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Journal:	<i>Environmental Reviews</i>
Manuscript ID	er-2016-0068.R2
Manuscript Type:	Review
Date Submitted by the Author:	15-Dec-2016
Complete List of Authors:	Mueller, Barbara; University of Basel,
Keyword:	arsenic, arsenic contamination, release of arsenic to the groundwater, removal of arsenic



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

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24 **Abstract:** As in several other countries of south east Asia (namely Bangladesh, India, Myanmar,
25 China, Vietnam, Cambodia) arsenic concentrations in the groundwater of the lowlands of Nepal (the
26 so called Terai) can reach concentrations which are unsafe to humans using the groundwater as
27 drinking water. Whereas Bangladesh has received much international attention concerning the
28 arsenic crisis, Nepal was more or less neglected. The first report about arsenic contamination of the
29 groundwater above toxic levels in Nepal was published in 1999. Twenty-four percent of samples
30 analyzed (n = 18,635) from the Terai Basin exceeded the WHO guideline of 10 µg/L. Since the first
31 overall survey from 2001, only sporadic information on the situation has been published. The
32 geological and geochemical conditions favour the release of the contaminant as arsenic can be easily
33 solubilized in groundwaters depending on pH, redox conditions, temperature, and solution
34 composition. The thin alluvial aquifers of the Terai are some of the most severely As contaminated.
35 These sediments constituting a high proportion of the Terai aquifers are derived from two main
36 sources, (i) sediments deposited by large rivers that erode the upper Himalayan crystalline rocks, (ii)
37 weathered meta-sediments carried by smaller rivers originating in the Siwalik forehills. The generally
38 low redox potential, low SO_4^{2-} , high DOC, PO_4^{3-} and HCO_3^- concentrations in groundwater signify
39 ongoing microbial mediated redox processes favoring As mobilization in the aquifer. Other
40 geochemical processes, e.g., Fe-oxyhydroxides reduction and carbonate dissolution are also
41 responsible for high As occurrence in groundwaters. Originally, Gagri filters (two filter system with
42 chemical powder) and later iron-assisted bio-sand filters were commonly used to remove arsenic and
43 iron from well water in Nepal – these two options were believed to be the best treatment option at
44 household levels. This review will focus on the description of the overall situation (geogenic issues,
45 occurrence of arsenic in the sediments of the Terai, mechanisms of the release of arsenic to the
46 groundwater, mitigation options).

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48 **Keywords:** arsenic; arsenic contamination; release of arsenic to the groundwater; removal of
49 arsenic; mitigation

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53 Introduction

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

63 Arsenic itself is not found in high abundance in the Earth's continental crust; it is less abundant
64 than several of the “rare-earth” elements. Unlike the rare-earth elements, however, arsenic is
65 commonly concentrated in sulphide-bearing mineral deposits, and it has a strong affinity for pyrite,
66 one of the more ubiquitous minerals in the Earth's crust. It is also concentrated in hydrous iron oxides
67 and clay minerals. Arsenic can be easily solubilized in groundwaters depending on pH, redox
68 conditions, temperature, and solution composition. Many geothermal waters contain high
69 concentrations of arsenic, too. Natural arsenic in groundwater at concentrations above the drinking
70 water guideline of 10 µg/L is not uncommon. A small number of source materials are now
71 recognized as significant contributors to arsenic in water supplies: organic-rich or black shales,
72 Holocene alluvial sediments with slow flushing rates, mineralized and mined areas (most often gold
73 deposits), volcanogenic sources, and thermal springs. Two other environments can lead to high
74 arsenic: (i) closed basins in arid-to-semi-arid climates (especially in volcanogenic provinces) and (ii)
75 strongly reducing aquifers, often composed of alluvial sediments but with low sulphate
76 concentrations. Young sediments in low-lying regions of low hydraulic gradient are characteristic of
77 many arsenic-rich aquifers. Ordinary sediments containing 1 to 20 mg/kg (near crustal abundance) of
78 arsenic can give rise to high dissolved arsenic (> 50 µg/L) if initiated by one or both of two possible
79 “triggers” - an increase in pH above 8.5 or the onset of reductive iron dissolution. Other important
80 factors promoting arsenic solubility are high concentrations of phosphate, bicarbonate, silicate,
81 and/or organic matter in the ground waters. These solutes can decrease or prevent the adsorption of
82 arsenate and arsenite ions onto fine-grained clays and especially iron oxides. Arsenite tends to adsorb
83 less strongly than arsenate often causing arsenite to be present at higher concentrations. The geologic
84 and groundwater conditions that promote high arsenic concentrations are now quite well known and
85 help identify high-risk areas (Nordstrom, 2002, Smedley and Kinniburgh, 2002). The water table
86 within the IGB (Indo-Gangetic Basin, including the Terai) alluvial aquifer is typically shallow (<5 m
87 below ground level). Abstraction of groundwater can also influence arsenic flux: It can flush aqueous

88 arsenic from the aquifer; irrigation pumping protects deeper groundwater in some instances, by
89 creating a hydraulic barrier, but it seems that high-capacity deep pumping may draw arsenic down to
90 levels in the Bengal aquifer system which are otherwise of good quality (MacDonald et al., 2016)

91 Whereas Bangladesh has received much international attention concerning the arsenic crisis (e. g.
92 Hug et al., 2011 and references therein), Nepal was more or less neglected, though the population of
93 the southern lowlands of Nepal (the so called Terai, the Indo-Gangetic Plain of southern Nepal) face
94 the same arsenic contamination of the groundwater (Nakano et al., 2014). The study of arsenic
95 concentrations in the groundwater in Nepal began only after the severity of the arsenic contamination
96 problem in the Bengal delta was recognized in 1998. The first report of arsenic contamination in
97 groundwater above toxic levels in Nepal was made from the Terai Basin (Sharma 1999). Twenty-
98 four percent of samples analyzed ($n = 18,635$) from the Terai Basin exceeded the WHO limit of 10
99 $\mu\text{g/L}$ (Shrestha and Shrestha 2004). Since the first overall survey conducted by WHO (2001), only
100 sporadic information on the situation has been published. Available documents later indicated that
101 the region of As contamination extends even into the Terai region and that 24 districts in Nepal,
102 including all 20 Terai districts and four hill districts, exhibit arsenic contamination (Bhattacharya et
103 al. 2003; Neku and Tandukar 2003; Shrestha et al. 2003; FAO, 2004; Tandukar et al. 2005; Panthi-
104 al., 2006; Maharjan et al., 2006; Pokhrel et al. 2009; Emerman et al., 2010; Thakur et al., 2011).
105 Although the Terai constitutes less than 20 % of the Nepal's surface, it contains over half of the total
106 arable land and is home to about 50 % of the total Nepalese population, i.e. 30 millions of
107 inhabitants. Groundwater is the main source of water for drinking and irrigation in the Terai area.
108 Over 90 % of the Terai population draws groundwater from tube wells for drinking, household use,
109 and irrigation (Guillot 2015).

110 According some publications, 25,058 tubewells in the Terai region have been tested for As, of
111 which 5,686 tubewells (22.7%) exceed the WHO (World Health Organization) As guideline (As =
112 0.01 mg/L) and 1,916 tubewells (7.6%) exceed the Nepal Interim As Standard (As = 0.05 mg/L)
113 (Panthi et al. 2006). It is estimated that there are perhaps 200,000 tubewells in the Terai region and
114 that 3.5 million Nepalese have no access to drinking water that does not exceed the WHO As
115 guideline (Mahat and Shrestha 2008; Mahat and Kharel, 2009; Pokhrel et al. 2009). In the most
116 recent report from NASC-NRCS (National Arsenic Steering Committee/National Red Cross Society,
117 2011) the total database covers 1.1 million wells tested between the years 2003 to 2008.
118 Approximately 1.73% showed values above the Nepal drinking water standard of 50 ppb, while
119 approximately 5.37% of tube wells contain 11-50 ppb of arsenic concentration. The percentage of all
120 tube wells exceeding 50 ppb varies from 0.05% of the wells in the district of Jhapa to 11.69% in the
121 district of Nawalparasi.

122 The most severe arsenic contamination is prevalent in several districts of the Terai namely

123 Nawalparasi, Bara, Parsa, Rautahat, Rupandehi, and Kapilivastu (Shrestha et al., 2014). Maharjan et
124 al. (2005) reported that 29% of more than 20,000 tube wells had arsenic concentrations exceeding the
125 World Health Organization (WHO) guideline (10 µg/L), that the prevalence of arsenicosis varied
126 between 1.3% and 5.1% (average of 2.6 %; see NRCS–ENPHO (Nepal Red Cross
127 Society/Environment & Public Health Organization), 2002; Yadav et. al., 2011) among four
128 independent surveys, and that approximately 0.5 million people in Terai were at risk of consuming
129 water with an arsenic concentration > 50 µg/L, the maximum permissible limit for Nepal (Shrestha et
130 al. 2003). It was found that overall prevalence of arsenicosis among the subjects ≥ 15 years old was
131 6.9%, which was comparable to those found by the same examiner in arsenic-contaminated areas in
132 Bangladesh, and that males had prevalence a twice as high as females, which could not be explained
133 by the difference in the exposure level Maharjan et al. (2005). These reports have alerted the decision
134 makers of the government as well as non-governmental agencies involved in controlling water
135 supply. As a consequence, in 2003 the National Arsenic Steering Committee (NASC) was formed,
136 involving major stakeholders from the drinking water and sanitation sector (Shrestha et al. 2003).
137 The NASC worked in collaboration with the Environment Public Health Organization (ENPHO) to
138 perform testing on 18,635 tubewells in 20 Terai districts, under a program called the “State of
139 Arsenic in Nepal 2003.” All the data collected revealed that concentration of As varied both spatially
140 and seasonally, suggesting the possibility of spatial variation due to geospatial conditions such as
141 latitude, longitude and depth of tubewell. The temporal distribution of As showed seasonal
142 dependence with lower concentration in winter and higher in summer (Yadav et al., 2012).

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145 **Geological situation of the Terai region**

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

158 Archean crystalline formations deep beneath the Alluvium of the Terai as well as the marine
159 sedimentary deposits forming the high Himalayas, and the Siwalik formation formed by the then
160 east–west flowing rivers can be find within confined space (Yadav et al., 2015).

161 The prominent mountain chain in Nepal – the Himalayas – is built up by four major Himalayan
162 tectonic units: (1) the Tethys Himalaya, delimited at the base by the South Tibetan Detachment
163 system (STDS); (2) the Higher Himalayan Crystallines (HHC) delimited at the base by the Main
164 Central Thrust I (MCT I); (3) the Lesser Himalaya (LH) divided into upper and lower Lesser
165 Himalaya, is delimited at the base by the Main Boundary Thrust (MBT); and (4) the Siwaliks,
166 delimited at its base by the Main Frontal Thrust (MFT) and the Quaternary foreland basin. These
167 units span a wide range of various rocks being metamorphic, sedimentary, and igneous in origin,
168 making it possible for their differential erosion to account for some of the groundwater arsenic
169 heterogeneity we see in the foreland and delta (i.e. Gurung et al. 2005; Shah 2008; van Geen et al.
170 2008; Guillot et al., 2015). The Terai Plain is an active foreland basin consisting of Quaternary
171 sediments that include molasse units along with gravel, sand, silt, and clay. Most of the rivers in the
172 Terai flow from north to south. All major rivers originate in the high Himalayas whereas minor rivers
173 also emanate from the nearby Siwalik Hills, and therefore deposit sediments in the form of a fan
174 along the flank of the Terai basin. Fine sediments and organic material are deposited in inter-fan
175 lowlands, in wetlands and swamps (Sharma 1995). The Siwalik lithofacies are strongly diachronous,
176 and further complicated by a variable addition of micaceous sands and arkose, locally derived from
177 southward-draining tributaries from the emerging Himalayas. The most typical Siwalik lithologies
178 are conglomerates, ‘salt and pepper’ micaceous sandstones, blue-grey siltstones, clay-stones, red
179 (iron-rich) shales, and minor lignite. Potential adsorption substrates and co-precipitation hosts for
180 arsenic are common throughout the finer-grained Siwalik facies, as iron mineralization, as sulphides,
181 or as clays. In Nepal, the groundwater arsenic is of relatively local provenance, being derived directly
182 from eroded Siwaliks (Stanger 2005).

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

188 The geology of the Terai region of Nepal itself is on the whole comparable to the Bengal Delta
189 Plain (BDP) and it is the continuation of Indo-gangetic trough. The Terai plain covered by recent and
190 older alluvium comprises channel sand and gravel deposits and outwash deposits. These fluvial
191 deposits are cross-bedded, eroded, reworked, and redeposited because of regular shifting of stream
192 channels. Geomorphologically, the Terai plain is divided into two zones: the Bhabar zone in the

193 north and the main Terai zone in the south. They have diverse hydrogeological characteristics and are
194 separated by a line of natural springs. The Bhabar zone is a narrow extension of a recent alluvial and
195 colluvial fan deposit at the bottom of Siwalik Hills (Kansakar 2004). It consists of thick deposits of
196 gravel, pebble, and boulder mixed with sand and silt. Sediments in the main Terai were deposited by
197 braided rivers, which regularly changed their course. As a result, clay, silt, sand, and gravel deposits
198 of varying thicknesses occur interlayered with each other. The Terai plain has a multiple aquifer
199 system (Yadav et al., 2011).

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

222 Small ephemeral rivers originating from the Siwalik frontal mountains disappear upon entering
223 into the Indo-Gangetic plain and reappear again in Nawalparasi. Hence, small natural ponds and river
224 meanderings were observed as characteristic geomorphic features of the area. Therefore, close to the
225 frontal mountain chain, the Indo-Gangetic plain consists of boulder- to gravel-sized sediments, while
226 soils further south consists dominantly of fine-grained sediments. Guillot et al. (2015) report about
227 the lithology of sledge core samples from the five drill holes showing various coarse (millimetric) to

228 fine-grained (micrometric) sediments in the Narayani basin. They distinguished light-grey to dark
229 grey sands; grey, greenish-grey to brown-grey and yellow– brown silts; and light-grey to black-grey,
230 yellow–brown and black clay with occasional gravel layers. Macroscopic observations showed that
231 on average, the drilled sediments are composed of 33 % of silts; 30 % of grey to black clays, 27 % of
232 brown clay, 9 % of fine-grained silt and sand and less than 1 % of calcrete. Sands, silt and clay
233 sediments often contained micas that were occasionally massive to laminated, bioturbated, and/or
234 also containing roots and plant debris. Binocular observations show that the detrital minerals in the
235 silt fraction are dominated by quartz, biotite, muscovite, K-feldspar, calcite and dolomite as major
236 phases and garnet, zircon, and monazite as heavy minerals. In the region of provenance of the
237 Narayani basin, the Tethys Himalaya includes 10 km of various metasedimentary rocks (limestones,
238 calc-schists, shales, quartzites) ranging from Cambrian to Jurassic. There is also the Manaslu
239 leucogranite emplaced within the Tethyan rocks. The Higher Himalayan Crystallines are a
240 metamorphic stack, including from base to top paragneisses (metapelites and metapsammites);
241 gneisses with calcsilicate minerals (diopside and amphibole) and orthogneisses representing
242 metamorphosed Lower Paleozoic granites. The Lesser Himalaya consists of mostly unfossiliferous
243 metasediments and some dolomitic meta-carbonates alternating with dominant black schists,
244 aluminium rich schists and quartzites. Amphibolites occur in both of these groupings. The Siwaliks
245 represent the Cenozoic foreland basin of the Himalayan belt with local thickness of 6 km in Nepal.
246 They are divided into three units having a typical coarsening-upward succession. The lower unit
247 consists of fluvial channel sandstones alternating with calcareous paleosols; the middle unit consists
248 of very thick channel sandstones with minor paleosols, and the upper unit mainly hosts
249 conglomerates of gravelly braided river deposits (after Guillot et al., 2015).

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251

252 **Arsenic in groundwater in the Terai**

253

254 The worst affected districts in Nepal include Nawalparasi (Western Region), Rautahat and Bara
255 (Central Region) and Bardia (Midwestern Region). These districts, together with Parsa, Rupandehi,
256 Kapilbastu and Banke had priority areas for testing, water-supply mitigation and health screening
257 (BGS, 2001). The spatial and temporal distribution of elevated groundwater arsenic in Nepal is
258 unique in South Asia. In the Terai districts, elevated arsenic is found exclusively in the foreland basin
259 south of the Main Frontal Thrust (MFT), on the undisturbed floodplain. Surficial aquifers here are
260 formed from material eroded from the thrust wedge (immediately north of the MFT), which is
261 composed of earlier floodplain and later debris fan sediments exhumed by thrusting. Arsenic
262 occurrences are further limited to the areas immediately downslope from exposures of the fine-

263 grained Lower Siwalik Formation (Smith et al., 2004), comprised of meandering stream deposits laid
264 down during the initial uplift of the Himalaya (Brikowski et al., 2014). The thin alluvial aquifers of
265 the Nawalparasi district are some of the most severely As contaminated in the Terai region. Diwakar
266 et al. (2015) state that the alluvial sediments comprising the Terai aquifers in this district are derived
267 from two main sources, (i) sediments deposited by large rivers that erode the upper-Himalayan
268 crystalline rocks, (ii) weathered meta-sediments carried by smaller rivers originating in the Siwalik
269 forehills. The aquifer itself is characterized by Ca-HCO₃ type water and is multi-contaminated, with
270 the WHO guideline values exceeded for As, Mn and F in 80%, 70% and 40% of cases respectively.
271 The middle portion of the floodplain is heavily contaminated with As, predominantly as As(III). The
272 river water displayed some evidence of reductive processes in the hyporheic zone contributing As, Fe
273 and Mn to baseflow and also had elevated fluoride Diwakar et al. (2015). Fifty-five percent of water
274 samples collected from streams that drain the Terai, sedimentary rocks of the Siwalik Group,
275 carbonate and low-grade metamorphic rocks of the Lesser Himalayas had ≥ 0.01 ppm of arsenic
276 (Mukherjee et al., 2009).

277 Provenance of the aquifer sediments is relevant to tracing the source of arsenic. As already
278 mentioned there are two possible sources for the Terai sediments, the Siwalik hill and the higher
279 Himalayas. Sediments carried from the Siwalik hills by the minor rivers seem to release more arsenic
280 than those carried by major rivers from the higher Himalayas. REE and other charged cation
281 elements like Th, Sc, Hf, and Zr are highly immobile in most geological processes, and thus can be
282 used for provenance studies. The observed enrichment of incompatible elements is also indicative of
283 a felsic source. Sediments hosting As-contaminated aquifers are therefore probably homogeneous
284 mixtures of different types of rocks, with a felsic source. Studies of As contamination of groundwater
285 of the Bengal delta have demonstrated the geological control and found that high concentration of As
286 is restricted to the Holocene sediments rich in organic matter. Average arsenic content of the Terai
287 sediments is within the range of normal sediments (9 ppm). Abundances are greater in finer
288 sediments such as black clay (maximum 31 ppm) than in coarser sediments (silt and fine sand, 3
289 ppm). The sediments represent homogeneous mixtures of a wide range of parent rocks of felsic
290 composition. Significant As leaching rates indicate that the Terai sediments have high potential for
291 arsenic release, and that pH and redox conditions play crucial roles Gurung et al. (2005). Paudyal
292 (2011) mention that at present, there exist several possible natural sources of arsenic in Nepal. On the
293 basis of chemical and mineralogical analysis of collected rock, minerals, soil and water samples from
294 different parts of Nepal, several primary sources of arsenic have been identified (Sharma 1999; Sah
295 et al. 2003). The sulphide minerals from the polymetallic deposit of Ganesh Himal, iron ore of
296 Phulchauki area, ferruginous concretions of Tertiary deposits, bituminous coal of Tosh area, Dang;
297 Kalimati clay of the Kathmandu Valley and sediments from hot spring water show high values of

298 arsenic concentrations. Ferruginous quartzite, sandstone and mudstone also show comparatively
299 higher values of arsenic. The above mentioned minerals, rocks and sediments could therefore
300 represent the primary sources of arsenic in Nepal (Paudyal, 2011).

301 In Nawalparasi district clays contained particularly high amounts of iron, in the range of 21.9–
302 59.9 g/kg (2-6 % in sediments). Together with the high levels of iron, high concentrations of
303 aluminum were also extracted from the sediments (2.75-34.1 g/kg). Iron and aluminum in the
304 sediments were positively correlated with arsenic, with correlation coefficients of 0.607 and 0.444,
305 respectively. Arsenic is retained abundantly in finer particles like clay minerals, where it forms
306 several different types of phases including ion exchange phases, carbonate and sulphide phases, ferric
307 or manganous oxide and hydroxide phases, and soil organic matter phases depending on pH and
308 redox potential (Eh) (Nakano et al. 2014). Yadav et al. (2015) in their article describe that As
309 concentration varied from 0.22 to 0.64 ppm (mean 0.36 ppm) in sediment samples. Comparatively,
310 higher concentration of As was observed mostly in the fine-grained clay sediments (black and
311 yellow) than in coarse-grained sediments. A variety of Fe minerals in the Nawalparasi aquifer system
312 are key host-phases for As. Specific examples include; goethite (α -FeOOH), authigenic pyrite (FeS₂)
313 in deeper organic-clays and ferrihydrite floes (Johnson et al., 2015). Vertical distribution of Fe
314 followed similar distribution pattern as that of As showing its higher and lower concentration in clay
315 and fine-sand, respectively. High As-yielding aquifers also contained higher percentage of calcium,
316 silica, aluminum, and iron. Arsenic occurs generally in oxyanionic forms in aqueous environment.
317 The hydrogeochemical data for groundwater of the TAP (Terai Alluvial Plain) aquifers suggest
318 predominantly reducing character, with high HCO₃⁻, low SO₄²⁻, and NO₃⁻ concentrations. Elevated
319 HCO₃⁻ levels result primarily due to the oxidation of organic matter while low SO₄²⁻ levels results as
320 a result of sulphate reduction (Bhattacharya et al. 2003). Yadav et al. (2012) found three types of
321 tubewells that are used as a source of drinking water in the Terai region which all vary by depth.
322 These three include shallow tubewells (STW) (< 50 m deep), deep tubewells (DTW) (> 50 m deep),
323 and dug wells (DW) (up to 20 m or more). A majority of them (98%) were STWs. The depth of DWs
324 displayed various As concentrations. The depth of deep tubewells ranged from 1 to 183 m. Virtually
325 all (97%) of the tested tubewells that had As levels exceeding WHO guidelines, were of less than a
326 depth of 20 m. At this depth range, more than 8% of tubewells had As levels above 10 mg/L, while
327 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
328 tubewells had As concentrations that exceeded the 10 and 50 mg/L guideline levels, respectively.
329 Similarly, at a depth greater than 50 m, tubewells having an As concentration that exceeded guideline
330 values (10 and 50 mg/L) were significantly fewer in number. Therefore, it seems that tubewells
331 having a depth less than 20 m had average higher As concentrations. Most of the known wells record

332 a high arsenic concentration in March, and a low value in May and September. A general pattern of
333 low arsenic - low piezometric level, high arsenic - high piezometric level can be observed (Shrestha
334 et al., 2004). According to Emerman (2005) central Nepal does not contain one geographically
335 limited source of As in that nearly all rivers showed elevated levels of As. Nearly all rivers also
336 showed elevated levels of Cu, Co, Fe, and Ni, while fluvial Zn was very close to the global
337 background level. Therefore, As mineralisation may be associated with mineralisation of Cu, Co, Fe,
338 or Ni, but probably not with Pb-Zn mineralisation (Pb and Zn are almost always associated). Bhusal
339 and Paudyal (2014) clearly state that the distribution and occurrence of arsenic is controlled by
340 geological material, much less by topography and not by land use, artificial fertilizers, pesticides and
341 other organic additives.

342

343

344 **Mechanism of arsenic release to groundwater**

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346 Since the fundamental work by Nickson et al. (2000) some scientific articles about the specific
347 situation and mechanisms of arsenic release to the groundwater in the Terai in Nepal have been
348 published. As outlined by Nickson et al. (2000) the As in the groundwater derives from reductive
349 dissolution of As-rich Fe oxyhydroxides that exists as a dispersed phase (e.g. as a coating) on
350 sedimentary grains. The reduction is driven by microbial degradation of sedimentary organic matter
351 (O_2 consuming, O_2 as electron acceptor) and the redox process that occurs after microbial oxidation
352 of organic matter takes place as soon as dissolved O_2 and NO_3 are disappeared. Strong correlation
353 between dissolved organic carbon (DOC) and As in groundwater suggests that the microbial
354 degradation of organic matter in the sediment results in an overall reducing environment and
355 facilitates the release of As in the groundwater (Halim et al. 2009). Whilst arsenic release by the
356 dissolution of arsenious pyrite is still recognized as a minor contributing but widespread process, a
357 consensus view emerges in which the dominant process is, initially, the fixation of aqueous arsenic
358 by sorption onto Fe-, Mn-oxide or clay surfaces during high-redox medium-pH conditions (i.e. about
359 5.5-6.5). Subsequently, desorptive release of arsenic occurs as groundwater becomes more reducing
360 and alkaline (i.e. negative Eh and $pH > 6.5$), principally as the byproduct of bacterially-mediated
361 $FeOOH$ dissolution. Since the reducing agent is buried organic material, such as peat, mangroves,
362 reed-swaps, etc., and since the predominant adsorption substrate is goethite or its analogues, with
363 clay, high-iron and high-organic sedimentary environments are evidently prerequisites for the
364 modern release of arsenic (Stanger 2005).

365 Arsenic (As, atomic number = 33) is a ubiquitous element, which ranks 20th in the earth's crust.
366 Arsenic exists in four oxidation states, +V (arsenate), +III (arsenite), 0 (arsenic), and -III (arsine).

367 Arsenic is unique among the heavy metalloids and oxy-anion forming elements. Its sensitivity to
368 mobilisation largely depends on the pH values typically found in groundwater (pH 6.5-8.5) under
369 both oxidizing and reducing conditions. The valency and species of inorganic arsenic highly
370 dependent on the redox conditions (E_h) and the pH of the groundwater. Arsenite, the reduced
371 trivalent form [As (III)], is normally present in groundwater (assuming anaerobic conditions) while
372 arsenate, the oxidized pentavalent form [As (V)], is present in surface water (assuming aerobic
373 conditions). In general, inorganic arsenic species are more toxic than organic forms of arsenic for
374 living organisms. As already mentioned, redox potential and pH basically control arsenic speciation
375 in natural environments. Inorganic arsenic primarily occurs as arsenic acid (H_3AsO_4) under
376 oxidizing conditions, and predominates only at extremely high Eh values and low pH (<2). Within a
377 pH range of 2 to 11 it is replaced by $H_2AsO_4^-$ and $HAsO_4^{2-}$. At low Eh values, H_3AsO_3 (arsenious
378 acid) exists up to moderately alkaline pH but is replaced by $H_2AsO_3^-$ at pH > 9.2 (Thakur et al.,
379 2011; Zakhaznova-Herzog et al., 2006).

380 Bhattacharya et al. (2003) report about the groundwater in the Terai to be mostly near-neutral to
381 alkaline within a pH range of 6.1-8. 1. Redox potential (E_h) levels between -0.20 to -0.11 V
382 suggesting fairly reduced condition in the aquifers. The groundwater is predominantly of Ca-Mg-Na-
383 HCO_3^- -type with HCO_3^- as the principal anion and low levels of Cl^- and SO_4^{2-} . Low NO_3^- coupled
384 with elevated NH_4^+ concentrations in these groundwater reflects the dissimilarity nitrate reduction in
385 the aquifers. Moreover, redox levels ($E_h < -0.2$ V) for sulphate reduction are sufficiently low, which
386 facilitates the reduction of Fe^{3+} and Mn^{4+} in the aquifer sediments. The source of As in the
387 subsurface environment is geogenic, and principally mobilized through natural interaction of the
388 aqueous phases with the aquifer sediments under anoxic conditions. The sequence of redox reactions
389 or terminal electron accepting processes (TEAP) prevalent in the aquifers plays critical role in
390 controlling the As-chemistry in groundwater. The predominant TEAPs in the sedimentary aquifers
391 are O_2 reduction (aerobic respiration), NO_3^- reduction (denitrification and dissimilatory nitrate
392 reduction), Mn^{4+} reduction, Fe^{3+} reduction and SO_4^{2-} reduction with oxygen (O_2), NO_3^- , Mn^{4+} ,
393 Fe^{3+} , and SO_4^{2-} as the prominent electron acceptors. High levels of Fe and Mn in the groundwater
394 together with the predominance of As(III) in the groundwater suggest that As is mobilized due to the
395 reductive dissolution Fe- and Mn-oxides and -hydroxides with sorbed As-oxyanions in the Terai
396 sediments. According Panthi et al. (2006) the reductive desorption theory is the most likely
397 explanation in which arsenic rich iron oxides break down and get dissolved into water regarding the
398 context of strongly reducing environments (E_h -110 to -200 mV) of groundwater in Nepal. Moreover,
399 the arsenic is thought to be closely associated with oxidation-reduction process of iron oxides and

400 pyrite. Evidence exists to support oxidizing/reducing desorption of iron oxides and pyrite oxidation
401 theories of releasing arsenic. But negative correlation between As and SO_4^{2-} demonstrates the As
402 may not be directly mobilized from sulphide minerals like arsenopyrite. In flooded soils, As is
403 mobilized into porewater due to reductive dissolution of Fe^{III} (hydr) oxides and to arsenate (As^{V})
404 reduction to the less competitively sorbing arsenite (As^{III}). By contrast, As concentrations in
405 porewater are markedly lower under oxic conditions and generally dominated by As^{V} (Roberts et al.
406 2011). Furthermore, the equilibrium of groundwater with respect to carbonate minerals and their
407 precipitation/dissolution seems to be controlling the overall groundwater chemistry. The low SO_4^{2-}
408 and high DOC, PO_4^{3-} and HCO_3^- concentrations in groundwater signify ongoing microbial mediated
409 redox processes favoring As mobilization in the aquifer. Multiple geochemical processes, e.g., Fe-
410 oxyhydroxides reduction and carbonate dissolution (pH!), are responsible for high As occurrence in
411 groundwaters Bhowmick et al. (2013).

412 The generally sub-oxic conditions, dominance of As^{III} and Fe^{2+} species and positive correlation
413 between As and both NH_3 and UV-absorbance at 254 nm suggests that oxidation of organic matter
414 coupled with microbial mediated reductive processes are important for mobilizing As in the aquifers
415 in the Terai. The generally low redox potential of tube well waters combined with the abundance of
416 reduced species of various redox sensitive elements (i.e. Fe^{2+} , As^{III} , NH_3) clearly indicates that
417 reductive processes are important controls on aquifer geochemistry Diwakar et al. (2015). For
418 example, McArthur et al. (2011) proposed that the absence or presence of a palaeo-weathering
419 surface was a key control on As heterogeneity at their study site in West Bengal, India. They
420 suggested that a palaeo-weathering surface formed during the last glacial maximum protects the
421 underlying Pleistocene aquifer from contamination with DOC and As enriched water. According
422 Brikowski et al. (2014) mitigation efforts concerning elevated arsenic in groundwater in Southeast
423 Asia are hindered by persistent uncertainty about the proximal source of arsenic and mechanisms for
424 its mobilization. At the core of this uncertainty seem to be the relative roles of surficial organic clays vs.
425 deeper aquifer matrix iron oxyhydroxides. Temporal variations in groundwater chemistry can serve
426 to distinguish the contributions of these two sources, and such variation is especially pronounced in
427 headwater areas of the Ganges floodplain immediately adjacent to the Himalayan foothills (e.g. the
428 Terai of Nepal). Monsoon recharge refreshes these aquifers, temporarily minimizing arsenic
429 concentrations. Post-monsoon, average groundwater compositions exhibit increasing trends in water-
430 rock interaction (higher TDS, with cation exchange to form increasingly Na- HCO_3 waters), as well
431 as in arsenic and iron concentrations. This cycle can be repeated during dry-season precipitation
432 events as well, revealing direct correlation between trends in degree of clay interaction (sodium
433 fraction of major cations) and arsenic concentrations. These observations strongly support a model of

434 reductive mobilization of arsenic from adjacent clays into aquifers, tempered by repeated flushing
435 during periods of appreciable rainfall. Surficial sediments in the Terai exhibit extreme heterogeneity.
436 Highly organic clays predominate in the shallow hydrologic system (the upper 50–100 m of surficial
437 sediments contain >70% clay), and aquifer hydraulic conductivities are two orders of magnitude
438 lower than in the delta. Low hydraulic conductivity of surficial fines limits infiltration, which likely
439 enhances reducing conditions and mobilization of arsenic. In the Terai these factors combine to yield
440 highly heterogeneous groundwater arsenic concentrations both in space and time, providing a
441 valuable setting for exploring the arsenic mobilization process.

442 Mukherjee et al. (2012) state that information on groundwater chemistry in the central Ganges
443 basin could provide insights into recharge, provenance, and fate of solutes in arsenic (As)-affected
444 areas upstream of the more intensively studied Bengal basin. The area they studied extends from the
445 northern edge of the Indian craton outcrops to the foothills of the Himalayas. Arsenic is probably
446 mobilized by reductive dissolution of Fe–Mn (oxyhydr)oxides in the alluvium, with possibility of
447 competitive anionic mobilization. Hence, relative to the Bengal basin, in addition to lower
448 groundwater abstraction influence, groundwater chemistry in their study area reflects a greater
449 variety of differences in the geological and geomorphological settings of the aquifers. Redox-
450 sensitive parameters indicate generally reducing, post-oxic, metal-reducing conditions. However,
451 redox conditions are highly spatially variable (oxic to methanic), with no systematic depth variation
452 within sampled depth of the aquifers. Nakano et al. (2014) assume that the brown color of the
453 sediments in the Terai arises from the presence of Fe(III) and the gray color can be induced by the
454 reduction of Fe(III). The dissolution of FeOOH seems to be mainly derived by microbial
455 fermentation under redox condition. They found that microbial degradation accompanying iron
456 reduction released As attached on the surface of iron-bearing solids, however, the released As
457 coupled with dissolution of iron can be continuously resorbed on the surface of solid phases like
458 aluminosilicates (clay minerals) and silty sediments. Another possibility of resorption are crystallized
459 iron bearing minerals which might be reproduced along with As during the sediment–water
460 interactions controlled by microbial activity and redox condition. Microbial activity will be strongly
461 affected by redox and pH changes. Upon saturation of adsorption sites, the As remains in the
462 groundwater. The dissolution of calcium-related minerals may also play an important role in the
463 process of releasing arsenic as this dissolution raises the pH locally, making the environment more
464 alkaline. Alkaline conditions favor the desorption of As from As-bearing oxides as well as from
465 organic matter. Low concentrations of NO_3^- and SO_4^{2-} together high Fe, as found in the geochemical
466 analysis, also indicates reducing conditions, being prevalent in Terai groundwater. In sequential
467 extraction techniques, chemical leaching by potassium chlorate and HCl releases As from sulphide
468 and silicate phases. As exhibited in regression analysis, weak interrelationship between As, Fe and

469 SO_4^{2-} suggests the absence of pyrite/arsenopyrite oxidation mechanism in present site. Further, if
470 pyrite would have been oxidized, its As would have been sorbed onto the resulting Fe-oxyhydroxide
471 rather than getting released in the groundwater. The leachable As content was high in organic matter
472 phase next to sulphide/silicate phase as observed in sequential leaching analysis. This is an indication
473 of the role of microbial population and organic matter in mobility of As under reducing condition.
474 Moreover, the microbial oxidation of organic matter consumes dissolved oxygen present in the
475 groundwater resulting in the formation of HCO_3^- . The distribution of grain size of the sediments in
476 groundwater may also play a vital role in the mobility of As. It is evident from XRF analysis that
477 high As concentration was mostly associated with fine-grained clay minerals. As the fine grain-size
478 fractions has larger surface area it adsorbs the major part of As on their surface. Since, Fe, Mn and Al
479 oxides and hydroxides are the major components of fine grained particles and thought to retain high
480 As under specific pH conditions, their abundant percentage in Terai groundwater also suggests a
481 reductive dissolution mechanism for As release Yadav et al. (2015). Finding presented by Johnston et
482 al., (2015) provide direct XAS-based quantification of solid-phase As and Fe speciation in the
483 alluvial aquifer sediments of the Terai region and help to shed light on key process controlling spatial
484 patterns of solid-phase As/Fe speciation. Their dataset is broadly consistent with the widely invoked
485 hypotheses that reductive dissolution of (near surface) Fe oxides and/or reductive desorption of
486 As(III) coupled with downward transport are largely responsible for As mobilization in Gangetic
487 floodplain aquifers (e.g. Fendorf et al., 2010). The findings also strongly affirm the critical role that
488 various Fe minerals can play as host-phases for As as it undergoes redox cycling throughout the
489 floodplain landscape. Most tube wells on the Nawalparasi floodplain are screened more than 15–20
490 m below ground level (Gurung et al., 2005) in order to tap permanently saturated thin sandy layers.
491 Data presented in this article for the various floodplain sites indicate that at these depths, solid phase
492 As(III) and lower valency As-sulphide species are the dominant species, while poorly crystalline
493 Fe(III) and Fe oxides are largely absent. Consistent with the findings of Polizzotto et al. (2005), the
494 paucity of Fe-oxides at the depth of tube well screens suggests that current mobilization of As(III)
495 within these sedimentary facies is more likely due to downward transport or desorption of As(III)
496 rather than contemporary in situ reductive dissolution of As-bearing Fe oxides occurring at the depth
497 of well screens. Johnson et al., (2015) state that downstream transport is likely to be followed by
498 some degree of floc reburial on the floodplain and therefore result in exposure of Fe(III) floc to
499 seasonally fluctuating redox conditions. The material is freshly precipitated, very poorly-crystalline –
500 hence susceptible to reductive dissolution – and contains readily exchangeable-As at concentrations
501 well above those of bulk sediments. As-bearing authigenic pyrite occurs within 12 m of the ground
502 surface at various floodplain sites and close (~5 m) to the current range of seasonal water table
503 fluctuations. While stable under reducing conditions, if there is some regional lowering of water

504 tables, either through prolonged drought, climate-induced shifts in monsoonal precipitation, or excess
505 groundwater abstraction, then these materials may be at risk of exposure and oxidation. Although
506 oxidation of pyrite may simply cause As to shift host-phases and become sequestered in the resulting
507 Fe(III) oxides (Polizzotto et al., 2006), it is conceivable there could be consequences for mobilizing
508 additional As in the aquifer, especially in the short-term.

509

510

511

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

513

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

524

525

526 **Gagri filters**

527 One of the first filters employed was the two Gagri filter system with chemical powder. The
528 system, consisting two earthen pots (Nepali language: gagri), uses chemical powder (a mixture of
529 FeCl_3 , NaOCl and charcoal). Ferric chloride is the compound that removes arsenic present in
530 affected water. The candle filter aids in filtration of the coagulants formed in the upper pot. The
531 second pot underneath the first one receives water free from arsenic, iron, bacteria and odour. This
532 system is 90% efficient in removing arsenic and is below the Nepal interim standard. Further
533 development led to the three gagri filter system. This filter replicates the three kulsi system of
534 Bangladesh and solves the problem of chemical powder. Oxidation, adsorption, precipitation and
535 filtration are the process for removal of arsenic and iron in this filter. This filter system can remove
536 up to 95% of the arsenic, even when the water is highly contaminated. Retardation of filtration
537 process due to clogging and presence of microbes in the treated water limits the filter's performance.
538 Therefore, techniques for improvement of microbiological quality should also have been used while

539 providing this option (Shrestha et al., 2004a). The 3-Gagri filter is a water container made of copper,
540 brass, steel, tin, and or clay pot. The Three-Gagri Filter unit consists of three clay pots staggered
541 vertically with a 1 cm in diameter hole in the bottom of the middle and top filters. The top and
542 middle filters work as a reactor, and the bottom filter stores the treated water. The top filter contains
543 the following, from bottom to top: a layer of polyester cloth, 3 kg of iron nails (3 cm depth), 2 kg of
544 coarse sand (4 cm depth) and raw water. The middle filter contains the following from bottom to top:
545 a layer of polyester cloth, about 50 kg of brickbats, 2 kg of fine sand (3.5 cm depth), 1 kg of charcoal
546 (6 cm depth), 2 kg of brickbats (3 cm depth), and filtered water from the top filter. This filter could
547 remove 95–99% of arsenic but there were problem with high iron in treated water and filter clogging
548 due to bacterial growth. This filter was quickly replaced by arsenic biosand filter (Neku and
549 Thandukar (2003); Thakur et al. (2011)).

550

551 **Kanchan filters**

552

553 These iron-assisted bio-sand filters were constructed on the basis of arsenic removal from water
554 using zero-valent iron (ZVI) media. Under conditions applicable to drinking water treatment,
555 arsenate removal by zero-valent iron media involves surface complexation only and does not involve
556 reduction to metallic arsenic. Under the pH and redox conditions of most groundwaters and surface
557 waters, dissolved arsenic exists as the As(V) (arsenate) species, H_2AsO_4^- and HAsO_4^{2-} , and the
558 As(III) (arsenite) species, H_3AsO_3^0 and H_2AsO_3^- . Removal of arsenic occurs through adsorption and
559 coprecipitation during the formation of iron(III) hydroxides. However, acceptable levels of removal
560 are achieved only when there is a filtration step to remove colloidal arsenic (Farrell et al., 2001).
561 Greater attention is required for the removal of As(III) from groundwater due to its higher toxicity
562 and mobility, which mainly arise from its neutral state (H_3AsO_3^0) in groundwater as compared to the
563 charged As(V) species (H_2AsO_4^- and HAsO_4^{2-}), which predominate near pH 6–9. The As(III)
564 removal mechanism is mainly due to spontaneous adsorption and coprecipitation of As(III) with
565 iron(II) and iron(III) oxides/hydroxides, which form in-situ during ZVI oxidation (corrosion).
566 Heterogeneous reactions at the corroding ZVI surface are complex and result in a variety of potential
567 adsorption surfaces for As(III) and As(V). Evidence has been presented showing that As(III) can be
568 removed by adsorption on NZVI (nonoscale zero valent iron) in a very short time (minute scale) and
569 is strongly adsorbed on NZVI over a wide range of pH and anion environments (Kanel et al., 2005).
570 Investigations by Neumann et al. (2013) regarding the SONO household filters used in Bangladesh
571 (an other version of an iron-assisted bio-sand filter) showed that over 95% of the As passing the top
572 sand layer was removed in the CIM (composite iron matrix) by sorption, coprecipitation, and

573 incorporation into solids formed during the corrosion of ZVI. The continued presence of dissolved
574 Fe(II) in the CIM appears to be important for the long-term operation of the filters. While young CIM
575 contained large fractions of As in amorphous or poorly crystalline phases, magnetite was dominant in
576 older CIM, consistent with an invariably deep black color. The transformation of As-rich
577 Fe(III)(hydr)oxides into magnetite is important for two reasons: (i) the much denser magnetite does
578 not lead to clogging of the filter and (ii) magnetite is more stable toward dissolution than freshly
579 formed amorphous phases and leaches less As during milder extraction steps. Leaching tests with
580 spent CIM in a previous study have shown very low remobilization of As, rendering used CIM
581 nonhazardous. Because As is removed predominantly in the CIM, the other filter components such as
582 sand, brick chips, and the plastic components can be disposed without special care.

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

597 The Kanchan Arsenic Filter (KAF), an award-winning household water filter, was constructed for
598 simultaneous arsenic and pathogen removal. The KAF is constructed using locally available labour
599 and materials and is optimized based on the local socio-economic conditions. The Kanchan Arsenic
600 Filter combines the concept of a slow sand filter for intermittent use (i.e. a biosand filter base) with
601 the innovation of a diffuser basin containing (rusty) iron nails for arsenic removal. Operating under
602 the water quality conditions encountered in the Terai region of Nepal (total arsenic <500mg/L,
603 phosphate < 2 mg/L, pH < 8) the iron nails can last 3 years before replacement is necessary (Ngai et
604 al. 2006). A two-year technical and social evaluation of over 1000 KAFs deployed in rural villages of
605 Nepal determined that the KAF typically removes 85–90% arsenic, 90–95% iron, 80–95% turbidity,
606 and 85–99% total coliforms. Then 83% of the households continued to use the filter after 1 year,
607 mainly motivated by the clean appearance, improved taste, and reduced odour of the filtered water, as

608 compared to the original water source. In the KAF, non-galvanized iron nails are exposed to air and
609 water, rusting quickly and producing ferric hydroxide on the iron nails' surface. When arsenic-
610 containing water is poured into the filter, arsenic is rapidly adsorbed onto the surface of the ferric
611 hydroxide. This mechanism is similar to arsenic adsorption on zero-valent iron and arsenic
612 adsorption on hydrous ferric oxides. Some arsenic-loaded iron particles in the KAF are flushed on to
613 the sand layer below, and are trapped in the top few centimeters of the fine sand due to straining. As
614 ferric hydroxide particles “exfoliate” from the iron nails, new iron surfaces are created, providing
615 additional arsenic adsorption capacity. A Kathmandu university study found that iron and arsenic do
616 not migrate through the sand media over time (Ngai et al. 2007). The filter container can be
617 constructed out of concrete or plastic. The container is about 0.9 m tall 0.3 m in diameter (Fig. 2).
618 The container is filled with layers of sieved and washed sand and gravel. There is a standing water
619 height of 5 cm above the sand layer. The diffuser basin is filled with 5 to 6 kg of non-galvanized iron
620 nails for arsenic removal. In addition, pathogens, iron and suspended material are removed from
621 water through a combination of biological and physical processes: mechanical trapping,
622 adsorption/attraction, predation and natural death. This filter can treat approximately 10–15 L/h of
623 arsenic contaminated water. The filters are locally available at a cost of about 1,400 to 1,800 NRs
624 (about US\$20) per filter (Thakur et al., 2011). Fig. 3 exhibits one of this operating filters in the
625 district Nawalparasi in October 2015. This zero-valent iron (ZVI)-based filters are able to remove
626 arsenic and other pollutants from drinking water, but their performance depends on the form of ZVI,
627 filter design, water composition and operating conditions. Kanchan filters use an upper bucket with
628 ZVI in the form of commercial iron nails, followed by a sand filter, to remove arsenic and pathogens.
629 Wenk et al. (2014) evaluated factors that influence the removal of arsenic and uranium with
630 laboratory columns containing iron nails with six different synthetic groundwater at pH 7.0 and 8.4
631 over 30 days. During the first 10 days, As removal was 65–95 % and strongly depended on the water
632 composition. As removal at pH 7.0 was better than at pH 8.4 and high P combined with low Ca
633 decreased As removal. From 10–30 days, As removal decreased to 45–60 % with all columns.
634 Phosphate, in combination with low Ca concentrations lowered As removal, but had a slightly
635 positive effect in combination with high Ca concentrations. The drop in performance over time can
636 be explained by decreasing release of iron to solution due to formation of layers of Fe^{III} phases and
637 calcite covering the iron surface. Mobile corrosion products contained ferrihydrite, Si-containing
638 hydrous ferric oxides, and amorphous Fe–Si–P phases. Comparisons with another type of ZVI filter
639 (SONO-filter, see Hussam and Munir, 2007) were used to evaluate filter design parameters. Higher
640 ZVI surface areas and longer contact times should lead to satisfactory As removal with Kanchan-type
641 filters. Economical and promising methods are co-precipitation of As with naturally occurring or
642 added Fe^{II} or Fe^{III}, sorption or adsorption to inexpensive prefabricated sorbents or ion exchangers, or

643 As removal with metallic iron. Phosphate interacts strongly with precipitating Fe^{III} (hydr)oxides and
644 outcompetes arsenic for sorption and incorporation, such that additional iron is necessary to remove
645 both arsenic and phosphate. Removal units using metallic zero-valent iron (ZVI) are promising for
646 several reasons: (1) ZVI filters can be constructed with locally available materials (typically sand and
647 iron in various forms such as turnings, filings, nails or cleaned scrap iron). Corroding iron can
648 potentially produce the largest amount of As-sorbing iron^{III} (hydr)oxides per mass of starting
649 material. Aerobic iron corrosion leads to oxidation of As^{III} to the more strongly sorbing As^V, without
650 the need of added oxidants. Two measures that could improve the performance of Kanchan filters are
651 (1) larger specific ZVI surface areas (e.g. by use of smaller nails) and (2) increased contact times by
652 more controlled and restricted flow from the upper diffuser bucket. As Singh et al. (2014) state in
653 their article, the Kanchan filters efficacy in field conditions operating for a long period has been
654 scarcely observed. They observed the efficacy of KAFs running over 6 months in highly arsenic-
655 affected households in Nawalparasi district. Of 62 tubewells, 41 had influent arsenic concentration
656 exceeding the Nepal drinking water quality standard value (50 µg/L). Of the 41 tubewells having
657 unsafe arsenic levels, KAFs reduced arsenic concentration to the safe level for only 22 tubewells, an
658 efficacy of 54%. In conclusion, they did not find significantly high efficacy of KAFs in reducing
659 unsafe influent arsenic level to the safe level under the in situ field conditions.

660

661

662 **Summary and future perspective**

663

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

676

677

678 **Acknowledgements**

679

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

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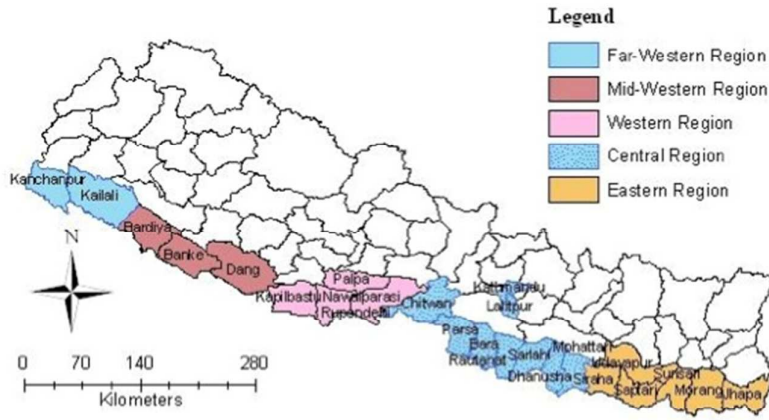
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915 Fig. 1. Groundwater arsenic tested districts in various development regions of Nepal (from Thakur
916 et al., 2011).

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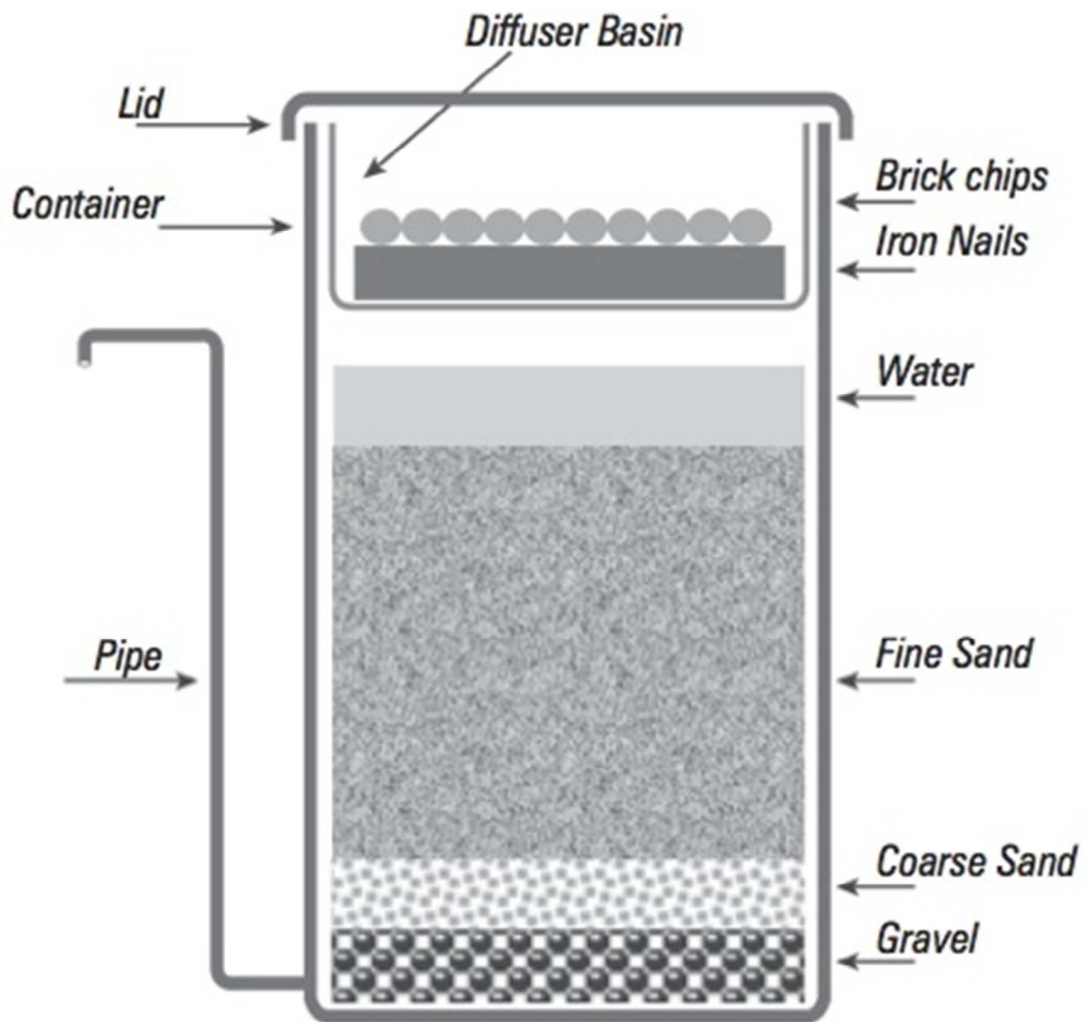


Groundwater Arsenic Tested Districts of Nepal

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923 Fig. 2. Diagram of the KAF, showing the location and arrangement of its components. Source:
924 Ngai et al. (2005).
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930 Fig. 3. Operating filter in Nawalparasi district, Nepal, October 2015. Photo: B. Mueller.
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