Kanchan arsenic filters in the lowlands of Nepal – mode of operation, arsenic removal and future improvements

Barbara Mueller [a,e], Bipin Dangol [b], Tommy K. K. Ngai [c], Stephan J. Hug [d]

[a] *Bamugeobiochem, Horbenstrasse 4, 8356 Ettenhausen, Switzerland*

[b] *ENPHO, Environment and Public Health Organization, New Baneshwor, Kathmandu, Nepal*

[c] *CAWST, Center for Affordable Water and Sanitation Technology, B12, 6020 – 2Street SE, Calgary, Alberta, T2H 2L8, Canada*

[d] *Eawag, Swiss Federal Institute of Aquatic Science and Technology, Ueberlandstrasse 133, CH-8600 Duebendorf, Switzerland*

[e] Corresponding author. Email: bar.mueller@unibas.ch

Abstract

In the lowlands of Nepal (Terai), the WHO drinking water guideline concentration of 10 µg/L for arsenic (As) is frequently exceeded. Since their introduction in 2006, iron-assisted bio-sand filters (Kanchan filters) are widely used to treat well water in Nepal. The filters are constructed on the basis of As-removal with corroding zero-valent iron (ZVI), with water flowing through a filter bed of iron nails placed above a sand filter. According to several studies, the performance of Kanchan filters varies greatly and depends on the size of the iron nails, filter design, water composition and operating conditions, leading to concerns about their actual efficiency. This study examined 38 Kanchan household filters for which insufficient As-removal was reported, to evaluate the reasons for limited removal efficiency and to define measures for improved performance. The measured arsenic removal ranged from 6.3% to 98.5 %. The most relevant factors were the concentrations of As and Fe in the raw water, with the best removal efficiency observed for water with low As (124 µg/l) and high Fe (4.94 mg/l). Although the concentrations of other elements, pH, flow rates, and contact time with ZVI also played a role, the combined evidence indicated that the reactivity of the frequently drying nail beds between filtrations was insufficient for efficient As-removal. Optimized filters with added top layers of sand and raised water outlets with flow restrictions to keep nails permanently immersed and to increase contact times, should be able to achieve higher and more consistent arsenic removal efficiencies.

Keywords: arsenic, arsenic contamination, Kanchan filters, efficiency

Introduction

In several countries of South East Asia (e.g. Bangladesh, India, Nepal, Myanmar, and others) contamination of groundwater by arsenic (As) causes adverse health effects as soon as the groundwater is used as drinking water for longer time periods. The current drinking water guideline of the World Health Organization (WHO) for As was set to a value of 10 µg/L. Changes in groundwater parameters such as pH, redox conditions, temperature and solution composition favor the release of As from solid phases in aquifers. In contrast to Bangladesh, the severity of this problem in Nepal was only addressed after 1999, when a first report was published (Sharma 1999). According to several publications (e.g. Bhattacharya *et al.* 2003; Neku and Tandukar 2003; Emerman *et al.* 2010; Thakur *et al.* 2011) residents of all 20 Terai (lowlands of Nepal) are at risk by consumption of As-contaminated water.

So far, 1.73 % of 1.1 million tube wells tested exhibited As concentration above the Nepal drinking water standard of 50 ppb, and in 5.37 % of these tube wells, As concentrations exceed the WHO guideline value of 10.0 µg/L. The district of Nawalparasi is by far the most affected by the arsenic crisis (NASC-NRCS; National Arsenic Steering Committee/National Red Cross Society, 2011). Hence it is imperative to decrease the concentrations of As in drinking water by either avoiding contaminated sources, or by appropriate water treatment where sources of safe drinking water are not available.

Geological situation of the Terai region and arsenic contamination of groundwater
Nepal's geography represents one of the most extreme on this planet: Squeezed between Tibet to the north and India to the south, east and west, it exhibits a predominantly mountainous structure. The Nepal Himalayas - the most prominent mountain chain of the country - is built up by different tectonic units including a broad range of various rocks of metamorphic, sedimentary, and igneous in origin. At least some of the groundwater arsenic

74　　heterogeneity we find in the foreland and delta is caused by differential erosion of these

75　　rocks (i.e. Gurung *et al.* 2005; Shah 2008; van Geen *et al.* 2008; Guillot *et al.* 2015).

76　　　　The southern lowlands of Nepal (the so called Terai) represents an active foreland basin

77　　including Quaternary sediments (molasse along with gravel, sand, silt, and clay). The vast

78　　ground water reservoirs are replenished by heavy monsoon precipitation and snow-fed river

79　　systems (Sharma 1995; Gurung *et al.* 2005).

80　　　　By far the most intensively studied Terai province regarding local geology and arsenic

81　　contaminated ground water is Nawalparasi. This district is part of the Terai plain which

82　　presents the continuation of Indo-Gangetic plain (Figure 1). The major river of this district

83　　(Narayani) having its origin in the Higher Himalayas exerts a major influence on the

84　　unconsolidated Holocene fluvial deposits. Small natural ponds and meandering rivers were

85　　delineated as characteristic geomorphic features of the area (Shrestha *et al.* 2004). According

86　　Guillot *et al.* (2015) the mostly fine grained sediments here include sands, silt and clay

87　　containing micas. In the areas with fine-grained sediments, elevated concentrations of As are

88　　typically recorded (Brikowski *et al.* 2004, 2014; Diwakar *et al.* 2015; Mueller & Hug 2018).

89　　The usually thin alluvial aquifers of the Nawalparasi district are known to be severely

90　　contaminated by As which is predominantly incorporated in finer particles like clay minerals

91　　(see Mueller 2017; Mueller & Hug, 2018). Considerable As leaching rates point out that the

92　　Terai sediments have high potential for arsenic release, and that pH and redox conditions

93　　favor the mobilization of As (Gurung *et al.* 2005; Mueller & Hug 2018).

94　　　　Nickson *et al.* (2000) described that As in the groundwater seems to be derived from

95　　reductive dissolution of As-rich Fe(III)hydr(oxides), driven by microbial degradation of

96　　sedimentary organic matter. Brikowski *et al.* (2014) mentioned differences in ground water

97　　composition regarding Fe and As concentration in pre- and post-monsoon seasons. Guillot *et*

98　　*al.* (2015) stated that As is concentrated in clayey sediments and is mostly associated with

99　　some specific elements (Fe, Al, K, and C), suggesting that biotite contributed to the

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sequestration of this element. Mueller & Hug (2018) also concluded that alumosilicates such as clay minerals represent a substantial source of As in Nepal. The apparent decoupling of the concentrations of Fe and As and the positive correlation between concentrations of As and Na, K in the ground water strongly indicate that clay minerals host a substantial quantity of arsenic (Mueller & Hug, 2018).

The current mitigation option in Nepal: Kanchan filters

The currently most used water treatment option in Nepal are the so called Kanchan filters. Kanchan filters were initially developed and installed in Nepal as a joint venture between Massachusetts Institute of Technology (MIT), Environment & Public Health Organization (ENPHO) and Centre for Affordable Water and Sanitation Technology (CAWST) (NRCS– ENPHO, 2003; Ngai and Walewijk, 2003; Ngai *et al.* 2005, Ngai *et al.* 2006; Ngai *et al.* 2007). The filters were designed to remove pathogens by physical straining and by predation of pathogens in the lower sand layer, as well as to eliminate arsenic by sorption on Fe(II,III)(hydr)oxide phases produced from corrosion of ZVI in the form widely available iron nails, placed in a perforated bucket above the sand layer, see Figures 2 and 3. First field tests indicated that Kanchan filters removed more than 95% arsenic on average (NRCS– ENPHO (2003); Ngai and Walewijk 2003). The microbiological quality of the treated water was declared satisfactory (Shrestha *et al,* 2004a). Ngai *et al.* (2006) estimated that in respect of the water quality conditions encountered in the Terai region of Nepal (total arsenic <500µg/L, phosphate < 2 mg/L, pH < 8) the iron nails can last 3 years before replacement is necessary. Yet, Chiew *et al.* (2009) found later that the performance of Kanchan filters installed in Cambodia was insufficient. Sing *et al.* (2014) later also reported that merely half of the Kanchan filters analyzed removed As to drinking water level standards and noted that the long-term performance of Kanchan filters in Nepal has rarely been tested. After six months of observing Kanchan filters in highly arsenic-affected households in Nawalparasi district, Sing et al. (2014) found that out of 62 tube wells, 41 had influent As concentrations

exceeding the Nepal drinking water quality standard value (50 µg/L). Out of the 41 tube wells

having unsafe As levels, KAFs reduced arsenic concentrations to the safe levels for only 22

tube wells. More recent surveys in Nepal also found that water treated with Kanchan filters

had effluent As concentrations exceeding the Nepal drinking water quality standard value (50

µg/L) (see Mueller 2017: Mueller and Hug, 2018). In a recent study, Ogata et al. (2020),

assessed the performance and long-term sustainability of 2833 Kanchan filters distributed in

Nepal. In a statistical analysis of the data, these authors found that treating water with average

concentrations of 92 µg As/L and 3.2 mg Fe/L, the filters reached an average As-removal

efficiency of 75%,. The main factors determining the removal efficiency were the

concentrations of As and Fe in the raw water, with filters generally performing better in the

first four years than after four to eight years use. The decreasing removal efficiency with time

was attributed to a loss of activity of the iron nails surfaces.

Removal of As with zero-valent iron media functions by formation of Fe(II) and

various Fe(II,III)(hydr)oxide phases by corrosion of ZVI material. Under the pH and redox

conditions of ground waters in Nepal, dissolved arsenic is predominantly present as As(III)

(H_3AsO_3 and $H_2AsO_3^-$), but in some wells, fractions up to 67 % As(V) ($H_2AsO_4^-$ and

$HAsO_4^{2-}$) were measured in this study. Corrosion of ZVI with oxygenated water leads to

formation of oxidizing species (e.g. H_2O_2, $^•OH$, Fe(IV)) that in turn leads to partial or

complete oxidation of As(III) and to adsorption and incorporation of As(V) into

Fe(II,III)(hydr)oxides (Katsoyiannis et al. 2008). Colloidal Fe-oxide particles with adsorbed

As that can pass the upper ZVI-filter are retained in the sand filter below (see e. g. Farrell et

al. 2001). As ferric hydroxide particles tend to "exfoliate" from the iron nails, new

Fe(III)(hyr)oxides are continuously formed at the ZVI surface, providing a long lasting

arsenic adsorption capacity.

Due to the necessary reaction of oxygenated water with Fe(0), the performance of

filters with ZVI depends on the form of ZVI (in this case the form and size of iron nails), filter

design, water composition and operating conditions. So far the most successful and widely

distributed filters using ZVI are the SONO-filters developed in Bangladesh (Hussam and

Munir 2007; Neumann *et al.* 2013). SONO filters use a layer of submillimeter and millimeter

sized iron particles embedded between two layers of sand. The small iron particles in SONO

filters provide more active surface area per weight and volume than iron nails, which might be

a reason for the overall good performance of SONO filters. However, the specially produced

iron particles used in SONO-filters are not available outside of Bangladesh, while iron nails

are globally available and inexpensive. Filters using iron nails or similarly available forms of

ion are thus of continued interest. Wenk *et al.* (2014) evaluated factors influencing the

removal of arsenic and uranium with iron nails in laboratory columns with six different

synthetic groundwaters at pH 7.0 and 8.4 over 30 days. During the first 10 days, As removal

increased up to 65–95 % but strongly depended on the water composition. From 10–30 days,

As removal decreased to 45–60 % with all six groundwaters. More recently, Bretzler et al.

(2020) tested different filters with iron nails embedded between two layers of sand (similar to

SONO-filters) and found 60–80% arsenic removal in field filters in Burkina Faso with input

concentrations of 400–1350 μg As /L and <0.1 mg Fe/L. Tests with filter columns with

reduced dianmeters in the laboratory showed >95% removal with inflow concentrations of

500-1000 ug/L for a duration of several months.

To investigate reasons for the insufficient performance of Kanchan filters, co-workers from

CAWST in cooperation with ENPHO and Eawag, Switzerland, started two sampling

campaigns for ground water in Nawalparasi in October 2015 (post-monsoon) and in April

2017 (pre-monsoon) and analyzed As and other elements in raw groundwater and in water

treated with Kanchan filters. The aim of this study was to measure the As removal

efficiencies of 38 selected Kanchan filters with reported insufficient removal within and

around the municipality of Ramgram (capital of the district of Nawalparasi), and to find the

178 reasons for limited of arsenic removal. A special focus was to inspect single filters in detail

179 and to analyze main and trace elements in the untreated (raw) ground water and in treated

180 (filtered) water. The overall goal are future improvements in the design of the filters leading

181 to higher arsenic removal efficiencies.

182

Material and Methods

Study site

185 Ground water samples were collected from privately owned hand pumps within the urban

186 area of Ramgram and in the villages Manari, Panchanagar, Sukauli and Tilakpur (within a

187 distance of 10 km of Ramgram). Generally, the depth of the tube wells did not exceed 25 m

188 and the soil is mostly comprised of clayey sediments. Small natural ponds and rivers are

189 found as typical geomorphic features of the area. As decribed above, the clayey sediments

190 are dominated by quartz, biotite, muscovite, K-feldspar, calcite and dolomite as major phase

191 and garnet, zircon and monazite as minor constituents (Guillot *et al.* 2015).

Collection of samples

193 Two series of 38 water samples from hand pumps were collected in post-monsoon 2015 and

194 again in pre-monsoon 2017. Pumps were well flushed (e.g. pumped for approx. 2 min,

195 corresponding to replacement of roughly twice the volume of water in the tubing (see text S1,

196 Supplementary Material) before sample collection to remove all standing water in the tube

197 wells (see Ngai *et al.* 2006 and 2007). Private ground water pumps for sample collections

198 were chosen randomly according a register provided by ENPHO for wells from which

199 filtered water exceeded the Nepal drinking water quality standard value (50 µg/L) in previous

200 measurements. Water samples from the field were collected into 4.5 mL pre-acidified

201 polypropylene vials (containing 150 µl 1M HNO_3) and were sent to Eawag for further

202 examination. To determine As(III) and As(V), non-acidified water samples were filtered

203 through metalsoft arsenic speciation cartridges specifically adsorbing As(V). All samples

204 were analyzed by ICP-MS (Agilent 7500cx, Agilent Technologies, Waldbronn

205 , Germany) at Eawag, after 1:5 or 1:20 dilution with 1% HNO_3. Each measurement was

206 conducted in duplicate. Quality control of the ICP-MS measurements was conducted with

207 duplicate and triplicate samples of multi element reference samples (Standard Solution

208 32195.2000 BKR, Stehelin AG, CH-4142 Muenchenstein, Switzlerland), diluted at various

209 concentrations in distilled water and in synthetic groundwater of similar composition as the

210 groundwater in Nepal. Results were also regularly cross-checked with ICP multi-element

211 standard solution X 109493 (Merck Millipore). Detection limits in undiluted samples after

212 dilution were < 2µg/L for As and other trace elements, and < 0.2 mg/L for Fe and Mn. All

213 ICP-MS determinations agreed to within 3–5% standard deviation.

214

215 **Results and Discussion**

216 **As and other elements in groundwater and filtered water**

217 Figure 4A illustrates the concentrations of arsenic in groundwater collected from 38 different

218 tubewells without treatment, after passing through the nail bed, and after additionally passing

219 the sand layer in the complete filtration process with Kanchan filters. Figure 4B shows the

220 corresponding arsenic removal efficiencies (values and the localities of the wells are listed in

221 the Supplementary Material Table S1). The sample designation SN refers to the sampled

222 filter number and the description of the filter indicated at the time of installation. Only 2 of

223 the 38 selected filters were capable of removing arsenic below the WHO guideline of 10

224 µg/L, and only 7 of 38 filters reached the Nepal interim standard of 50 µg/L. The overall

225 removal efficiency after full filtration ranges from 6.3 % (SN15) and 98.5 % (SN9). In

226 several of the Kanchan filters, the arsenic concentrations were higher after passing the sand

227 layer than measured just past the nail bed. These are indications that the fine sand has to be

9

228 cleaned or replaced after a certain time of usage in order to remove exfoliated particles from

229 the nail bed from the sand layer, as these particles can release As again into solution under

230 possibly reducing conditions in the biofilm on top of the sand layer.

231   Figure 4C shows the molar concentrations of Fe, As and P in the untreated tubewell water.

232 Different from the situation in Bangladesh (Table 1), concentrations of P (predominantly

233 phosphate) are low and in the same range as As-concentrations and thus do not significantly

234 lower As-removal. Interestingly, there is a positive correlation between As and the lithophile

235 elements Na, B and Mo, and a negative correlation between Fe and As in the untreated

236 groundwater (Figure S1), showing the decoupling between As and Fe, as described in

237 Mueller and Hug (2018). In contrast, there is the expected positive correlation between As-

238 removal and Fe concentrations (Figures 5 and S2A).

239   Figure 5 shows the fractions of As removal as a function of Fe in the raw groundwater

240 (full and empty blue circles), and the expected fractions of As-removal by aeration and co-

241 precipitation with naturally present Fe(II) according to a co-precipitation model described

242 previously (Roberts *et al.* 2003) (black triangles, red squares and red diamonds). The model

243 assumes oxidation of Fe(II) to a Fe(III)-phase with $[Fe(II)]_{initial}/1.40$ adsorption sites, to

244 which silicate, phosphate, As(III) and As(V) adsorb competitively with sorption constants

245 $\log(K_{Si})$, $\log(K_P)$, $\log(K_{As(V)})$, $\log(K_{As(III)})$ of 2.70 5.80, 5.65, 3.67 in groundwater with pH 7

246 and typical Mg and Ca- concentrations. The red connected diamonds show the optimal

247 calculated removal efficiency that could be achieved if all As were present as As(V) (or if all

248 As(III) were oxidized before Fe(II) oxidizes and Fe(III) precipitates). The black connected

249 triangles show the calculated removal efficiency if all As is present as As(III) and no

250 oxidation occurs during the oxidation of Fe(II). The red empty squares show the calculated

251 As-removal accounting for initially present fractions of As(V) and for co-oxidation of As(III)

252 with the oxidation of Fe(II) as described previously (Hug & Leupin 2003; Roberts *et al.*

253 2003). The empty blue circles show the measured removal. The results shown in Figure 5

254   illustrate that arsenic removal in the investigated filters can be explained by removal of As

255   with the naturally present iron alone. The ability of the filters to partly oxidize As(III) and/or

256   to release more Fe to remove un-oxidized As(III) may have contributed to the As-removal,

257   but it did not lead to significantly better As removal than what can be expected with the

258   naturally present Fe in the groundwater in the tested filters. This finding indicates that the

259   iron nails are not sufficiently reactive to provide additional iron for efficient As-removal.

260   Figure 6a and more clearly Figure 6b depict the dependence of the removal efficiencies on

261   the contact time of the water with the iron nails. Contrary to expectations, even filters with

262   the longest contact times did not remove arsenic to acceptable levels. Although the contact

263   time is expected to be a crucial factor in the performance of As-removal filters (Wenk *et al.*

264   2014), an increase of the contact time to 30 min did not lead to satisfactory As-removal.

265   Additional changes in the filter design will most likely be required.

266   Various major and trace elements appeared to influence the removal efficiencies. The

267   dependence of As-removal on the concentrations of Fe, Na, P, B and Mo in raw water is

268   shown in Figure S2. A clear positive correlation is only observed with Fe. Higher

269   concentrations of Na, B and Mo appear to lower the adsorption of As. However, the negative

270   influence can be explained by the negative correlations between Fe and Na, B, and Mo in the

271   raw groundwater (Figure S1).

272   Although P (phosphate) strongly competes with the adsorption of As(III) and As(V) (Dixit

273   and Hering 2003; Roberts *et al.* 2004; Tyrovola *et al.* 2006; Wenk *et al.* 2014; Senn *et al.*

274   2018), P-concentrations did not show a clear influence (Figure S2C). Phosphate

275   concentrations in ground water in Nepal are relatively low (0.02 to 0.89 mg/L in

276   Nawalparasi) and the effect of phosphate is thus limited. In their report Ngai *et al.* (2007)

277   state that, based on a study from Meng *et al.* (2002), phosphate interference on arsenic

278   adsorption on Fe(III)hydr(oxides) starts at about 2 mg/L – concentrations by far higher than

279   concentrations of phosphate in the ground water from Nawalparasi. Furthermore, in ZVI

280   filters, phosphate has varying effects on adsorption and corrosion. Wenk *et al.* (2014) used

281   phosphate concentrations of up to 2mg P/L and found that although P competes with arsenic

282   for sorption and incorporation, phosphate prevented formation of dense $CaCO_3$ layers on ZVI

283   which potentially slow corrosion rates. Smith *et al.* (2014) showed in their field study that the

284   influent concentration of 0.45 mg P/L positively influenced the removal rate in their filters.

285   Within our range of P concentrations, the influence of phosphate on arsenic removal was

286   small.

287   Figure S3 again illustrates the dependence of the concentration of arsenic after complete

288   filtration on the concentration of Fe in the raw water. An elevated concentration of Fe in raw

289   water was essential to obtain acceptable removal efficiencies of As by non-iron-assisted sand

290   filters working with naturally present Fe(II), as is the case in Vietnam (Berg *et al.* 2006). In

291   Nawalparasi, we found much lower Fe/As ratios in the groundwater compared to other

292   countries affected by the arsenic crisis (Table 1). Conversely, the contents of P in ground

293   water in Nawalparasi are rather low and the filters should perform well if the corrosion of the

294   iron nails would be an efficient source of additional Fe for As-removal.

295   **Design of different ZVI-filters and As-removal efficiencies**

296   Groundwaters in Bangladesh also frequently have low Fe-concentrations and high

297   concentration of As and P. Meng *et al.* (2002), Roberts *et al.* (2004), Hug et al. (2008), and

298   Chiew *et al.* (2009) found that Fe/As molar ratios of >54 are needed for effective As-

299   removal from ground water with high concentrations of phosphate, silicate and carbonate. In

300   Cambodia, with concentration of As from 146 – 372 µg/L) and P above 0.5 mg/L, Fe

301   concentrations of 15 – 20 mg/L would be needed for efficient As-removal (Chiew *et al.*

302   2009). Iron concentrations in Nepal are typically much lower, such that additional sources of

303   iron and oxidants are needed to oxidize As(III) and remove As(V) and remaining As(III).

304   The SONO filters (Hussam & Munir, 2007), as mentioned above, work with a layer of

305   millimeter and sub-millimeter sized iron scraps (composite matrix CIM)) located in the

center of the filter between two layers of sand. With these filters, more than 95% of the As

passing the top sand layer was removed in the CIM by sorption, coprecipitation, and

incorporation into solids formed during the corrosion of ZVI (Hussam and Munir 2007;

Neumann *et al.* 2013).   Efficient formation of Fe(II) and Fe(III)(hdyr)oxides by corrosion of

ZVI is thus essential for As-removal. In comparison with the SONO filters, Kanchan filters

provide less ZVI-surface, without increasing the contact time correspondingly (Wenk *et al.*

2014). In addition, the iron nails in Kanchan filters are exposed to air when the filters are not

used. In SONO filters, the CIM layer with iron particles remains anoxic between filtrations,

which leads to formation of magnetite that can incorporate As(III) and As(V) on surfaces and

As(V) in the lattice (Coker *et al.* 2006; Neumann *et al.* 2013).

Factors influencing the performance of the tested Kanchan filters

Table 2 specifies important properties and characteristics of the filters and of the raw water

for best or worst observed performance, respectively. For each important factor, two filters

with the best and the poorest performance are described, in order to obtain insight into the

dependence of the As-removal efficiency on specific variables.

Nail bed and contact time. A deciding factor is the mass, size and distribution of nails in

the nail bed. Not surprisingly, the most efficient filters that contained the highest amount of

small, bright brown, rusty and loosely kept nails include SN9 (4.05 kg, Figure S4), SN65 (5.2

kg), SN21 (4.7 kg) and SN10 (5.0 kg). Poor performers like SN15 (Figure S5) and SN14 also

contain more than 5 kg so the nails mass itself does not seem to play an important role, but

the distribution of nails was not even, leaving flow channels with no contact with the nails.

Nail bed and sand layers of filters SN9, SN21 and SN10 (good performers) were immersed in

water at the time of inspection, and nails and sand were at least wet in filter SN65. The nail

beds of filters SN15 and SN14 (poor performers) were almost kept dry. In order to promote

corrosion of the nails efficiently, the nails should be immersed in water all time. Yet users of

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332 the filters are not aware of this fact and pour water only for filtering, letting the nails run dry

333 between filtrations. A direct inspection of the filter with the highest performance regarding

334 removal efficiency (SN9, Figure S4) in the field revealed uniformly distributed, small, rusty

335 nails in the flat nail bed (no holes between the nails, no clogging of the nails) as well as

336 standing water in the filter (e. g. sand layer was usually covered with water). Another good

337 performer is filter SN65: Nail bed presented similar to SN9 but SN65 was completely

338 cleaned and the sand replaced the day before inspection. SN63 is another good performer

339 (removal efficiency: 92.6 %) with a pH of 7.4, medium residence time, medium As, Fe, Na,

340 B and Mo concentration; the nail bed presented similar to filter SN9 with very fine sand,

341 thick rusty layer compose of Fe(III)hydr(oxides) on top of the sand layer which is essential to

342 deposit exfoliated particles from the nail bed. Other good performers include SN21 (removal

343 efficiency 2: 80.7 %), SN10 (removal efficiency 2: 79.0 %) – these two filters had a short

344 contact time but are fed with water of low pH and low As in raw water, high Fe, low Na, B

345 and Mo. The highest Fe concentration was analyzed in raw water for filter SN45. But this

346 filter is a mediocre performer only (removal efficiency 2: 70.7 %) – the most important

347 reasons is obvious: Low residence time – but medium pH, low As concentration, medium

348 concentration of Na, Ba and Mo; nails were scarcely corroded. The best performers (SN9,

349 SN10, SN21) were located in Manari, SN 63 and SN 65 in Ramgram (see Fig. 4B and Table

350 S1).

351 **pH and concentration of As, Fe and other ions.** Another factor influencing the overall

352 operation of the Kanchan filters is the pH of the ground water (see e.g. Ramakrishna *et al*.

353 2006). The pH of the ground water in Nawalparasi is typically between 7 and 8. Within this

354 pH-range, Fe in the groundwater is negatively and As positively correlated with pH, due

355 more sorption of Fe(II) and less sorption of As(V) in the sediments with increasing pH (see

356 Mueller and Hug, 2018). Wenk *et al.* (2014) mentioned that the As-removal efficiency with

357 nails declined with increasing pH, due a decrease of corrosion rates. Even though the pH of

filter SN9 is the highest measured during the field campaign (7.81) high concentration of Fe

combined with a low concentration of As, Na, B and Mo and a high residence time

counteracted the adverse effect of pH. Poor performers concerning removal efficiency were

filters SN15 (6.3 %) (Figure S5) and SN14 (13.2 %), both located in Manari. The reasons for

this unacceptable efficiency seem apparent: Low residence time (SN15: 4.57 min.; SN14:

15.1 min.); medium pH (7.5); high As concentration (SN15: 353.3 µg/L; SN14: 657.0µg/L);

lowest iron concentration measured for all filters included in this study (SN15: 0.51 mg/L;

SN14: 1.54 mg/L); medium concentrations of Na, B and Mo concentration for filter SN15

but high concentrations of the latter elements for filter SN14.

The factors discussed above reflect both the varying composition of the ground water and

the differences in the construction and maintainance of the filters, such that good and poor

performers were found within the same municipality (Manari). Chiew *et al.* (2009) studied

the efficiency of arsenic in Kanchan-type filters in Cambodia. The overall average arsenic

removal was just 39.4, 74.9, and 45.4 %, respectively. They found that the main reasons for

poor arsenic removal was due to the combination of high influent P (> 0.5 mg/L) and low Fe

(< 5 mg/L) concentrations and that the added iron nails were largely ineffective due to

insufficient contact time with the water. In their report Ngai et al. (2007) state that based on a

study from Meng *et al.* (2002) phosphate interference on arsenic adsorption on

Fe(III)hydr(oxides) starts at about 2 mg/L – concentrations by far higher than contents of

arsenic in the ground water from Nawalparasi. Smith *et al.* (2017) clearly mentioned in their

field study that the design of the filters where crucial to achieve a good performance.

Especially an increased contact time between iron nails and water is imperative in order to

enhance the removal efficiency. Chiew *et al.* (2009) mentioned the limited contact time of

the nails with the water as a major concern regarding the performance of the filters. In all the

filters examined for this study the water passes too quickly through the nail bed presenting a

highly permeable zone. The contact time for formation of sufficient Fe(II) and

384   Fe(II,III)(hydr)oxides by corrosion of ZVI for the oxidation and removal of As(III) is too

385   limited. Inspection of the data from best performing filters (e.g. filter SN9) indicated a

386   contact time of at least 20 min.

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389   **Limitations of Kanchan filters**

390   Beside the low concentration of Fe, short residence time, high pH and high concentrations of

391   As, Na, B, Mo and other trace elements (see Mueller & Hug, 2018), other factors, such as

392   drying of the nail bed instead of a permanent immersion in ground water or poor

393   maintenance limit the proper performance of the filters. As long as the nail bed of the filters

394   remains wet and oxygen free, the continuously formed Fe(II) leads to transformation of

395   Fe(III)hydr(oxides) to black mixed Fe(II,III)-phase solids and to magnetite (Fe_3O_4) with

396   incorporation of As(V). Fe(II,III)(hydr)oxides exhibit a stronger adsorption affinity for As(V)

397   than for As(III) (Neumann et al. 2013, Wenk *et al.* 2014). As described by Noubactep (2018),

398   the in-situ generated Fe(III)hydr(oxides) are larger in volume than the parent Fe(0). This

399   means that the initial porosity of a packed bed containing Fe(0) will be progressively filled

400   and the surface of iron covered with a crust of Fe(III)(hydr)oxides, causing a loss of

401   permeability and reactivity in the filter media. Corroded nails in the filters in the various

402   municipalities in the district Nawalparasi showed a rusty color and were rarely or only partly

403   black (Figure S6) – a good indication for the formation of Fe(III)hydr(oxides), but not

404   magnetite which occupies a smaller volume than Fe(III)hydr(oxides). Figure S5 illustrates

405   an absolute "no-go" concerning maintenance and usage of the Kanchan-filters: An uneven

406   nail bed with holes between the loosely arranged nails, leading to a lower ZVI-surface and

407   reaction time between ground water and nails. An even, regular nail bed is imperative in

408   order to avoid preferential flow therein. Application of a thin layer of fabric and an upper

409   sand layer could help to keep the nails in place. Further investigations are in progess.

410

Future improvements of Kanchan filters

Appropriate measures have to be taken to overcome the above mentioned limitations, mainly

the limited corrosion during the short contact of the nails with water. Additionally, drying of

the nailbed between filtrations, leading to dense brown crusts of iron(III) phases on the

surface of nails (Figure S6), has to be avoided. The limited contact time can be addressed

with lowering of the flow rates by restricting the flow at the outlet.

In SONO filter, the ZVI layer consisting of fine iron scrap is embedded between two

sand layers and develops a black color (due to green rusts and magnetite) instead of brown

iron(hydr)oxides. (Neumann *et al.* 2013). Smith *et al.* (2017) used a modified version of the

biosand filter with nails embedded in sand to achieve acceptable removal efficiencies of

arsenic, most likely due to increased contact time and sustained corrosion. They found

average removal efficiencies of 92% and never below 86%. Smiech *et al.* (2018) found in a

pilot study that filters containing sand mixed with corroding iron matter (CIM-Filter) could

remove arsenic below the WHO guideline concentration of 10 µg/L, for inlet concentrations

above 150 µg/L As. According to Caré *et al.* (2012), for the pH range of natural waters (pH >

4.5), the ZVI medium should be mixed with a non-expansive material (see also Noubactep

2010), but this is not done in the successful SONO filters. Mixing small iron particles (e.g.

iron filings) with sand might be beneficial, but filling the pore space between loosely packed

iron nails with sand leads to a decrease of the pore volume in the filter bed. The recent study

of Bretzler et al. (2020) showed that efficient arsenic removal with constantly immersed beds

of small iron nails in layers of 60 mm thickness between two sand layers can achieve efficient

As-removal from water with high As-concentrations (500-1000 μg/L As(V) or As (III)). In

laboratory experiments with columns of 32 mm diameter, and with controlled flow

conditions, As removal was >95% for a duration of several months. Achieving controlled

conditions in larger field filters was more challenging; In filters of 320 mm diameter in

17

436   Burkina Faso, As-reomval was 60-80% (inflow 400–1350 μg As/L). In transparent columns

437   in the laboratory, we observed that trapped air in the nail bed can significantly lower the

438   contact between water and iron nails and we suspect that trapped air and temporarily

439   uncontrolled fast flow were the cause of lower As-removal efficiencies in the field filters.

440   Based on the observation with SONO-Filters and the nail filters described above, we

441   will attempt to imrove the performance of Kanchan filters in Nawalparasi by various

442   measures. Improvements include a top layer of sand or gravel above the nail bed to protect

443   and support the integrity of the nail bed and achieve a more homogenous flow across the

444   filter. Additionally, an uplift of the water level by raising the outlet of the filter to above the

445   level of the nail bed, and a flow restriction will ensure that the nails will remain immersed

446   between filtrations and that he contact time of water with the nails is be increased. We will

447   separate the sand from the nails with fabric in order to prevent the mixing of nail and sand or

448   gravel and to facilitate the cleaning of the nails, if necessary. As long as the nails remain

449   immersed in oxygen-depleted water between filtrations, most of the brown and voluminous

450   iron(hydr)oxides are expected to be converted to dense magnetite.

451

Conclusion

453   Despite of the currently varying and often insufficient As-removal with ZVI filters using

454   iron nails, ZVI filters are promising for several reasons: (1) Other types of ZVI filters. e.g.

455   SONO filters (Hussam and Munir, 2007) remove As efficiently from groundwater in

456   Bangladesh. (2) ZVI filters can be constructed with locally available materials (typically sand

457   and iron in various forms such as turnings, filings, nails or cleaned scrap iron). (3) As(III) is

458   oxidized to As(V) in properly working ZVI filter without additional oxidants. For lack of

459   alternatives in many countries and regions, further improvement of arsenic removal by

460   optimized ZVI-based As-removal filters is urgently needed.

461   Ongoing investigations show that the most promising approach for improvements of As-

removal filters is a combination of field and laboratory studies. In field campaigns, untreated water, filtered water, and solid samples of the filter materials have to be collected repeatedly, followed by analytical investigations in laboratories. Moreover, inclusion of reports and experiences of local users of the filters in respect of handling and maintenance of the filters are essential. Filters have to be constructed to insure longterm reliability and acceptance, and users have to be instructed properly and repeatedly.

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Tables and Figures

Table 1. Average molar ratio Fe/As and average concentration of P (mg/L) in various countries in South East Asia

	Vietnam (Hanoi area)	Bangladesh	Nawalparasi (Nepal)
Fe/As	60-68	15-91	6.0-9.6
P	0.8	1.9	0.12
	Berg et al., 2008	Achmed et al., 2010 Rahman et al., 2015	Mueller and Hug, 2018

756 *Table 2.* Factors expected to influence the removal efficiency of Kanchan filters, including

757 several trace elements as examples. Removal efficiencies (RE) of the whole filters in %;

758 contact time in min.; concentration of Fe and Na in mg/L, concentration of As, B, Mo in

759 µg/L). Performance mentioned regarding single filters.

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Factor	Range of value	Filter with best values # (Factor) RE	As/Fe	Filters with poor values # (Factor) RE	As/Fe
Removal efficiency 2	6.28 - 98.5 %	SN9 (98.5 %)	123/5.0	SN15 (6.3 %)	393/0.4
		SN65 (93.9 %)	278/2.6	SN14 (14.0 %)	620/1.4
Contact time (min)	2.67 - 29.8	SN72 (29.8) 50.6%	205/2.4	SN13 (2.67) 39.3%	759/1.7
		SN12 (27.2) 48.4%	309/1.9	SN11 (3.84) 49.1%	339/1.6
pH	7.04-7.81	SN45 (7.04) 71.1%	170/5.8	SN9 (7.81) 98.5%	123/5.0
		SN65 (7.05) 93.8%	278/2.6	SN76 (7.59) 7.2%	139/1.2
As (µg/L)	95.5 - 798.7	SN21 (95.5) 80.5%	89/3.5	SN13 (798.7) 39.3%	759/1.7
		SN9 (128.9) 98.5%	123/4.5	SN14 (657.0) 13.2%	620/1.4
Fe (mg/L)	0.51 - 5.91	SN45 (5.8) 71.1%	170/5.8	SN15 (0.51) 6.3%	339/0.4
		SN9 (5.0) 98.5%	123/5.0	SN34 (0.52) 34.7%	389/0.4
Na (mg/L)	19.9 - 70.9	SN9 (19.9) 98.5%	123/5.0	SN68 (70.9) 55.9%	192/1.2
		SN33 (26.9) 59.1%	365/2.6	SN72 (65.5) 50.6%	205/2.4
B (µg/L)	10.7 - 100.8	SN9 (10.7) 98.5%	123/5.0	SN79 (100.8) 72.6%	211/1.6
		SN10 (11.3) 79.2%	168/3.5	SN68 (64.4) 55.9%	192/1.2
Mo (µg/L)	1.03 - 20.2	SN21 (1.03) 80.5%	89/3.5	SN18 (20.2) 37.1%	505/1.3
		SN9 (1.11) 98.5%	123/5.0	SN12 (18.6) 48.4%	309/1.9

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762 Best performing filter: **SN9** (locality: Manari, see Table S1). Raw water with a low As and a

763 high Fe concentration. Low in Na, B and Mo. Long residence time (22.5 min.), but high pH.

764 Raw water from filter **SN65** with a high removal rate exhibits a low pH, medium As (289.9

765 µg) and Fe concentration (2.72 mg/L). Medium in Na (53.9 mg/L), B (56.4 µg/L) and Mo

766 (5.23 µg/L). Medium residence time: 11.42 min.

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Fig. 1. Groundwater arsenic tested districts in various development regions of Nepal (from

Yadav et al., 2011).
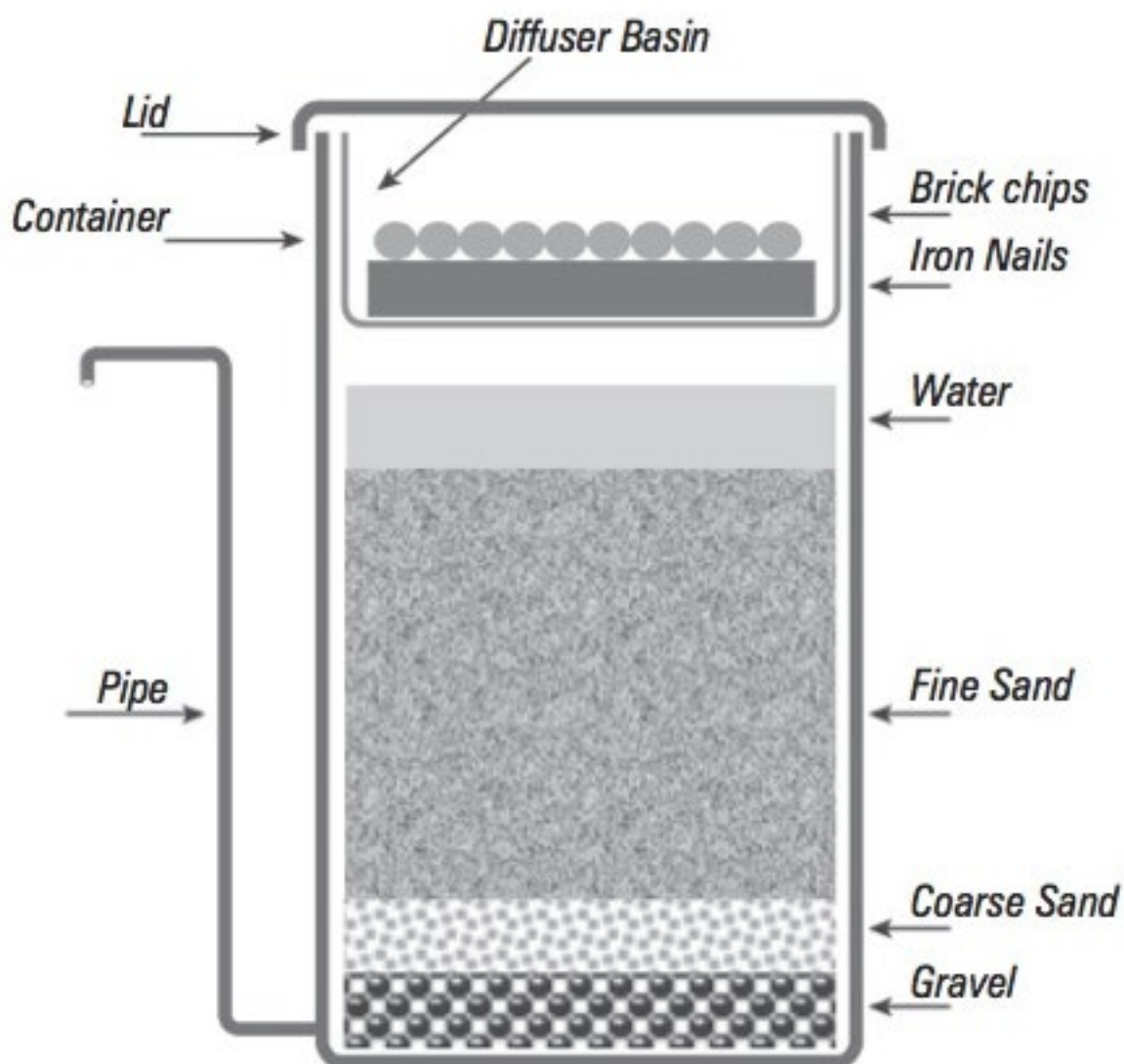
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783   *Fig. 2*. Operating filter (SN54) in Nawalparasi district, Nepal, October 2015. Photo: B.

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789   *Fig. 3*. Diagram of the KAF, showing the location and arrangement of its components.
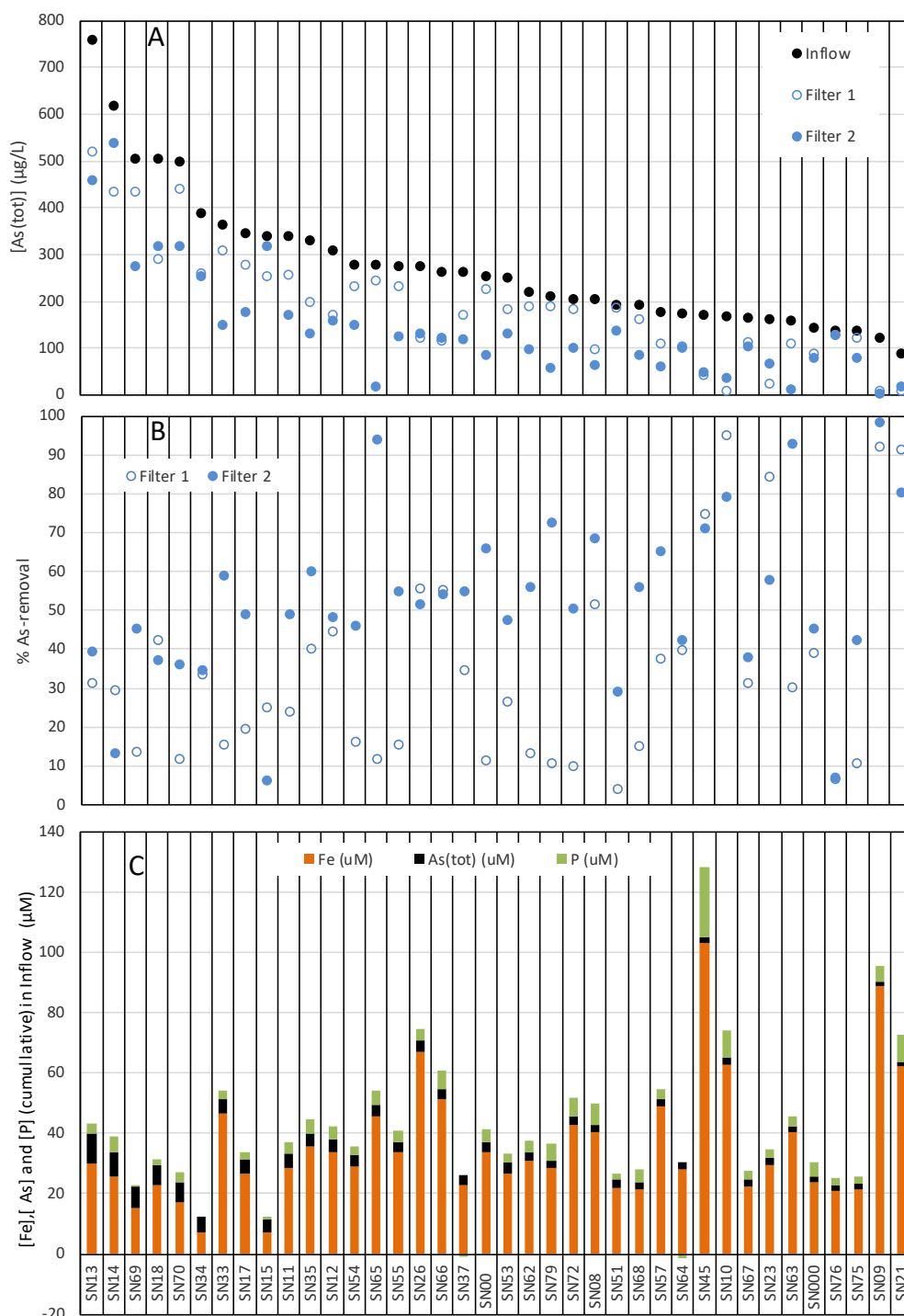
790   Source: Ngai et al. (2005).

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Fig. 4. (A) Concentrations of arsenic in untreated tubewell water (black circles), after passing through the nailbed (empty blue circles) and after the complete filtration process with Kanchan filters (filled blue circles). **(B)** Removal efficiencies (% As-removal) by the nailbed (empty blue circles, filter 1) and by the whole filter (filled blue circles, filter 2). (**C**) Molar concentrations of iron (dissolved Fe(II)), As(tot) (As(III) and As(V)) and of P (phosphate) in the ground water, as indicated in the Figure. Error ranges from duplicate measurements (not shown for clarity) were ±5%. Localities: SN00-SN23 Manari, SN26-SN35 Panchanagar, SN45-SN72 Ramgram, SN75-SN76 Sakauli, SN79 Tilakpur.

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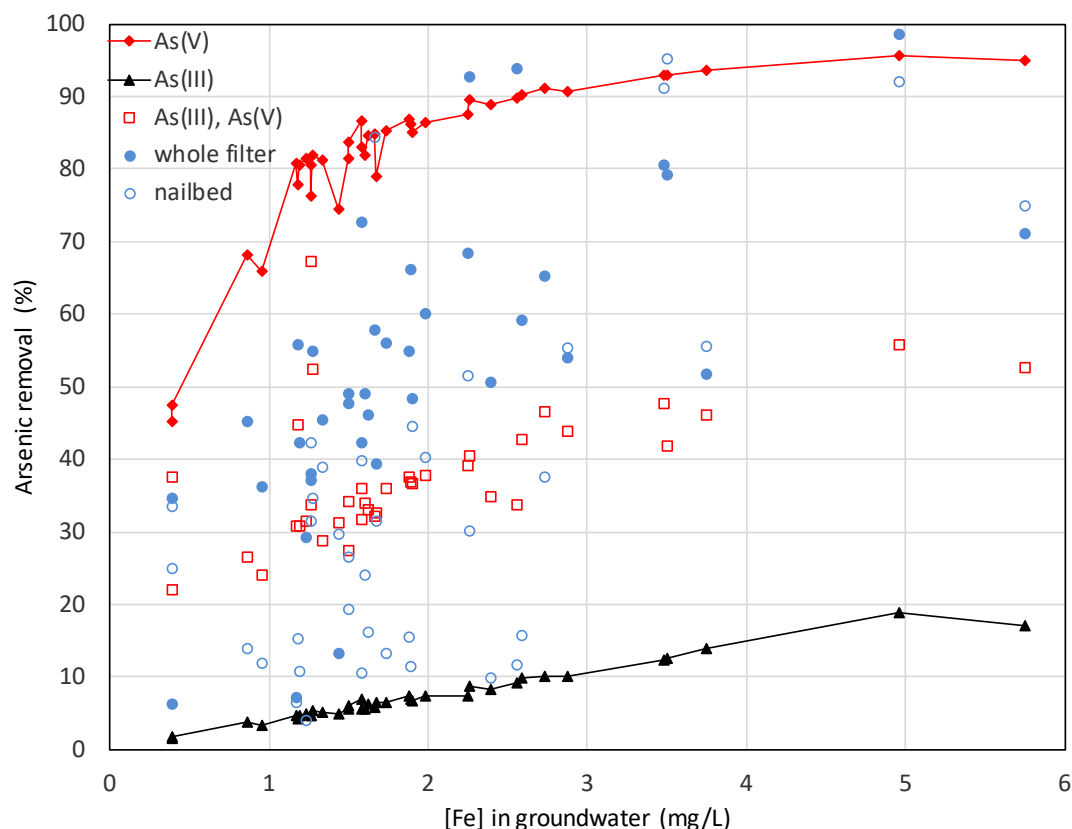
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806 *Fig. 5*. Observed and calculated arsenic removal as a function of the naturally present Fe-
807 concentrations. The observed As-removal by the nailbed alone is shown by empty blue
808 circles and the removal by the whole filter by filled blue circles. The red diamonds and line
809 show the calculated As-removal if all As were oxidized to As(V), the red squares for
810 calculated As(III) and As(V) fractions formed by co-oxidation of As(III) with the present
811 Fe(II), and the black triangles for As present as As(III). For the calculations, the
812 co-precipitation model described by Roberts et al. 2004 was used, as explained in the text.
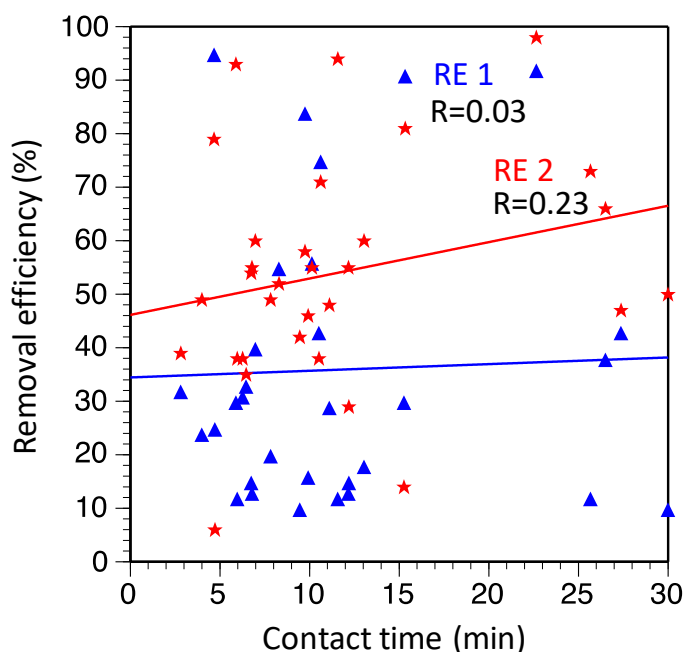
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Fig. 6a. Arsenic removal efficiency (RE in %) as a function of contact time in ground water

partially filtered by the nails (RE 1) and after complete filtration process (RE 2). Data from

32 filters where removal efficiencies could be calculated. Due to sampling or analytical

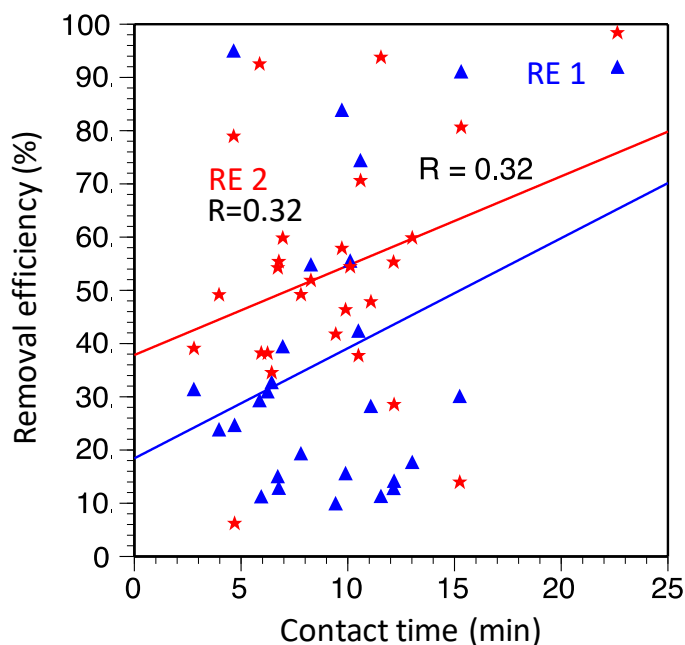errors some filters are not included in the table. No obvious correlation was detected.

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820

Fig. 6b. Arsenic removal efficiency (RE in %) as a contact time (min.) plotted against

removal efficiencies of arsenic in ground water partially filtered by the nails (RE 1) and after

complete filtration process (RE 2). Data from filters SN12, SN57, SN72 and SN79 (with

highest contact time) were omitted. The correlation is more apparent with the omitted filters.

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Electronic Supplementary Material

Kanchan arsenic filters in the lowlands of Nepal – mode of operation, arsenic removal and future improvements

Barbara Mueller [a,e], Bipin Dangol [b], Tommy K. K. Ngai [c], Stephan J. Hug [d]

[a] *Bamugeobiochem, Horbenstrasse 4, 8356 Ettenhausen, Switzerland*

[b] *ENPHO, Environment and Public Health Organization, New Baneshwor, Kathmandu,*

Nepal

[c] *CAWST, Center for Affordable Water and Sanitation Technology, B12, 6020 – 2Street SE,*

Calgary, Alberta, T2H 2L8, Canada

[d] *Eawag, Swiss Federal Institute of Aquatic Science and Technology, Ueberlandstrasse 133,*

CH-8600 Duebendorf, Switzerland

[e] Corresponding author. Email: bar.mueller@unibas.ch
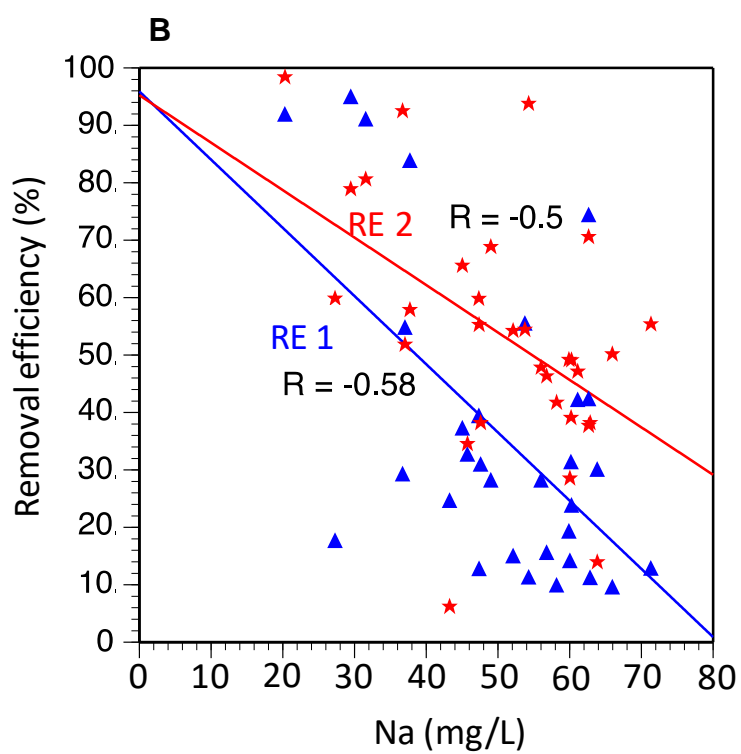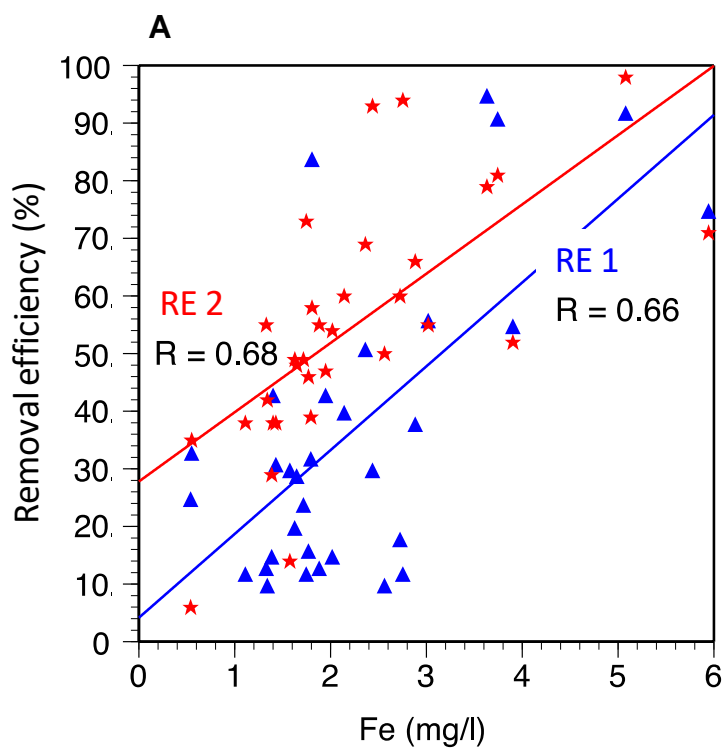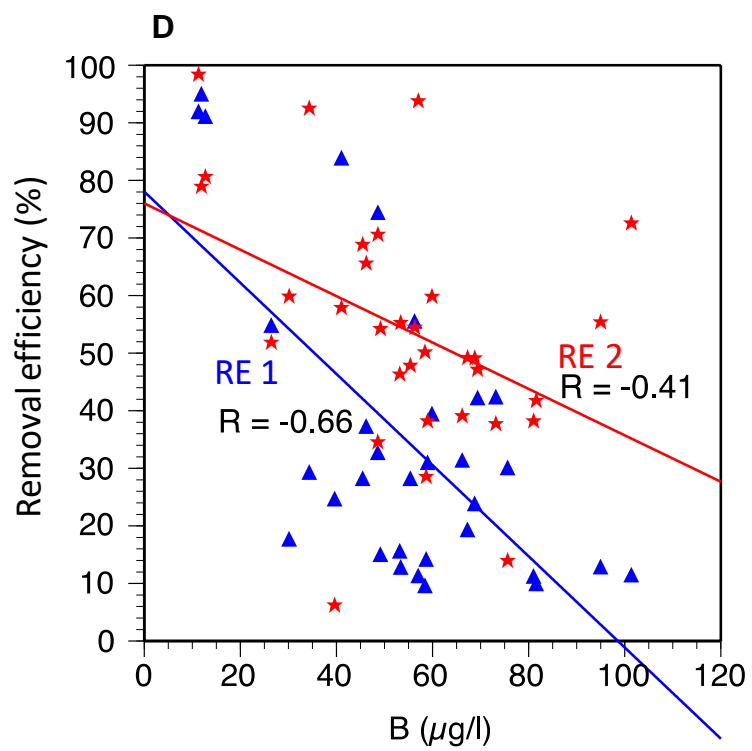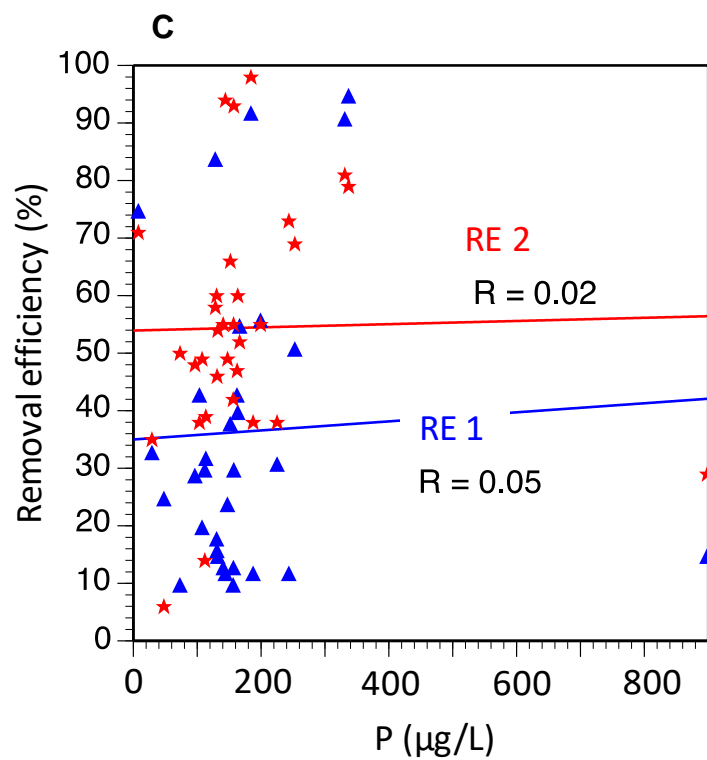
Number of pages: 10

Content

Table S1. Locality, concentration of As in raw water (µg/l) and % Arsenic removal efficiencies in 38 filters, after passing the iron nails (nailbed) and after the complete filtration process (effluent). SN = Sample number, according to the register provided by ENPHO.

Filter number	Locality	Raw water (µg/l)	After nailbed (% removal)	Filter effluent (% removal)
SN00	Manari	256	11.4	66.0
SN000	Manari	145	38.9	45.5
SN8	Manari	203	51.4	68.5
SN9	Manari	124	92.0	98.5
SN10	Manari	169	95.2	79.2
SN11	Manari	341	24.0	49.1
SN12	Manari	300	44.6	48.4
SN13	Manari	758	31.4	39.3
SN14	Manari	623	29.6	13.2
SN15	Manari	341	25.0	6.3
SN17	Manari	349	19.4	49.0
SN18	Manari	510	42.3	37.1
SN21	Manari	90	91.2	80.5
SN23	Manari	164	84.4	57.8
SN26	Panchanagar	277	55.6	51.7
SN33	Panchanagar	376	15.6	59.1
SN34	Panchanagar	393	33.4	34.7
SN35	Panchanagar	334	40.3	60.1
SN37	Panchanagar	171	34.7	55.0
SN45	Ramgram	170	74.9	71.1
SN51	Ramgram	195	3.9	29.2
SN53	Ramgram	253	26.6	47.6
SN54	Ramgram	281	16.2	46.1
SN55	Ramgram	279	15.6	54.8
SN57	Ramgram	179	37.6	65.2
SN62	Ramgram	219	13.2	55.9
SN63	Ramgram	159	30.0	92.7
SN64	Ramgram	100	39.9	42.2
SN65	Ramgram	623	11.7	93.8
SN66	Ramgram	268	55.4	54.1
SN67	Ramgram	166	31.4	37.9
SN68	Ramgram	189	15.2	55.9
SN69	Ramgram	273	13.8	45.3
SN70	Ramgram	498	11.9	36.2
SN72	Ramgram	202	9.9	50.6
SN75	Sukauli	136	10.7	42.3
SN76	Sukauli	139	6.6	7.2
SN79	Tilakpur	212	10.5	72.6

	Li	B	Na	Mg	Si	P	K	Ca	Mn	Fe	Zn	As	Sr	Mo
Li	1.00													
B	0.16	1.00												
Na	0.36	0.70	1.00											
Mg	0.13	0.59	0.56	1.00										
Si	0.82	-0.02	0.10	0.04	1.00									
P	-0.52	-0.17	0.06	-0.04	-0.50	1.00								
K	0.36	-0.07	0.22	0.13	0.35	-0.13	1.00							
Ca	-0.28	-0.53	-0.55	-0.49	-0.17	0.27	-0.09	1.00						
Mn	-0.77	-0.44	-0.42	-0.34	-0.70	0.38	-0.20	0.48	1.00					
Fe	-0.55	-0.50	-0.41	-0.30	-0.43	0.76	-0.10	0.63	0.53	1.00				
Zn	-0.16	-0.28	-0.11	-0.31	0.02	0.05	-0.06	0.07	0.31	0.01	1.00			
As	0.49	0.16	0.42	0.07	0.29	-0.25	0.23	-0.36	-0.43	-0.35	0.00	1.00		
Sr	0.33	0.82	0.59	0.70	0.21	-0.39	0.03	-0.44	-0.50	-0.60	-0.17	0.12	1.00	
Mo	0.57	0.35	0.64	0.47	0.42	-0.38	0.38	-0.76	-0.58	-0.61	-0.13	0.71	0.41	1.00

Fig. S1. Correlations between element concentrations in the untreated raw groundwater. The negative correlations between Fe and B, Na, and Mo can explain the corresponding negative correlations between As-removal and Na, B and Mo in Figures S2 B and S2 D-E.
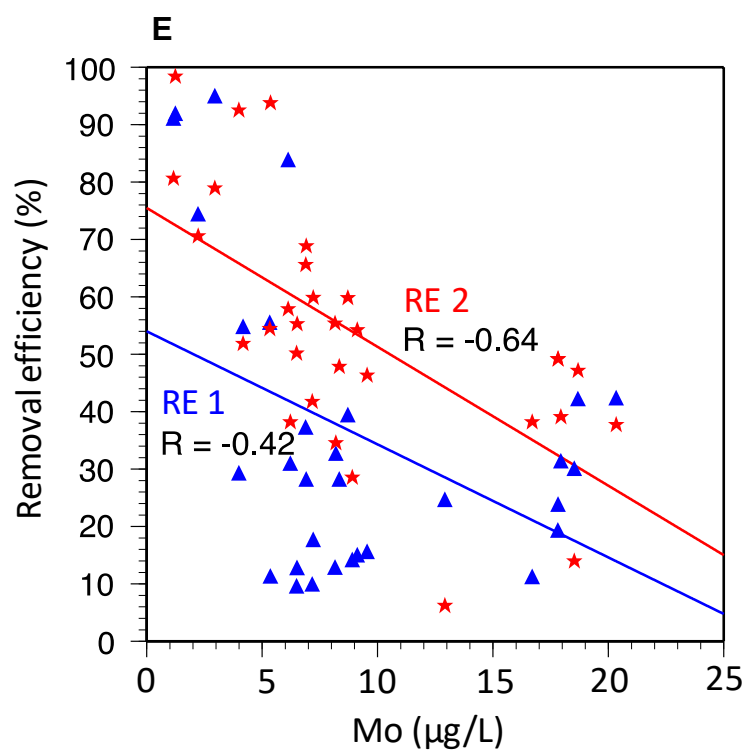
A

Removal efficiency (%) vs Fe (mg/l)

RE 2
R = 0.68

RE 1
R = 0.66

B

Removal efficiency (%) vs Na (mg/L)

RE 2
R = -0.5

RE 1
R = -0.58

Fig. S2. % Removal efficiencies of arsenic in ground water partially filtered by the nails

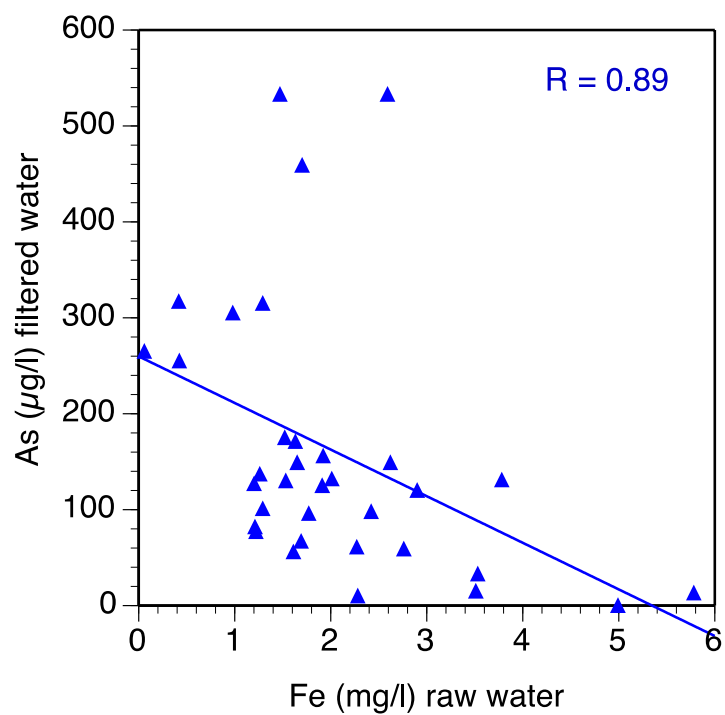(rate 1) and after complete filtration process (rate 2) in dependence of Fe, Na, P, B and Mo.

Fig. S3. Correlation of arsenic after complete filtration with the concentration of Fe in the raw water

Fig. S4. Nailbed immersed in water from the best performing filter SN9. This filter is of the rare type "metal round".

Fig. S5. Nailbed of filter SN15 (removal rate 6.3 %). The ground water can freely pass through the nail bed without contact to the nails.

Fig. S6. Nailbed of filter SN11 (removal rate 49.2 %). Although this nail bed looks ok (rusty, partly black nails) the ground water can freely pass at the rim of the filter. The "glued" nails impede the common functioning of the filter.

Text S1. Flushing of tubewells

The diameter of tubewells for household drinking water wells in Nepal is generally 40 mm (1.5 inch). With a common tube length of 25 m, the volume of water in the tube is 31.4 L. Hand pumps typically pump around 30 L water per minute (Ferguson *et al.* 2011). Pumping for 2 min thus flushes the entire tube and pump with about twice the volume of water in the tube.

Ferguson et al. (2011). Hand-pumps as reservoirs for microbial contamination of well water. Journal of Water Health 9(4), 708–717. doi:10.2166/wh.2011.106.

https://sswm.info/sites/default/files/reference_attachments/OLLEY%202008%20Human%20Powered%20Handpumps%20for%20Water%20Lifting.pdf
(Piston 24-36 litres/min at 7 m depth Shallow Well Household and Small Community Industrial Village Level with local skills and purchase of spare parts). Shallow well piston pumps lift 24-36 litres of water per minute.