal Red Cross Society/Environment & Public Health Organization), 2002; Yadav et. al., 2011) among four independent surveys, and that approximately 0.5 million people in Terai were at risk of consuming water with an arsenic concentration > 50 µg/L, the maximum permissible limit for Nepal (Shrestha et al. 2003). It was found that overall prevalence of arsenicosis among the subjects ≥ 15 years old was 6.9%, which was comparable to those found by the same examiner in arsenic-contaminated areas in Bangladesh, and that males had prevalence a twice as high as females, which could not be explained by the difference in the exposure level Maharjan et al. (2005). These reports have alerted the decision makers of the government as well as non-governmental agencies involved in controlling water supply. As a consequence, in 2003 the National Arsenic Steering Committee (NASC) was formed, involving major stakeholders from the drinking water and sanitation sector (Shrestha et al. 2003). The NASC worked in collaboration with the Environment Public Health Organization (ENPHO) to perform testing on 18,635 tubewells in 20 Terai districts, under a program called the “State of Arsenic in Nepal 2003.” All the data collected revealed that concentration of As varied both spatially and seasonally, suggesting the possibility of spatial variation due to geospatial conditions such as latitude, longitude and depth of tubewell. The temporal distribution of As showed seasonal dependence with lower concentration in winter and higher in summer (Yadav et al., 2012).

Geological situation of the Terai region

Nepal is a landlocked country in South Asia, located between Tibet to the north and India to the south, east and west. With a total land area of 147,181 km², the country is characterized by a diverse, rugged and undulating topography, geology and in general by a cold climate. Nepal is predominantly mountainous, with elevations ranging from 64 m above sea level to 8,848 m at the peak of the world’s highest mountain, Sagarmatha (Everest), within a span of 200 km. Approximately 6,000 rivers and rivulets, with a total drainage area of about 194,471 km², flow through Nepal, whereof 76% of this drainage area is contained within Nepal. The topographic variations in Nepal are largely controlled by geology (BGS, 2001; Thakur et al., 2011). The geology of Nepal marks the transition where the Southern Gondwanaland collided with the Northern Eurasianland lifting the sediments of the then Tethys Sea to form the Himalayas. As a consequence, the southern and northern parts of Nepal differ widely in their formations. 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 formed by the then east–west flowing rivers can be find within confined space (Yadav et al., 2015).

The prominent mountain chain in Nepal – the Himalayas – is built up by four major Himalayan tectonic units: (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) and the Quaternary foreland basin. These units span a wide range of various rocks being metamorphic, sedimentary, and igneous in origin, making it possible for their differential erosion to account for some of the groundwater arsenic heterogeneity we see in the foreland and delta (i.e. Gurung et al. 2005; Shah 2008; van Geen et al. 2008; Guillot et al., 2015). The Terai Plain is an active foreland basin consisting of Quaternary sediments that include molasse units along with gravel, sand, silt, and clay. Most of the rivers in the Terai flow from north to south. All major rivers originate in the high Himalayas whereas minor rivers also emanate from the nearby Siwalik Hills, and therefore deposit sediments in the form of a fan along the flank of the Terai basin. Fine sediments and organic material are deposited in inter-fan lowlands, in wetlands and swamps (Sharma 1995). The Siwalik lithofacies are strongly diachronous, and further complicated by a variable addition of micaceous sands and arkose, locally derived from southward-draining tributaries from the emerging Himalayas. The most typical Siwalik lithologies are conglomerates, ‘salt and pepper’ micaceous sandstones, blue-grey siltstones, clay-stones, red (iron-rich) shales, and minor lignite. Potential adsorption substrates and co-precipitation hosts for arsenic are common throughout the finer-grained Siwalik facies, as iron mineralization, as sulphides, or as clays. In Nepal, the groundwater arsenic is of relatively local provenance, being derived directly from eroded Siwaliks (Stanger 2005).

High monsoon precipitation (1,800–2,000 mm) and year-round snow-fed river systems recharge the Terai sediments, giving them a high potential for groundwater resources. Shallow aquifers (<50 m) are generally unconfined or semi-confined, whereas the deep aquifers (>50 m) are mostly confined by impermeable clay layers. The aquifer system is highly sensitive to precipitation (Gurung et al., 2005).

The geology of the Terai region of Nepal itself is on the whole comparable to the Bengal Delta Plain (BDP) and it is the continuation of Indo-gangetic trough. The Terai plain covered by recent and older alluvium comprises channel sand and gravel deposits and outwash deposits. These fluviatile deposits are cross-bedded, eroded, reworked, and redeposited because of regular shifting of stream channels. Geomorphologically, the Terai plain is divided into two zones: the Bhabar zone in the
north and the main Terai zone in the south. They have diverse hydrogeological characteristics and are separated by a line of natural springs. The Bhabar zone is a narrow extension of a recent alluvial and colluvial fan deposit at the bottom of Siwalik Hills (Kansakar 2004). It consists of thick deposits of gravel, pebble, and boulder mixed with sand and silt. Sediments in the main Terai were deposited by braided rivers, which regularly changed their course. As a result, clay, silt, sand, and gravel deposits of varying thicknesses occur interlayered with each other. The Terai plain has a multiple aquifer system (Yadav et al., 2011).

So far the most intensive studied Terai province concerning local geology and arsenic contaminated groundwater is Nawalparasi. This district lies in the Terai plain as the continuation of Indo-Gangetic plain (Fig. 1). It has gentle slope toward south from an elevation of 200–300 m in the north to as low as 63 m in the south near Indian border from the mean sea level (Upreti 2001). From Indian border, Nawalparasi district extends northward across Narayani River (one of the major river of Nepal) alluvium then across the low gradient fan of locally derived alluvium and finally into the Himalayan foothill (also known as Churia hills) (Hagen 1969). The lithology of the Terai sedimentary basin belongs to Holocene alluvium that includes the present day alluvial deposits, channel sand and gravel deposits and outwash deposits (Yadav et al., 2014). The district has three distinct hydrogeological zones: (1) the Siwalik Hills, (2) the Bhabar recharge zone and (3) the Terai plain unconsolidated Holocene floodplain sediments. The northern part of the district is bounded by the steeply sloped Siwalik Hills which are composed of sedimentary rocks such as sandstone, siltstone, mudstone, shale, and conglomerates. Immediately south lies the Bhabar zone, which is composed of unconsolidated sediments that are porous, coarse such as gravel, cobbles and boulder material, thereby making the Bhabar zone highly permeable (Kansakar, 2004; Shrestha, 2007). A major river, the Narayani/Gandaki, which descends from the Higher Himalaya, flows along the eastern boundary of the Nawalparasi district and has had a major influence on the underlying unconsolidated Holocene fluvial deposits that comprise the floodplain aquifer system. Unlike other regions of Terai, where finer of sediments typically increase toward the south, fines predominate in the north and sand and gravels are found near the Nepal–India border (Shrestha et al., 2004). In the areas with fine-grained sediments, elevated concentrations of As are typically recorded (Brikowski et al., 2004, 2014; Diwakar et al. 2015).

Small ephemeral rivers originating from the Siwalik frontal mountains disappear upon entering into the Indo-Gangetic plain and reappear again in Nawalparasi. Hence, small natural ponds and river meanderings were observed as characteristic geomorphic features of the area. Therefore, close to the frontal mountain chain, the Indo-Gangetic plain consists of boulder- to gravel-sized sediments, while soils further south consists dominantly of fine-grained sediments. Guillot et al. (2015) report about the lithology of sledge core samples from the five drill holes showing various coarse (millimetric) to
fine-grained (micrometric) sediments in the Narayani basin. They distinguished light-grey to dark
grey sands; grey, greenish-grey to brown-grey and yellow–brown silts; and light-grey to black-grey,
 yellow–brown and black clay with occasional gravel layers. Macroscopic observations showed that
on average, the drilled sediments are composed of 33 % of silts; 30 % of grey to black clays, 27 % of
brown clay, 9 % of fine-grained silt and sand and less than 1 % of calcrete. Sands, silt and clay
sediments often contained micas that were occasionally massive to laminated, bioturbated, and/or
also containing roots and plant debris. Binocular observations show that the detrital minerals in the
silt fraction are dominated by quartz, biotite, muscovite, K-feldspar, calcite and dolomite as major
phases and garnet, zircon, and monazite as heavy minerals. In the region of provenance of the
Narayani basin, the Tethys Himalaya includes 10 km of various metasedimentary rocks (limestones,
calc-schists, shales, quartzites) ranging from Cambrian to Jurassic. There is also the Manaslu
leucogranite emplaced within the Tethyan rocks. The Higher Himalayan Crystallines are a
metamorphic stack, including from base to top paragneisses (metapelites and metapsammites);
gneisses with calc-silicate minerals (diopside and amphibole) and orthogneisses representing
metamorphosed Lower Paleozoic granites. The Lesser Himalaya consists of mostly unfossiliferous
metasediments and some dolomitic meta-carbonates alternating with dominant black schists,
aluminium rich schists and quartzites. Amphibolites occur in both of these groupings. The Siwaliks
represent the Cenozoic foreland basin of the Himalayan belt with local thickness of 6 km in Nepal.
They are divided into three units having a typical coarsening-upward succession. The lower unit
consists of fluvial channel sandstones alternating with calcareous paleosols; the middle unit consists
of very thick channel sandstones with minor paleosols, and the upper unit mainly hosts
conglomerates of gravelly braided river deposits (after Guillot et al., 2015).

Arsenic in groundwater in the Terai

The worst affected districts in Nepal include Nawalparasi (Western Region), Rautahat and Bara
(Central Region) and Bardia (Midwestern Region). These districts, together with Parsa, Rupandehi,
Kapilbastu and Banke had priority areas for testing, water-supply mitigation and health screening
(BGS, 2001). The spatial and temporal distribution of elevated groundwater arsenic in Nepal is
unique in South Asia. In the Terai districts, elevated arsenic is found exclusively in the foreland basin
south of the Main Frontal Thrust (MFT), on the undisturbed floodplain. Surficial aquifers here are
formed from material eroded from the thrust wedge (immediately north of the MFT), which is
composed of earlier floodplain and later debris fan sediments exhumed by thrusting. Arsenic
occurrences are further limited to the areas immediately downslope from exposures of the fine-
grained Lower Siwalik Formation (Smith et al., 2004), comprised of meandering stream deposits laid
down during the initial uplift of the Himalaya (Brikowski et al., 2014). The thin alluvial aquifers of
the Nawalparasi district are some of the most severely As contaminated in the Terai region. Diwakar
et al. (2015) state that the alluvial sediments comprising the Terai aquifers in this district are derived
from two main sources, (i) sediments deposited by large rivers that erode the upper-Himalayan
crystalline rocks, (ii) weathered meta-sediments carried by smaller rivers originating in the Siwalik
forehills. The aquifer itself is characterized by Ca-HCO$_3$ type water and is multi-contaminated, with
the WHO guideline values exceeded for As, Mn and F in 80%, 70% and 40% of cases respectively.
The middle portion of the floodplain is heavily contaminated with As, predominantly as As(III). The
river water displayed some evidence of reductive processes in the hyporheic zone contributing As, Fe
and Mn to baseflow and also had elevated fluoride Diwakar et al. (2015). Fifty-five percent of water
samples collected from streams that drain the Terai, sedimentary rocks of the Siwalik Group,
carbonate and low-grade metamorphic rocks of the Lesser Himalayas had $\geq$ 0.01 ppm of arsenic
(Mukherjee et al., 2009).

Provenance of the aquifer sediments is relevant to tracing the source of arsenic. As already
mentioned there are two possible sources for the Terai sediments, the Siwalik hill and the higher
Himalayas. Sediments carried from the Siwalik hills by the minor rivers seem to release more arsenic
than those carried by major rivers from the higher Himalayas. REE and other charged cation
elements like Th, Sc, Hf, and Zr are highly immobile in most geological processes, and thus can be
used for provenance studies. The observed enrichment of incompatible elements is also indicative of
a felsic source. Sediments hosting As-contaminated aquifers are therefore probably homogeneous
mixtures of different types of rocks, with a felsic source. Studies of As contamination of groundwater
of the Bengal delta have demonstrated the geological control and found that high concentration of As
is restricted to the Holocene sediments rich in organic matter. Average arsenic content of the Terai
sediments is within the range of normal sediments (9 ppm). Abundances are greater in finer
sediments such as black clay (maximum 31 ppm) than in coarser sediments (silt and fine sand, 3
ppm). The sediments represent homogeneous mixtures of a wide range of parent rocks of felsic
composition. Significant As leaching rates indicate that the Terai sediments have high potential for
arsenic release, and that pH and redox conditions play crucial roles Gurung et al. (2005). Paudyal
(2011) mention that at present, there exist several possible natural sources of arsenic in Nepal. On the
basis of chemical and mineralogical analysis of collected rock, minerals, soil and water samples from
different parts of Nepal, several primary sources of arsenic have been identified (Sharma 1999; Sah
et al. 2003). The sulphide minerals from the polymetallic deposit of Ganesh Himal, iron ore of
Phulchauki area, ferruginous concretions of Tertiary deposits, bituminous coal of Tosh area, Dang;
Kalimati clay of the Kathmandu Valley and sediments from hot spring water show high values of
arsenic concentrations. Ferruginous quartzite, sandstone and mudstone also show comparatively higher values of arsenic. The above mentioned minerals, rocks and sediments could therefore represent the primary sources of arsenic in Nepal (Paudyal, 2011).

In Nawalparasi district clays contained particularly high amounts of iron, in the range of 21.9–59.9 g/kg (2-6 % in sediments). Together with the high levels of iron, high concentrations of aluminum were also extracted from the sediments (2.75-34.1 g/kg). Iron and aluminum in the sediments were positively correlated with arsenic, with correlation coefficients of 0.607 and 0.444, respectively. Arsenic is retained abundantly in finer particles like clay minerals, where it forms several different types of phases including ion exchange phases, carbonate and sulphide phases, ferric or manganous oxide and hydroxide phases, and soil organic matter phases depending on pH and redox potential (Eh) (Nakano et al. 2014). Yadav et al. (2015) in their article describe that As concentration varied from 0.22 to 0.64 ppm (mean 0.36 ppm) in sediment samples. Comparatively, higher concentration of As was observed mostly in the fine-grained clay sediments (black and yellow) than in coarse-grained sediments. A variety of Fe minerals in the Nawalparasi aquifer system are key host-phases for As. Specific examples include; goethite (α-FeOOH), authigenic pyrite (FeS₂) in deeper organic-clays and ferrihydrite flocs (Johnson et al., 2015). Vertical distribution of Fe followed similar distribution pattern as that of As showing its higher and lower concentration in clay and fine-sand, respectively. High As-yielding aquifers also contained higher percentage of calcium, silica, aluminum, and iron. Arsenic occurs generally in oxyanionic forms in aqueous environment. The hydrogeochemical data for groundwater of the TAP (Terai Alluvial Plain) aquifers suggest predominantly reducing character, with high HCO₃⁻, low SO₄²⁻, and NO₃⁻ concentrations. Elevated HCO₃⁻ levels result primarily due to the oxidation of organic matter while low SO₄²⁻ levels results as a result of sulphate reduction (Bhattacharya et al. 2003). Yadav et al. (2012) found three types of tubewells that are used as a source of drinking water in the Terai region which all vary by depth. These three include shallow tubewells (STW) (< 50 m deep), deep tubewells (DTW) (> 50 m deep), and dug wells (DW) (up to 20 m or more). A majority of them (98%) were STWs. The depth of DWs displayed various As concentrations. The depth of deep tubewells ranged from 1 to 183 m. Virtually all (97%) of the tested tubewells that had As levels exceeding WHO guidelines, were of less then a depth of 20 m. At this depth range, more than 8% of tubewells had As levels above 10 mg/L, while only 2% of tubewells had levels above 50 mg/L. At a depth of 21-50 m, 4.7 and 1.3 % of the water in tubewells had As concentrations that exceeded the 10 and 50 mg/L guideline levels, respectively. Similarly, at a depth greater than 50 m, tubewells having an As concentration that exceeded guideline values (10 and 50 mg/L) were significantly fewer in number. Therefore, it seems that tubewells having a depth less than 20 m had average higher As concentrations. Most of the known wells record
a high arsenic concentration in March, and a low value in May and September. A general pattern of low arsenic - low piezometric level, high arsenic - high piezometric level can be observed (Shrestha et al., 2004). According to Emerman (2005) central Nepal does not contain one geographically limited source of As in that nearly all rivers showed elevated levels of As. Nearly all rivers also showed elevated levels of Cu, Co, Fe, and Ni, while fluvial Zn was very close to the global background level. Therefore, As mineralisation may be associated with mineralisation of Cu, Co, Fe, or Ni, but probably not with Pb-Zn mineralisation (Pb and Zn are almost always associated). Bhusal and Paudyal (2014) clearly state that the distribution and occurrence of arsenic is controlled by geological material, much less by topography and not by land use, artificial fertilizers, pesticides and other organic additives.

Mechanism of arsenic release to groundwater

Since the fundamental work by Nickson et al. (2000) some scientific articles about the specific situation and mechanisms of arsenic release to the groundwater in the Terai in Nepal have been published. As outlined by Nickson et al. (2000) the As in the groundwater derives from reductive dissolution of As-rich Fe oxyhydroxides that exists as a dispersed phase (e.g. as a coating) on sedimentary grains. The reduction is driven by microbial degradation of sedimentary organic matter (O₂ consuming, O₂ as electron acceptor) and the redox process that occurs after microbial oxidation of organic matter takes place as soon as dissolved O₂ and NO₃ are disappeared. Strong correlation between dissolved organic carbon (DOC) and As in groundwater suggests that the microbial degradation of organic matter in the sediment results in an overall reducing environment and facilitates the release of As in the groundwater (Halim et al. 2009). Whilst arsenic release by the dissolution of arsenious pyrite is still recognized as a minor contributing but widespread process, a consensus view emerges in which the dominant process is, initially, the fixation of aqueous arsenic by sorption onto Fe-, Mn-oxide or clay surfaces during high-redox medium-pH conditions (i.e. about 5.5-6.5). Subsequently, desorptive release of arsenic occurs as groundwater becomes more reducing and alkaline (i.e. negative Eh and pH > 6.5), principally as the byproduct of bacterially-mediated FeOOH dissolution. Since the reducing agent is buried organic material, such as peat, mangroves, reed-swaps, etc., and since the predominant adsorption substrate is goethite or its analogues, with clay, high-iron and high-organic sedimentary environments are evidently prerequisites for the modern release of arsenic (Stanger 2005).

Arsenic (As, atomic number = 33) is a ubiquitous element, which ranks 20th in the earth’s crust. Arsenic exists in four oxidation states, +V (arsenate), +III (arsenite), 0 (arsenic), and −III (arsine).
Arsenic is unique among the heavy metalloids and oxy-anion forming elements. Its sensitivity to mobilisation largely depends on the pH values typically found in groundwater (pH 6.5-8.5) under both oxidizing and reducing conditions. The valency and species of inorganic arsenic highly dependent on the redox conditions (Eh) and the pH of the groundwater. Arsenite, the reduced trivalent form [As (III)], is normally present in groundwater (assuming anaerobic conditions) while arsenate, the oxidized pentavalent form [As (V)], is present in surface water (assuming aerobic conditions). In general, inorganic arsenic species are more toxic than organic forms of arsenic for living organisms. As already mentioned, redox potential and pH basically control arsenic speciation in natural environments. Inorganic arsenic primarily occurs as arsenic acid (H₃AsO₄) under oxidizing conditions, and predominates only at extremely high Eh values and low pH (<2). Within a pH range of 2 to 11 it is replaced by H₂AsO₄⁻ and HAsO₄²⁻. At low Eh values, H₃AsO₃ (arsenious acid) exists up to moderately alkaline pH but is replaced by H₂AsO₃⁻ at pH > 9.2 (Thakur et al., 2011; Zakhaznova-Herzog et al., 2006).

Bhattacharya et al. (2003) report about the groundwater in the Terai to be mostly near-neutral to alkaline within a pH range of 6.1-8.1. Redox potential (Eh) levels between -0.20 to -0.11 V suggesting fairly reduced condition in the aquifers. The groundwater is predominantly of Ca-Mg-Na-HCO₃⁻-type with HCO₃⁻ as the principal anion and low levels of Cl⁻ and SO₄²⁻. Low NO₃⁻ coupled with elevated NH₄⁺ concentrations in these groundwater reflects the dissimilarity nitrate reduction in the aquifers. Moreover, redox levels (Eh < -0.2 V) for sulphate reduction are sufficiently low, which facilitates the reduction of Fe³⁺ and Mn⁴⁺ in the aquifer sediments. The source of As in the subsurface environment is geogenic, and principally mobilized through natural interaction of the aqueous phases with the aquifer sediments under anoxic conditions. The sequence of redox reactions or terminal electron accepting processes (TEAP) prevalent in the aquifers plays critical role in controlling the As-chemistry in groundwater. The predominant TEAPs in the sedimentary aquifers are O₂ reduction (aerobic respiration), NO₃⁻ reduction (denitrification and dissimilatory nitrate reduction), Mn⁴⁺ reduction, Fe³⁺ reduction and SO₄²⁻ reduction with oxygen (O₂), NO₃⁻, Mn⁴⁺, Fe³⁺, and SO₄²⁻ as the prominent electron acceptors. High levels of Fe and Mn in the groundwater together with the predominance of As(III) in the groundwater suggest that As is mobilized due to the reductive dissolution Fe- and Mn-oxides and -hydroxides with sorbed As-oxyanions in the Terai sediments. According Panthi et al. (2006) the reductive desorption theory is the most likely explanation in which arsenic rich iron oxides break down and get dissolved into water regarding the context of strongly reducing environments (Eh -110 to -200 mV) of groundwater in Nepal. Moreover, the arsenic is thought to be closely associated with oxidation-reduction process of iron oxides and
Evidence exists to support oxidizing/reducing desorption of iron oxides and pyrite oxidation theories of releasing arsenic. But negative correlation between As and $\text{SO}_4^{2-}$ demonstrates the As may not be directly mobilized from sulphide minerals like arsenopyrite. In flooded soils, As is mobilized into porewater due to reductive dissolution of Fe$^{\text{III}}$ (hydr) oxides and to arsenate (As$^V$) reduction to the less competitively sorbing arsenite (As$^{\text{III}}$). By contrast, As concentrations in porewater are markedly lower under oxic conditions and generally dominated by As$^V$ (Roberts et al. 2011). Furthermore, the equilibrium of groundwater with respect to carbonate minerals and their precipitation/dissolution seems to be controlling the overall groundwater chemistry. The low $\text{SO}_4^{2-}$ and high DOC, $\text{PO}_4^{3-}$ and $\text{HCO}_3^-$ concentrations in groundwater signify ongoing microbial mediated redox processes favoring As mobilization in the aquifer. Multiple geochemical processes, e.g., Fe-oxyhydroxides reduction and carbonate dissolution ($\text{pH}!$), are responsible for high As occurrence in groundwaters Bhowmick et al. (2013).

The generally sub-oxic conditions, dominance of As(III) and Fe$^{2+}$ species and positive correlation between As and both NH$_3$ and UV-absorbance at 254 nm suggests that oxidation of organic matter coupled with microbial mediated reductive processes are important for mobilizing As in the Terai. The generally low redox potential of tube well waters combined with the abundance of reduced species of various redox sensitive elements (i.e. Fe$^{2+}$, As(III), NH$_3$) clearly indicates that reductive processes are important controls on aquifer geochemistry Diwakar et al. (2015). For example, McArthur et al. (2011) proposed that the absence or presence of a palaeo-weathering surface was a key control on As heterogeneity at their study site in West Bengal, India. They suggested that a palaeo-weathering surface formed during the last glacial maximum protects the underlying Pleistocene aquifer from contamination with DOC and As enriched water. According Brikowski et al. (2014) mitigation efforts concerning elevated arsenic in groundwater in Southeast Asia are hindered by persistent uncertainty about the proximal source of arsenic and mechanisms for its mobilization. At the core of this uncertainty seem to the relative roles of surficial organic clays vs. deeper aquifer matrix iron oxyhydroxides. Temporal variations in groundwater chemistry can serve to distinguish the contributions of these two sources, and such variation is especially pronounced in headwater areas of the Ganges floodplain immediately adjacent to the Himalayan foothills (e.g. the Terai of Nepal). Monsoon recharge refreshes these aquifers, temporarily minimizing arsenic concentrations. Post-monsoon, average groundwater compositions exhibit increasing trends in water–rock interaction (higher TDS, with cation exchange to form increasingly Na–$\text{HCO}_3^-$ waters), as well as in arsenic and iron concentrations. This cycle can be repeated during dry-season precipitation events as well, revealing direct correlation between trends in degree of clay interaction (sodium fraction of major cations) and arsenic concentrations. These observations strongly support a model of
reductive mobilization of arsenic from adjacent clays into aquifers, tempered by repeated flushing during periods of appreciable rainfall. Surficial sediments in the Terai exhibit extreme heterogeneity. Highly organic clays predominate in the shallow hydrologic system (the upper 50–100 m of surficial sediments contain >70% clay), and aquifer hydraulic conductivities are two orders of magnitude lower than in the delta. Low hydraulic conductivity of surficial fines limits infiltration, which likely enhances reducing conditions and mobilization of arsenic. In the Terai these factors combine to yield highly heterogeneous groundwater arsenic concentrations both in space and time, providing a valuable setting for exploring the arsenic mobilization process.

Mukherjee et al. (2012) state that information on groundwater chemistry in the central Ganges basin could provide insights into recharge, provenance, and fate of solutes in arsenic (As)-affected areas upstream of the more intensively studied Bengal basin. The area they studied extends from the northern edge of the Indian craton outcrops to the foothills of the Himalayas. Arsenic is probably mobilized by reductive dissolution of Fe–Mn (oxyhydr)oxides in the alluvium, with possibility of competitive anionic mobilization. Hence, relative to the Bengal basin, in addition to lower groundwater abstraction influence, groundwater chemistry in their study area reflects a greater variety of differences in the geological and geomorphological settings of the aquifers. Redox-sensitive parameters indicate generally reducing, post-oxic, metal-reducing conditions. However, redox conditions are highly spatially variable (oxic to methanic), with no systematic depth variation within sampled depth of the aquifers. Nakano et al. (2014) assume that the brown color of the sediments in the Terai arises from the presence of Fe(III) and the gray color can be induced by the reduction of Fe(III). The dissolution of FeOOH seems to be mainly derived by microbial fermentation under redox condition. They found that microbial degradation accompanying iron reduction released As attached on the surface of iron-bearing solids, however, the released As coupled with dissolution of iron can be continuously resorbed on the surface of solid phases like aluminosilicates (clay minerals) and silty sediments. Another possibility of resorption are crystalized iron bearing minerals which might be reproduced along with As during the sediment–water interactions controlled by microbial activity and redox condition. Microbial activity will be strongly affected by redox and pH changes. Upon saturation of adsorption sites, the As remains in the groundwater. The dissolution of calcium-related minerals may also play an important role in the process of releasing arsenic as this dissolution raises the pH locally, making the environment more alkaline. Alkaline conditions favor the desorption of As from As-bearing oxides as well as from organic matter. Low concentrations of NO$_3^-$ and SO$_4^{2-}$ together high Fe, as found in the geochemical analysis, also indicates reducing conditions, being prevalent in Terai groundwater. In sequential extraction techniques, chemical leaching by potassium chlorate and HCl releases As from sulphide and silicate phases. As exhibited in regression analysis, weak interrelationship between As, Fe and
SO$_4^{2−}$ suggests the absence of pyrite/arsenopyrite oxidation mechanism in present site. Further, if pyrite would have been oxidized, its As would have been sorbed onto the resulting Fe-oxyhydroxide rather than getting released in the groundwater. The leachable As content was high in organic matter phase next to sulphide/silicate phase as observed in sequential leaching analysis. This is an indication of the role of microbial population and organic matter in mobility of As under reducing condition. Moreover, the microbial oxidation of organic matter consumes dissolved oxygen present in the groundwater resulting in the formation of HCO$_3^{−}$. The distribution of grain size of the sediments in groundwater may also play a vital role in the mobility of As. It is evident from XRF analysis that high As concentration was mostly associated with fine-grained clay minerals. As the fine grain-size fractions has larger surface area it adsorbs the major part of As on their surface. Since, Fe, Mn and Al oxides and hydroxides are the major components of fine grained particles and thought to retain high As under specific pH conditions, their abundant percentage in Terai groundwater also suggests a reductive dissolution mechanism for As release Yadav et al. (2015). Finding presented by Johnston et al., (2015) provide direct XAS-based quantification of solid-phase As and Fe speciation in the alluvial aquifer sediments of the Terai region and help to shed light on key process controlling spatial patterns of solid-phase As/Fe speciation. Their dataset is broadly consistent with the widely invoked hypotheses that reductive dissolution of (near surface) Fe oxides and/or reductive desorption of As(III) coupled with downward transport are largely responsible for As mobilization in Gangetic floodplain aquifers (e.g. Fendorf et al., 2010). The findings also strongly affirm the critical role that various Fe minerals can play as host-phases for As as it undergoes redox cycling throughout the floodplain landscape. Most tube wells on the Nawalparasi floodplain are screened more than 15–20 m below ground level (Gurung et al., 2005) in order to tap permanently saturated thin sandy layers. Data presented in this article for the various floodplain sites indicate that at these depths, solid phase As(III) and lower valency As-sulphide species are the dominant species, while poorly crystalline Fe(III) and Fe oxides are largely absent. Consistent with the findings of Polizzotto et al. (2005), the paucity of Fe-oxides at the depth of tube well screens suggests that current mobilization of As(III) within these sedimentary facies is more likely due to downward transport or desorption of As(III) rather than contemporary in situ reductive dissolution of As-bearing Fe oxides occurring at the depth of well screens. Johnson et al., (2015) state that downstream transport is likely to be followed by some degree of floc reburial on the floodplain and therefore result in exposure of Fe(III) floc to seasonally fluctuating redox conditions. The material is freshly precipitated, very poorly-crystalline – hence susceptible to reductive dissolution – and contains readily exchangeable-As at concentrations well above those of bulk sediments. As-bearing authigenic pyrite occurs within 12 m of the ground surface at various floodplain sites and close (~5 m) to the current range of seasonal water table fluctuations. While stable under reducing conditions, if there is some regional lowering of water
tables, either through prolonged drought, climate-induced shifts in monsoonal precipitation, or excess groundwater abstraction, then these materials may be at risk of exposure and oxidation. Although oxidation of pyrite may simply cause As to shift host-phases and become sequestered in the resulting Fe(III) oxides (Polizzotto et al., 2006), it is conceivable there could be consequences for mobilizing additional As in the aquifer, especially in the short-term.

Mitigation options: Type of arsenic removal filters used in the Terai of Nepal

Following the study of Sharma (1999), several organizations and agencies have conducted surveys into arsenic contamination of well water in Nepal. NRCS/ENPHO in 2003 have provided the following six types of mitigation options to the arsenicosis patients in all VDCs of Rautahat district: (i) two-Gagri (water vessel) filter, (ii) innovated dug well, (iii) arsenic iron removal plant (AIRP), (iv) tube wells from arsenic free aquifer (v) modified biozsand filter and (vi) awareness program on nutrition. Of these, the option of two-Gagri filter and awareness program has been provided in Bagahi (Pradhan et al. 2006). According Nakano et al. (2014), Gagri filters and iron-assisted biozsand filters were later commonly used to remove arsenic and iron from well water in Nepal, which are believed to be the best treatment option at household levels (Yadav et al. 2011). This review will focus on the description of these two household filter types.

Gagri filters

One of the first filters employed was the two Gagri filter system with chemical powder. The system, consisting two earthen pots (Nepali language: gagri), uses chemical powder (a mixture of FeCl$_3$, NaOCl and charcoal). Ferric chloride is the compound that removes arsenic present in affected water. The candle filter aids in filtration of the coagulants formed in the upper pot. The second pot underneath the first one receives water free from arsenic, iron, bacteria and odour. This system is 90% efficient in removing arsenic and is below the Nepal interim standard. Further development led to the three gagri filter system. This filter replicates the three kulsi system of Bangladesh and solves the problem of chemical powder. Oxidation, adsorption, precipitation and filtration are the process for removal of arsenic and iron in this filter. This filter system can remove up to 95% of the arsenic, even when the water is highly contaminated. Retardation of filtration process due to clogging and presence of microbes in the treated water limits the filter’s performance. Therefore, techniques for improvement of microbiological quality should also have been used while
providing this option (Shrestha et al., 2004a). The 3-Gagri filter is a water container made of copper, brass, steel, tin, and or clay pot. The Three-Gagri Filter unit consists of three clay pots staggered vertically with a 1 cm in diameter hole in the bottom of the middle and top filters. The top and middle filters work as a reactor, and the bottom filter stores the treated water. The top filter contains the following, from bottom to top: a layer of polyester cloth, 3 kg of iron nails (3 cm depth), 2 kg of coarse sand (4 cm depth) and raw water. The middle filter contains the following from bottom to top: a layer of polyester cloth, about 50 kg of brickbats, 2 kg of fine sand (3.5 cm depth), 1 kg of charcoal (6 cm depth), 2 kg of brickbats (3 cm depth), and filtered water from the top filter. This filter could remove 95–99% of arsenic but there were problem with high iron in treated water and filter clogging due to bacterial growth. This filter was quickly replaced by arsenic biosand filter (Neku and Thandukar (2003); Thakur et al. (2011)).

Kanchan filters

These iron-assisted bio-sand filters were constructed on the basis of arsenic removal from water using zero-valent iron (ZVI) media. Under conditions applicable to drinking water treatment, arsenate removal by zero-valent iron media involves surface complexation only and does not involve reduction to metallic arsenic. Under the pH and redox conditions of most groundwaters and surface waters, dissolved arsenic exists as the As(V) (arsenate) species, H$_2$AsO$_4^-$ and HAsO$_4^{2-}$, and the As(III) (arsenite) species, H$_3$AsO$_3^0$ and H$_2$AsO$_3^-$. Removal of arsenic occurs through adsorption and coprecipitation during the formation of iron(III) hydroxides. However, acceptable levels of removal are achieved only when there is a filtration step to remove colloidal arsenic (Farrell et al., 2001). Greater attention is required for the removal of As(III) from groundwater due to its higher toxicity and mobility, which mainly arise from its neutral state (H$_3$AsO$_3^0$) in groundwater as compared to the charged As(V) species (H$_2$AsO$_4^-$ and HAsO$_4^{2-}$), which predominate near pH 6–9. The As(III) removal mechanism is mainly due to spontaneous adsorption and coprecipitation of As(III) with iron(II) and iron(III) oxides/hydroxides, which form in-situ during ZVI oxidation (corrosion). Heterogeneous reactions at the corroding ZVI surface are complex and result in a variety of potential adsorption surfaces for As(III) and As(V). Evidence has been presented showing that As(III) can be removed by adsorption on NZVI (nonoscale zero valent iron) in a very short time (minute scale) and is strongly adsorbed on NZVI over a wide range of pH and anion environments (Kanel et al., 2005). Investigations by Neumann et al. (2013) regarding the SONO household filters used in Bangladesh (an other version of an iron-assisted bio-sand filter) showed that over 95% of the As passing the top sand layer was removed in the CIM (composite iron matrix) by sorption, coprecipitation, and
incorporation into solids formed during the corrosion of ZVI. The continued presence of dissolved Fe(II) in the CIM appears to be important for the long-term operation of the filters. While young CIM contained large fractions of As in amorphous or poorly crystalline phases, magnetite was dominant in older CIM, consistent with an invariably deep black color. The transformation of As-rich Fe(III)(hydr)oxides into magnetite is important for two reasons: (i) the much denser magnetite does not lead to clogging of the filter and (ii) magnetite is more stable toward dissolution than freshly formed amorphous phases and leaches less As during milder extraction steps. Leaching tests with spent CIM in a previous study have shown very low remobilization of As, rendering used CIM nonhazardous. Because As is removed predominantly in the CIM, the other filter components such as sand, brick chips, and the plastic components can be disposed without special care.

The bio-sand filter (the modified model now used in Nepal is known as Kanchan filter) as a point-of-use drinking water treatment option was initially designed by Dr. David Manz of the University of Calgary, Canada in the late 1990 with support of numerous organizations and individuals. The biosand filters were modified to remove arsenic and tested in Nepal jointly by Massachusetts Institute of Technology (MIT) researchers; ENPHO, Nepal; Rural Water Supply and Sanitation Support Programme (RWSSSP), Nepal; and CAWST, Canada, based on slow sand filtration and iron hydroxide adsorption principles (Thakur et al., 2011). Now such filters can be used for removal of arsenic, iron, bacteria and turbidity. This filter uses the process of aeration, adsorption and filtration. As this system has a high flow rate of 30 litres per hour, the bio-sand filter has become in high demand in communities, not only for arsenic removal, but also due to higher flow rate. Field test showed that this filter removes more than 95% arsenic on average and up to 99% in some cases (NRCS–ENPHO (2003); Ngai and Walewijk, 2003). The filter also removes high levels of iron - up to 99%, with an average of 95%. Microbiological quality of this treated water is satisfactory (Shrestha et al., 2004a).

The Kanchan Arsenic Filter (KAF), an award-winning household water filter, was constructed for simultaneous arsenic and pathogen removal. The KAF is constructed using locally available labour and materials and is optimized based on the local socio-economic conditions. The Kanchan Arsenic Filter combines the concept of a slow sand filter for intermittent use (i.e. a biosand filter base) with the innovation of a diffuser basin containing (rusty) iron nails for arsenic removal. Operating under the water quality conditions encountered in the Terai region of Nepal (total arsenic <500mg/L, phosphate < 2 mg/L, pH < 8) the iron nails can last 3 years before replacement is necessary (Ngai et al. 2006). A two-year technical and social evaluation of over 1000 KAFs deployed in rural villages of Nepal determined that the KAF typically removes 85–90% arsenic, 90–95% iron, 80–95% turbidity, and 85–99% total coliforms. Then 83% of the households continued to use the filter after 1 year, mainly motivated by the clean appearance, improved taste, and reduced odour of the filtered water, as
compared to the original water source. In the KAF, non-galvanized iron nails are exposed to air and
water, rusting quickly and producing ferric hydroxide on the iron nails' surface. When arsenic-
containing water is poured into the filter, arsenic is rapidly adsorbed onto the surface of the ferric
hydroxide. This mechanism is similar to arsenic adsorption on zero-valent iron and arsenic
adsorption on hydrous ferric oxides. Some arsenic-loaded iron particles in the KAF are flushed on to
the sand layer below, and are trapped in the top few centimeters of the fine sand due to straining. As
ferric hydroxide particles “exfoliate” from the iron nails, new iron surfaces are created, providing
additional arsenic adsorption capacity. A Kathmandu university study found that iron and arsenic do
not migrate through the sand media over time (Ngai et al. 2007). The filter container can be
constructed out of concrete or plastic. The container is about 0.9 m tall 0.3 m in diameter (Fig. 2).
The container is filled with layers of sieved and washed sand and gravel. There is a standing water
height of 5 cm above the sand layer. The diffuser basin is filled with 5 to 6 kg of non-galvanized iron
nails for arsenic removal. In addition, pathogens, iron and suspended material are removed from
water through a combination of biological and physical processes: mechanical trapping,
adsorption/attraction, predation and natural death. This filter can treat approximately 10–15 L/h of
arsenic contaminated water. The filters are locally available at a cost of about 1,400 to 1,800 NRs
(about US$20) per filter (Thakur et al., 2011). Fig. 3 exhibits one of this operating filters in the
district Nawalparasi in October 2015. This zero-valent iron (ZVI)-based filters are able to remove
arsenic and other pollutants from drinking water, but their performance depends on the form of ZVI,
filter design, water composition and operating conditions. Kanchan filters use an upper bucket with
ZVI in the form of commercial iron nails, followed by a sand filter, to remove arsenic and pathogens.
Wenk et al. (2014) evaluated factors that influence the removal of arsenic and uranium with
laboratory columns containing iron nails with six different synthetic groundwater at pH 7.0 and 8.4
over 30 days. During the first 10 days, As removal was 65–95 % and strongly depended on the water
composition. As removal at pH 7.0 was better than at pH 8.4 and high P combined with low Ca
decreased As removal. From 10–30 days, As removal decreased to 45–60 % with all columns.
Phosphate, in combination with low Ca concentrations lowered As removal, but had a slightly
positive effect in combination with high Ca concentrations. The drop in performance over time can
be explained by decreasing release of iron to solution due to formation of layers of Fe\text{III}
phases and calcite covering the iron surface. Mobile corrosion products contained ferrihydrite, Si-containing
hydrous ferric oxides, and amorphous Fe–Si–P phases. Comparisons with another type of ZVI filter
(SONO-filter, see Hussam and Munir, 2007) were used to evaluate filter design parameters. Higher
ZVI surface areas and longer contact times should lead to satisfactory As removal with Kanchan-type
filters. Economical and promising methods are co-precipitation of As with naturally occurring or
added Fe\text{II} or Fe\text{III}, sorption or adsorption to inexpensive prefabricated sorbents or ion exchangers, or
As removal with metallic iron. Phosphate interacts strongly with precipitating Fe\textsuperscript{III} (hydr)oxides and outcompetes arsenic for sorption and incorporation, such that additional iron is necessary to remove both arsenic and phosphate. Removal units using metallic zero-valent iron (ZVI) are promising for several reasons: (1) ZVI filters can be constructed with locally available materials (typically sand and iron in various forms such as turnings, filings, nails or cleaned scrap iron). Corroding iron can potentially produce the largest amount of As-sorbing iron\textsuperscript{III} (hydr)oxides per mass of starting material. Aerobic iron corrosion leads to oxidation of As\textsuperscript{III} to the more strongly sorbing As\textsuperscript{V}, without the need of added oxidants. Two measures that could improve the performance of Kanchan filters are (1) larger specific ZVI surface areas (e.g. by use of smaller nails) and (2) increased contact times by more controlled and restricted flow from the upper diffuser bucket. As Singh et al. (2014) state in their article, the Kanchan filters efficacy in field conditions operating for a long period has been scarcely observed. They observed the efficacy of KAFs running over 6 months in highly arsenic-affected households in Nawalparasi district. Of 62 tubewells, 41 had influent arsenic concentration exceeding the Nepal drinking water quality standard value (50 µg/L). Of the 41 tubewells having unsafe arsenic levels, KAFs reduced arsenic concentration to the safe level for only 22 tubewells, an efficacy of 54%. In conclusion, they did not find significantly high efficacy of KAFs in reducing unsafe influent arsenic level to the safe level under the in situ field conditions.

Summary and future perspective

As mentioned above the factors that influence the removal of arsenic with laboratory columns containing iron nails were evaluated. As stated, the drop in performance over time could be explained by decreasing release of iron to solution due to formation of layers of Fe\textsuperscript{III} phases and calcite covering the iron surface. Inspection of operating filters in Nawalparasi district during a field campaing in October 2015 often revealed corrosion products covering the nails as well as insufficient contact time with the nails. Higher ZVI surface areas and longer contact times should lead to satisfactory As removal with Kanchan-type filters. Three critical measures that could improve the performance of Kanchan filters include: Larger specific ZVI surface areas (e.g. by use of smaller nails); Increased contact times by more controlled and restricted flow from the upper diffuser bucket and also immersed and anoxic conditions in the nailbed under no-flow conditions. Further improvements concerning these questions are under investigation.
Acknowledgements

I am grateful for the assistance of Dr. Stephan Hug, Eawag, Dübendorf, Prof. Dr. Christian de Capitani, Dept. Environmental Science, University of Basel and Dr. Marcel Guillong, Earth Sciences Department, ETH Zürich. My great appreciation for all support is also expressed to Tommy Ngai, Candice Young-Rojanschi, Finn Macdonald and Laura MacDonald from CAWST, Calgary, Canada; Bipin Dangol and Hari Boudhatoki ENPHO, Kathmandu, Nepal; Gyan Prakash Yadav, Parasi, Nepal and last but not least to Shankar Rai and Som Rai, my loyal expedition and trekking guides in Nepal and responsible for all logistics over many years.

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Fig. 1. Groundwater arsenic tested districts in various development regions of Nepal (from Thakur et al., 2011).
Fig. 2. Diagram of the KAF, showing the location and arrangement of its components. Source: Ngai et al. (2005).
Fig. 3. Operating filter in Nawalparasi district, Nepal, October 2015. Photo: B. Mueller.