

**Impact of water components on iron nail corrosion and arsenic removal efficiency of  
Kanchan™ arsenic filter (KAF)**

Minh Phuong Nguyen

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By: Minh Phuong Nguyen

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And submitted in partial fulfillment of the requirements for the degree of

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Complies with the regulation of this University and meets the accepted standards with respect to originality and quality.

Signed by the final examining committee:

\_\_\_\_\_ Chair and Examiner

Dr. S. Rahaman

\_\_\_\_\_ External to the program

Dr. A. De Visscher

\_\_\_\_\_ Examiner

Dr. C. An

\_\_\_\_\_ Supervisor

Dr. Catherine N. Mulligan

Approved by \_\_\_\_\_

Chair of Department or Graduate Program Director

\_\_\_\_\_

Dean of Faculty

Date May 21<sup>st</sup>, 2019

Abstract

**Impact of water components on iron nail corrosion and arsenic removal efficiency of Kanchan™ arsenic filter (KAF)**

Minh Phuong Nguyen

The Kanchan Arsenic Filter (KAF) is an intermittent, point-of-use, affordable drinking water technology for the removal arsenic from groundwater. Performance of KAFs in Nepal has been variable, with high removal in some settings and low removal in others. This study consisting of two parts (laboratory and field research) was conducted to improve the performance of the KAFs. In the laboratory research, four different synthetic groundwaters spiked with arsenic were fed to the filters once a day for 35 days to evaluate the effect of water components on corrosion of iron nails and of arsenic removal in downscaled KAFs. Composition of water samples collected from those filters was analyzed using UV-Vis spectroscopy, Flame - atomic absorption spectrophotometry (F-AAS), and inductively coupled plasma mass spectrometry (ICP-MS). Morphology of iron nail surface was done by scanning electron microscopy – energy dispersive spectroscopy (SEM-EDS), followed by a characterization of iron rust powder using X-ray powder diffraction (XRD). Colloids of iron corrosion products were analyzed using transmission electron microscopy (TEM) for their phase, images and structure. Batch tests were conducted to examine iron corrosion rates of iron nails in the presence of 2,2-bipyridine under different testing conditions. The field research in Nepal containing water quality assessments, filter checks, and household interviews was carried out to explore potential factors causing a bias and inadequate filter performance.

Results from the lab research showed remarkable impacts of water conditions on iron corrosion and arsenic removal by the KAFs, with the strong enhancing effect of hardness surpassing the inhibiting effect of phosphate. Filters running hardness-rich water (200 mg/L Ca) removed arsenic

(initial concentration of 2000  $\mu\text{g/L}$ ) to under the limit of Nepali water standard (50  $\mu\text{g/L}$ ) while filters running low hardness water (40 mg/L Ca) removed much less arsenic, especially in the presence of phosphate. On average, 35.7 to 47.9% of arsenic was removed after contacting with iron nails while total arsenic removal was 85.2 to 99.5% over the duration of this study.

The field research revealed that inconsistencies in performance of the full-scale KAFs were mostly resulting from variations of groundwater quality, improper methods of maintenance and operation of the filter users and unfavorable water conditions. A change in nail type showed a moderate difference on filter performance. Addition of embedded nails in filter media significantly facilitated arsenic removal and is a promising solution for enhancing the KAF performance.

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List of Abbreviations

BPY	2,2-bipyridine
BSF	Biosand filter
CAWST	Center for Affordable Water and Sanitation Technology
DI	Deionized water
DO	Dissolved oxygen
EDX	Energy dispersive X-ray spectroscopy
ENPHO	Environment and Public Health Organization
EW	Effluent water
F-AAS	Flame -atomic absorption spectrophotometry
GW	Groundwater
HFO	Hydrous ferric oxide
ICP-MS	Inductively coupled plasma mass spectrometry

KAF	Kanchan Arsenic Filter
LSI	Langelier Saturation Index
MDI	Morrill Dispersion Index
ORP	Oxidation-reduction potential
RW	Raw water
SAED	Selected area electron diffraction
SEM-EDS	Scanning electron microscopy – Energy Dispersive Spectroscopy
SW	Supernatant water
TDS	Total dissolved solids
TEM	Transmission electron microscopy
UV-Vis	Ultraviolet-visible spectroscopy
WHO	World Health Organization
XRD	X-ray powder diffraction
ZVI	Zero-valent iron

## CHAPTER 1 Introduction

### 1.1 Research context

Arsenic contamination is a current global issue of concern due to its extensive distribution, high toxicity and the complexity of the arsenic removal process. Arsenic exposure is well-known as a dangerous threat to the health of hundreds of million people in the world (Tantry et al., 2015).

Among numerous water treatment techniques, the Kanchan Arsenic Filter is suitable for developing countries facing elevated arsenic levels (Chiew et al., 2009) because of its simple design with 1) locally available constructing materials, 2) minimal requirements in operation training, 3) no energy or chemicals needed in the operating process, 4) sustainability, and 5) easy and periodic maintenance with no or very little replacement needed (Zaman et al., 2014). The Kanchan Arsenic Filter (KAF) is an affordable household water treatment device removing turbidity, pathogens, contaminants, and especially arsenic from drinking water based on slow sand filtration and iron corrosion adsorption mechanisms (Thakur et al., 2010). In a two-year research study, over 1000 KAFs implemented in rural area of Nepal showed a 85-90% arsenic removal and excellent removal of iron, turbidity and total coliforms with a very high acceptance of users after one year of use (83%) (Mueller, 2016).

However, data analysis of the water quality assessment provided by CAWST taken in Nawalparasi district in three years showed that the performance of KAFs was not uniform even for filters using the same source of input unfiltered water. Filters were installed and operated the same day and used by closely located households had very different removal efficiency. Even within a KAF, the removal efficiency was unstable and inadequate in different testing time points of the study. Furthermore, some filters had negative contaminant removal efficiency. In other words, the quality of filtered water was poorer than that of the input or supernatant water in the same filter. The

arsenic removal efficiency of the KAFs was not related to arsenic concentration in the raw water as well. In general, the performance of KAFs in Nawalparasi, Nepal was not as reliable as expected after a period of use. That might be one of the reasons contributing to the permanent or occasional abandonment of the KAF by a few households in the community.

The KAF performance, as widely shown in previous studies, varies largely and depends highly on environmental conditions (Chiew et al., 2009). This study focused on iron nail corrosion which has been expected to play a crucial role on the arsenic removal process taking place in the KAF (CAWST 2012).

The study consists of two parts: lab research conducted in Environmental laboratories at Concordia University, Montreal, Canada and field research in the Nawalparasi district, Nepal collaborated with researchers from the Center for Affordable Water and Sanitation Technology (CAWST), Calgary, Canada and staff from the Environment and Public Health Organization (ENPHO), Nepal.

## 1.2 Objectives of the study

### 1.2.1 Objectives of the laboratory research at Concordia University, Montreal, Canada

In this lab research, corrosiveness of iron nails and the role iron corrosion on arsenic removal of the KAF were critical factors of concern. For a throughout investigation on this, the objectives of the study were as follows:

- To investigate the corrosion behavior of the nails by testing the corrosion rate using batch experiments and comparing to those in previous studies.
- To design and build lab-scale BSFs that are representative of the vertical cross-section of the full-scale KAF to test their performance.

- To evaluate how different components of synthetic groundwater affect the iron corrosion process and arsenic removal efficiency of down-scaled filters.

### 1.2.2 Purpose of the field research in the Nawalparasi district, Nepal

The field research in Nepal took place over a two-week period from late September to early October of 2018 with two main parts: user interviews and a water quality assessment. The research was designed to have a comprehensive understanding of the real-world context of operation and maintenance of the KAFs in the Nawalparasi district for some long-term objectives as follows:

- To identify the sources of problems by studying user habits of employing the KAFs, evaluating filter parameters, and assessing water quality. Discovering the main reasons for KAF abandonment in general.
- To improve the performance of KAFs by examining the impact of changing iron nail types and adding embedded nails in filter media.
- To enhance user satisfaction with the KAF.

## 1.3 Scope and limitations of the thesis

### 1.3.1 Scope of the thesis

The first part of the thesis was a lab research composed of batch tests and column experiments for a set of downscaled filters which were designed and constructed with respect to the fundamental operational parameters of the real KAF. The results and scientific conclusions obtained from the lab research together with a data analysis from a Nawalparasi water quality assessment in the database at CAWST were applied to plan a field study in the Nawalparasi district, Nepal. The field research was done from late September to the beginning of October 2018 conducted of three main parts: user interviews, filter inspections and water quality tests in order to investigate all possible

factors preventing KAFs from its sustainably high performance and to increase filter efficiency as well as satisfaction of filter users.

### 1.3.2 Limitations of the lab research

In order to investigate the impact of water on iron nail corrosion, experiments on lab-scale filters were stopped when the filters began to clog, the iron nail surface and iron rust cumulated in the standing water were sampled and analyzed. No filter cleaning was done for either the sand surface or iron nails over the period of water treatment. Thus, the long-term use of the KAF, especially the performance of the KAF after maintenance was not examined in this study.

Among various factors of groundwater influencing iron corrosiveness and arsenic removal of the KAF, hardness and phosphate were chosen to be the independent parameters to be investigated in this study. Other factors, such as water components (alkalinity, silicate, and chloride) and water conditions (pH and temperature) were dependent parameters which could not be analyzed for their individual effect on the performance of the lab-scale filters.

### 1.3.3 Limitations of the field research in Nawalparasi district, Nepal

The research group fed the filter and collected a sample once a day in the morning to minimize the inconvenience caused to the households while all households employed their KAFs multiple times a day. As a result, filtered water was not corresponding to the raw water and supernatant water of the same day because they were of different water doses. The filtered water belonged to the latest dose fed on the previous day that stayed in the filter media and was flushed out by the newly fed raw water. Hence, the arsenic removal efficiency of each dose of water could not be calculated. Only comparisons between the quality of input and output water on the same day of the KAFs were made.

#### 1.4 Organization of the thesis

Chapter 2 of the thesis is a literature review of the context of arsenic contamination in the world, arsenic chemistry, an overview of current arsenic removal techniques, an introduction to the Kanchan arsenic filter (KAF), the mechanism of arsenic removal by the KAF and factors influencing the arsenic removal.

Chapter 3 describes the materials and methods used in the whole study. In part A of this chapter, the design of lab-scaled filters, operational tests and research methodology are illustrated. Methods of user interviews, filter check and water quality assessment of the field research in Nawalparasi district are reported in part B of the chapter.

Chapter 4 presents the results and discussions of the work done in two parts of this study. The contribution of the laboratory work to a field study in Nepal is also mentioned at the end of this chapter.

Chapter 5 summarizes the findings and observations of the study, as well as suggestions for future research.

The last part is the references of previous studies and knowledge mentioned in this study.



2.1 Arsenic-related problems in the world

Arsenic is found at elevated levels in the groundwater of numerous countries worldwide such as Argentina, Bangladesh, India, China, Chile, Mexico, the United States of America, West Bengal, Nepal, Mongolia, Cambodia, Myanmar, Iran, Vietnam, Afghanistan Pakistan (Mukherjee et al., 2006). Arsenic can leach to groundwater and surface water from natural geological sources (such as the dissolution of arsenic in rocks, sediments, minerals, and ores), industrial processes (such as mine-digging and paints), and agricultural activities (such as pesticides, fertilizers, and herbicides) (Mukherjee et al., 2006).

The severity of arsenic poisoning largely depends on the types of arsenic, dose, and duration of arsenic exposure (WHO, 2011). In 2003, Ratnaike affirmed that “As(III) is 60 times more toxic than As(V). Organic arsenic is non-toxic whereas inorganic arsenic is toxic” (p.1, paragraph 10). Arsenic poisoning caused by drinking arsenic-contaminated water can result in immediate, short-term (acute) and long-term (chronic) health effects, beginning with headaches, confusion, drowsiness, and diarrhea. Arsenic poisoning reduces infant weight while increases infant mortality, miscarriage, and childhood cancer (Tantry et al., 2015). Acute arsenic poisoning symptoms may include diarrhea, vomiting, vomiting blood, dark urine or blood in the urine, cramping muscles, convulsions and abdominal pain (Ratnaike, 2003). In the worse case scenario, it can result in coma and death. Chronic arsenic exposure can lead to severe health problems such as cancers (skin, lung, and bladder cancers are the most commonly found), diabetes, heart diseases, and neurological diseases (Vaishya & Gupta, 2005). Therefore, to save people from arsenic poisoning, national limitations of arsenic levels in water were established over the world.

Currently accepted national standards for arsenic in drinking water vary due to each country between 10 to 50  $\mu\text{g/L}$ . The WHO provisional guideline value for arsenic is 10  $\mu\text{g/L}$  because the minimum detection level of modern analytical technologies for arsenic is 10  $\mu\text{g/L}$  and it is challenging to remove arsenic to a concentration below that level (WHO, 2011). The WHO also mentioned some studies indicating the low risk of being exposed to arsenic at a concentration of 10  $\mu\text{g/L}$  and the health impact of arsenic exposure on the people living in areas with arsenic concentration levels around 50  $\mu\text{g/L}$  is still ambiguous. Table 1 shows guideline values for arsenic implemented in different countries and organizations.

Table 1. Common arsenic maximum acceptable levels worldwide

Country/Organization	Revised arsenic maximum acceptable level $\mu\text{g/L}$ )	References
WHO	10	(WHO, 2011)
US	10	(USEPA, 2018)
Canada	10	(CDW, 2017)
European Community	10	(European Commission Directive, 1998)
Australia	10	(NHMRC, 2011)
Bangladesh, India, Nepal, China, Vietnam, etc.	50	(Mondal et al., 2008)

## 2.2 Arsenic chemistry

The presence of arsenic in the natural environment varies considerably based on its forms and oxidation states. Arsenic exists in two forms (organic and inorganic) and four oxidation states:  $\text{As}^-$

$As^3$  (arsine),  $As^0$  (metalloid arsenic),  $As^{+3}$  (arsenite, As(III)), and  $As^{+5}$  (arsenate, As(V)) (Nguyen et al., 2009; Nicomel et al., 2015; Thakur et al., 2010). Nicomel et al. (2015) stated that the most common compounds of inorganic arsenic are arsenite As(III) and arsenate As(V). As(III) is known to be more toxic, more mobile, more soluble and harder to be removed from water than As(V) (Anjum et al., 2011).

Arsenic oxidation species are highly dependent on pH and oxidation-reduction conditions as shown in Figures 1 and 2 (Chowhury, 2015).

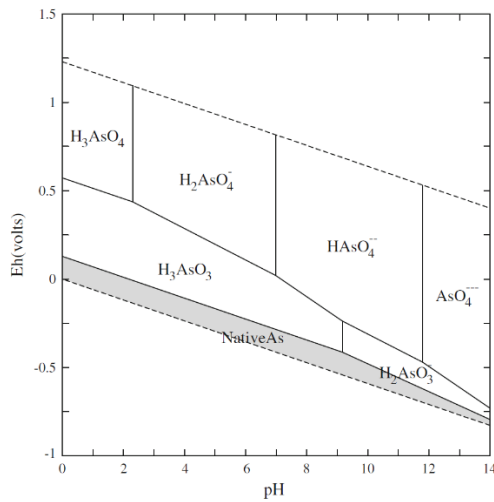
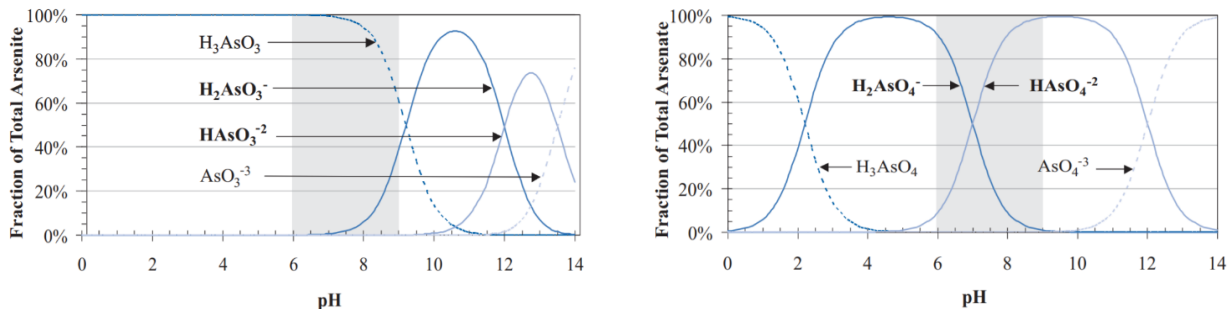


Figure 1. Eh-pH diagram for the As-O-H system at 25<sup>0</sup>C and 1 bar (Retrieved from Lu & Zhu, 2011). The grey area illustrates the solid phase of arsenic.

In anoxic and/or reducing conditions arsenic exists mostly in As (III) form as  $H_3AsO_3^0$ ,  $H_2AsO_3^-$ ,  $HAsO_3^{2-}$ , and  $AsO_3^{3-}$  while in aerobic and/or oxidizing condition, As(V) form is favored, distributing in arsenate derivatives as  $H_3AsO_4^0$ ,  $H_2AsO_4^-$ ,  $HAsO_4^{2-}$ , and  $AsO_4^{3-}$  (Chowdhury & Yanful, 2010; Müller et al., 2010; Thakur et al., 2010).



a) As(III) species

b) As(V) species

Figure 2. pH dependence of arsenic species (USEPA, 2003). The grey area demonstrates the most common pH range

Sadiq (1997) showed that at neutral pH 7 regardless of redox conditions, the distribution in descending order of As(V) and As(III) species in soil was  $\text{HAsO}_4^{2-} > \text{H}_2\text{AsO}_4^- > \text{AsO}_4^{3-} > \text{H}_3\text{AsO}_4^0$  and  $\text{HAsO}_2^0 = \text{H}_3\text{AsO}_3^0 > \text{AsO}_2^- = \text{H}_2\text{AsO}_3^- > \text{HAsO}_3^{2-} > \text{AsO}_3^{3-}$ , respectively. Concentrations of As(III) species escalated with respect to a reduction in the redox potential (pE). The dependence on pH and pE of arsenic species in soil goes along with those in water as demonstrated by USEPA (2003). Sadiq (1997) also described that As(III) was the dominant species in anoxic soil with  $\text{pE} + \text{pH} < 6$ , while As(V) was the major species in oxic soil where  $\text{pE} + \text{pH} > 10$ , and in suboxic soil with a  $\text{pE} + \text{pH}$  range of 6 to 8 both As(III) and As(V) species are present and distributed in accordance with the soil conditions.

### 2.3 Arsenic removal techniques

To lower arsenic levels in water under national allowable values or WHO guideline limit, a good deal of established methods has been investigated and conducted to treat different types of water and fit various conditions of usage. A research carried by Nicomel et al. (2015) listed current arsenic removal technologies in 5 categories: coagulation-flocculation, ion exchange, adsorption (zero valent iron, activated alumina, iron-based sorbents, etc.), membrane filtration (low pressure:

microfiltration and ultrafiltration, high pressure: reverse osmosis and nanofiltration), and oxidation (oxidation and filtration, photochemical, photocatalytic, biological, and in situ oxidation).

Liu et al. (2015) claimed that “among many techniques currently available for As removal from water, the adsorption process is considered one of the most promising techniques because it is economical, effective and socially acceptable” (p.7726). This conclusion agrees with Nicomel et al. (2015) and Philippi (2016).

## 2.4 Household arsenic removal technology in this study: the Kanchan™ arsenic filter (KAF)

### 2.4.1 Design of the Kanchan™ Arsenic Filter (KAF)

The Kanchan arsenic filter (KAF) is an affordable household water treatment device for rural communities lacking access to clean water supply from water treatment plants. The KAF is a low footprint and no energy consumption filter derived from an intermittent slow sand filter (biosand filter) by integration with a diffuser basin containing rusty iron nails for arsenic removal. Ngai et al. (2007) described KAFs as an advanced biosand filter for a better performance in terms of arsenic and pathogen removal. According to previous studies and technical documents, the pathogen removal is done by a combination of bio-physical processes happening on a thin bio layer on the surface of the sand (so called biofilm or Schmutzdecke layer) such as mechanical straining, attachment, predation and natural death while the arsenic removal is a result of interactions with iron corrosion products (adsorption, co-precipitation) followed by physical straining by the filter media (CAWST, 2012a; Espinoza, 2011; Mueller, 2016).

During the KAF development process, several versions of KAFs were released in different shapes (round and square) and filter body materials (concrete, metal or plastic) with some small changes in technical design. The cross-sectional design of the KAF version 10.0 is illustrated in Figure 3 from information obtained in the Biosand Filter Construction Manual released by CAWST (2012)

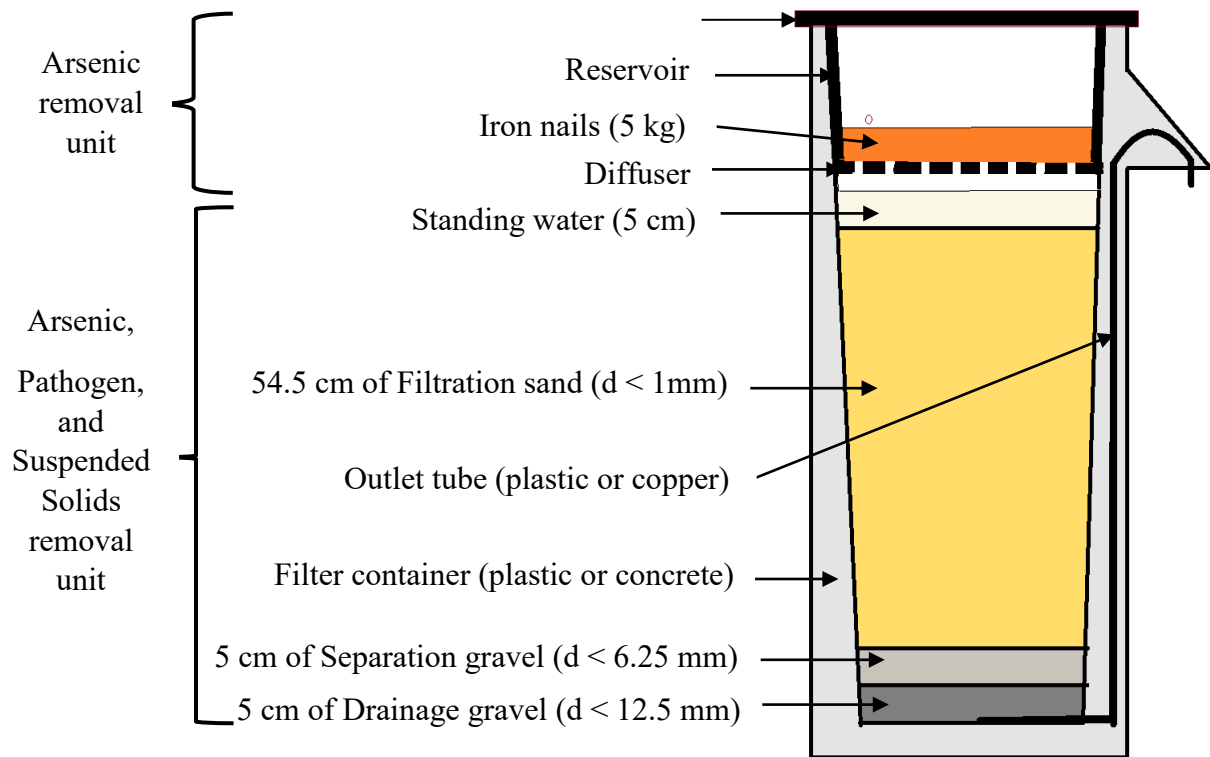


Figure 3. Diagram of KAF version 10 cross-section (CAWST, 2012a)

The design of the KAF and instructions for construction, installation, operation, and maintenance of the KAF are well described and available for public use on the website of Center for Affordable Water and Sanitation Technology (<https://www.cawst.org/>).

The KAF can be built by locally trained technicians using simple construction tools. Materials of the KAF production such as bricks, iron nails, sand, gravels, pipe, and plastic containers or concrete filter body are locally available, sustainable and affordable. The KAF operation and maintenance are easily done by filter users after training by local technicians. The only cost for maintenance is the purchase of new iron nails after at least three years of use (Ngai et al., 2007). Hence, the KAF was expected to be economical, sustainable, environmentally friendly and practical for vulnerable populations deeply affected by water contamination and severe arsenic exposure problems (Chiew et al., 2009; Elliott et al., 2015; Ngai et al., 2007).

#### 2.4.2 Arsenic removal mechanism of the KAF

A significant difference between the KAF with the conventional biosand filter is the diffuser basin containing rusty iron nails which play an essential role for arsenic removal. The KAF can remove arsenic from contaminated water by the physical-chemical interaction between iron and arsenic (Charlet et al., 2011; Mak, Rao, & Lo, 2009a; Sadiq, 1997) as well as physical removal of arsenic-contained compounds by straining on the sand surface and filter media of the filter (CAWST, 2012a; Ngai et al., 2007).

After contact with water, iron nails rust, forming various iron corrosion products which are ferric (Fe(III)) and ferrous (Fe(II)) hydroxides and oxides (Farrell & Chaudhary, 2013; Liu et al., 2015). Arsenic adsorption is significantly dependent on iron corrosion products' surface characteristics rather than their compounds' structures (Onoda Jr & De Bruyn, 1966; Yean et al., 2005). Iron corrosion products and their surface can play numerous significant roles in arsenic removal processes such as a reducing or oxidizing substance, co-precipitant, contaminant-immobilizing agent, or a sorbent (Nicomel et al., 2015). Arsenic may adsorb onto active sorption sites on the iron surface, or form precipitates with iron (hydro)oxides which then get trapped in the pores of fine sand of the KAF (Ngai et al., 2007).

In terms of arsenic removal, several studies indicated an inadequate performance of KAFs (Chiew et al., 2009; Wenk, 2008). Chiew et al. (2009) employed three KAFs in five and a half months to filter water from three arsenic-contaminated tube wells in Cambodia with different arsenic concentrations. None of the three filters could reduce arsenic concentrations of treated water under 50 µg/L. The arsenic removal efficiency of those filters was 39.4% (for the influent water with the highest As concentration), 74.9% (moderate As concentration) and 45.4% (lowest As concentration), but there was no correlation between arsenic concentrations in influent and effluent

water. Wenk (2008) conducted batch and column experiments for downscaled filters to investigate the influence of different iron corrosion products on the arsenic removal process and filter performance regarding different water components. After one month running the column experiments, the results showed that the arsenic removal efficiency of those filters was just about 50%.

Nevertheless, both studies had serious weaknesses affecting the value of their conclusions. The drawback of the Chiew et al. (2009) study was the lack of repetition. The Wenk (2008) study had two main limitations. First of all, the column experiments had no repetition. Second, the small filter columns were not representative of the KAFs as they were only the downscaled top part of KAF with the flow rate kept constant over time by a peristaltic pump. The bottom part of the KAF in which sophisticated biological, physical and chemical removal processes happened was eliminated in the Wenk (2008) study.

Therefore, more comprehensive studies on the arsenic removal by iron corrosion products should be done to verify effectiveness and/or to improve the KAF's performance.

#### 2.4.3 Iron corrosion products and arsenic removal

The mechanism of arsenic adsorption onto the surface of iron-corrosion products (i.e., iron(oxy)hydroxides, iron oxides, etc.) remains ambiguous and controversial. Farrell et al. (2013) and Yang et al. (2015) demonstrated kinetics of arsenic adsorption on iron hydroxides where some arsenic adsorption processes occur in seconds while full equilibrium may take weeks to be reached.

When iron nails contact water, the oxidation of Fe(0) surface is a corrosion process forming Fe(II) and Fe(III) as the following equations:





Or in the presence of O<sub>2</sub>,



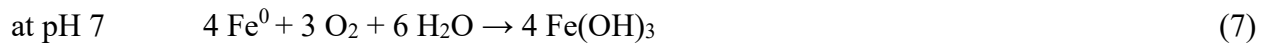
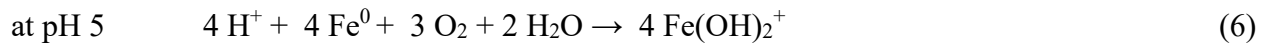
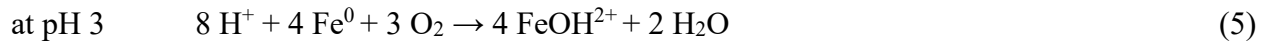
Then,



Fe(III) can be formed by the further oxidation of Fe(II) as the following equation:



or from Fe(0) at different pH conditions (Katsoyiannis et al., 2008) as follows:



Fe (II) / Fe (III) mixed products can be formed from hours to days (Manning et al., 2002). Iron corrosion products include iron oxides, iron hydroxides, etc. Yean et al. (2005) found that not the structures of different iron oxide, but the properties of iron oxides' surface are critical factors determining arsenic adsorption on iron oxides. Katharina et al. (2010) reported that goethite and hematite have much lower surface area compared to ferrihydrite and ferroxhyte, therefore are less available for As adsorption. Magnetite is capable of arsenic adsorption, especially for As(III) (Wang et al. 2014; Chowdhury et al., 2010).

pH also plays a significant role on arsenic adsorption on iron corrosion surface as it determines the charge of the iron's surface. Depending on the pH, magnetite can generate Fe(II) and its hydrolysis

products such as  $\text{Fe}(\text{OH})^+$ ,  $\text{Fe}(\text{OH})_2$ , and  $\text{Fe}(\text{OH})_3^-$  (Manning et al., 2002). At  $\text{pH} < 5.6$ , iron surface will be positively charged groups  $\text{Fe}^{2+}$  or  $\text{Fe}(\text{OH})^+$ , which attract negatively charged As compounds. At higher pH, iron corrosion may exist in the neutral or negatively charged forms repelling negatively charged arsenic, and as a sequence, lowering As adsorption on the iron surface (Chowdhury et al., 2010; Farrell et al., 2013).

#### 2.4.4 Factors affecting iron corrosion and arsenic removal efficiency of the KAF

The arsenic removal process is complex and highly dependent on environmental conditions. According to previous research on arsenic removal by zero valent iron (ZVI) or iron corrosion products, the main elements determining the arsenic removal performance of the KAF are: corrosiveness of iron nails, co-precipitation process between iron rust and arsenic, availability of sorption sites for arsenic, arsenic speciation (due to reduction-oxidation conditions), and arsenic desorption/mobility in water. Which factors favor or inhibit arsenic removal and to what extent those factors affect the removal process are still not well established.

In terms of water compositions that can influence the iron – arsenic interaction process, Mak et al. (2009b) and Tanboonchuy et al. (2012) described calcium hardness ( $\text{Ca}^{2+}$ ) and chloride ( $\text{Cl}^-$ ) as enhancing factors, the former provided additional adsorption sites for arsenic anion, and fostered iron corrosion process while the latter favor corrosive conditions. Mak et al. (2009b) also reported that calcium hardness in the presence of bicarbonate could form calcium carbonate precipitates which can enhance arsenic removal by sweep precipitation, but on the other hand, it may block iron active sites and reduce its reactivity.

Phosphate ( $\text{PO}_4^{3-}$ ), bicarbonate ( $\text{HCO}_3^-$ ), silicate ( $\text{SiO}_3^{2-}$ ), humic acid (HA) and sulphate were listed as major preventing factors as they competed with arsenic for adsorption sites on the surface of iron corrosion products (Nicomel et al., 2015; Tanboonchuy et al., 2012; Su & Puls, 2001;

Yean et al., 2005). Hsu et al. (2008) arranged the competitive strength of those ions were in a descending order of  $\text{PO}_4^{3-} > \text{SiO}_3^{2-} > \text{HA} > \text{HCO}_3^- > \text{Cl}^-$ . Moreover, HA also prevents the formation of iron corrosion product (Mak et al., 2009b).  $\text{HCO}_3^-$ , nevertheless, has a biased influence on arsenic removal efficiency because it can promote the iron corrosion process by stabilizing the solution pH but may also form carbonate minerals which can block the sorption sites and passivate the surface of iron (Mak et al., 2009b).

In addition to the presence of inhibiting chemical components in water, an insufficient amount of iron produced by iron nail corrosion also results in a poor arsenic removal performance of the KAF. Hug et al. (2008) stated that Fe/As and Fe/P ratios are the most important factors for arsenic removal. A Fe/As weigh ratio of  $>40$  or a Fe/As molar ratio of 53 is normally required to reduce As concentration to below 50 ppm even after a complete oxidation of As (III) (Meng et al., 2001). In the presence of P, at neutral pH, a Fe/P molar ratio of 1.5 to 2.0 is needed to remove phosphate, therefore, As removal only occurs when Fe amount in the solution is in excess of this ratio (Hug et al., 2008). Limited contact time of the contaminated water with iron nails and dissolved oxygen depletion were listed as two main elements contributing to the lack of iron corrosion products (Chiew et al., 2009; Wenk, 2008).

Arsenic dissolution/desorption is another factor strongly impeding its removal process. Arsenic adsorbed on the surface of hydrous ferric oxide (HFO) may be 1) reduced by a) natural organic matter (Yean et al., 2005), b) bacteria (Berg et al., 2001), and c) anoxic reducing conditions (Le Luu, 2017; Nguyen et al., 2009) or 2) replaced by bicarbonate and other competing compounds such as phosphate, silicate, sulphate (Mueller, 2016; Su & Puls, 2001). Arsenic mineral dissolution also contributes to the mobilization of arsenic in water (Dang et al., 2013).

## 2.5 Context of arsenic contamination in groundwater in Nawalparasi district, Nepal

The Nawalparasi district lies in the Terain province of Terai plain in the southern part of Nepal where groundwater is the major source of drinking water (Yadav et al., 2014). Arsenic contamination in groundwater of Nawalparasi has been very severe with the average concentration of 350  $\mu\text{g/L}$  and 98% groundwater exceeded the WHO guideline value of 10  $\mu\text{g/L}$ , higher As concentration at more than 400  $\mu\text{g/L}$  in wells with depths at 18-22 m and 50-80 m (Tran, 2017). Hydrogeochemical studies on the shallow tubewells of the Nawalparasi indicated that As concentration in groundwater was depth dependent (Yadav et al., 2014). Mueller (2016) claimed 11.69% of all tube wells in Nawalparasi had As concentration exceeding 50  $\mu\text{g/L}$  (Nepal's standard). Thakur et al. (2010) estimated that 85,344 people in Nawalparasi drank water containing As concentration higher than 50  $\mu\text{g/L}$  in 2011.

Previous groundwater quality assessments in Nawalpari reported that the groundwater was near-neutral to alkaline pH within a range of 6.1-8.1 in suboxic and moderately reduced conditions; multi-contaminated, Ca-Mg-Na-  $\text{HCO}_3^-$  water type with  $\text{HCO}_3^-$  as a dominant ion, high amounts of organic matter and variable levels of  $\text{PO}_4^{3-}$ ,  $\text{SO}_4^{2-}$ , and arsenic contaminated with As(III) as the dominant species (Diwakar et al., 2015; Mueller, 2016; Yadav et al., 2014). Pathak et al. (2013) indicated that groundwater sampled during wet season was more contaminated than that of the dry season. Nguyen et al. (2009) described the same phenomenon of contaminated groundwater in Ha Nam province, Vietnam. A possible explanation for this was the high rainfall intensity and groundwater recharge carrying high levels of contaminants and the reducing condition of groundwater favoring arsenic mobilization (Pathak et al, 2013; Yadav et al., 2014).

Yadav et al. (2011) and other researchers addressed arsenic desorption from natural rocks and sediment as a major source of high levels of arsenic in groundwater. Tran (2017) demonstrated a

strong correlation between arsenic and iron levels in sediment of groundwater, indicating that the reduction of arsenic-iron oxyhydroxides complexes could be responsible for the presence of arsenic in groundwater. Decomposition of organic matter was also mentioned as another source of arsenic leaching (Gurung et al., 2005, Tran, 2017). The use of cowdung during tubewell drilling was pinpointed by Bisht et al. (2003) as a possible source of organic matter promoting reductive processes and subsequent As leaching in groundwater (Diwakar et al., 2015). Oxidation of organic matter along with reductive dissolution of Fe and Mn-bearing minerals releasing As-oxyanions associated with these minerals (Diwakar et al., 2015, Gurung et al. 2005, Mueller, 2016).

After years of consuming highly arsenic-contaminated water and food, 3.6% population in Nawalparasi had chronic arsenic-related diseases so-called arsenicosis, of which the male population was prevalent, particularly those in their late 50s (Yadav et al., 2011). Some of the most typical symptoms are black and white spots on legs and trunk, skin lesions, gangrene of limbs (Thakur et al., 2010). Therefore, KAFs were supplied to households with family member(s) suffering from long-term arsenic poisoning in Nawalparasi. However, the efficiency and consistency of arsenic removal by these KAFs were inadequate and needed to be improved.

## CHAPTER 3 Materials and Methods

### A. Laboratory research at Concordia University, Montreal, Canada

#### 3.1 Material preparation

##### 3.1.1 Filter materials

Sand and gravels were locally purchased. Fisherbrand U.S. stainless steel sieves ½”, ¼”, No. 18, No. 40, No. 50, No. 80 and No. 100 with the pore sizes of 12.5 mm, 6.35 mm, 1 mm, 600 µm, 425 µm, 300 µm, 180 µm, and 150 µm were used to separate gravels and sand to different size ranges recommended by CAWST (2012b). The sieved materials were washed and oven-dried for 24 hours, followed by natural cooling.

A sand grain size test and a porosity test were conducted according to established manuals from CAWST for the filter media (clean sand) to measure its physical parameters. The filter media had a maximum diameter of 0.6 mm, an effective diameter ( $d_{10}$ ) of 0.18 mm, a uniformity coefficient (UC) of 2.22 and a porosity of 0.41, which meet the criteria of a  $d_{10}$  range of 0.15 to 0.20 mm, and a UC range of 1.5 to 2.5 (CAWST, 2012b).

##### 3.1.2 Iron nails

The iron nails used for lab-scale filters in this study were wire nails with heads produced by Hillman and purchased in Canada. The nails had a size of 1.5 x 26 mm. The surface area and average weight of one nail was approximately 158.7 mm<sup>2</sup> and 596.8 mg which results in an initial active surface area of 265.9 mm<sup>2</sup>/g. The nails in all filters of this study were cut in half to better fit the lab-scale filters.

### 3.1.3 Artificial groundwater types and experimental methods

Groundwater (after greensand treatment) provided by Dr. Mulligan from 10L and 20L containers was mixed well together and tested for concentrations of water compositions on the receiving day, containing  $55.05 \pm 0.94$  ppm calcium,  $7.85 \pm 0.58$  ppm magnesium,  $163.5 \pm 0.5$  ppm bicarbonate, and neglected amount of other chemical (iron, phosphate, etc.) and biological water components defined as raw water 1 (RW1). RW1 was used to prepare all types of artificial water in batch and column tests of this laboratory research. All chemicals in this study were ACS grade purchased from VWR and Fisher Scientific.

#### *3.1.3.1 Artificial groundwater spiked with BPY for ferrous (Fe (II)) measurement*

2,2-bipyridine (BPY,  $(C_5H_4N)_2$ ) is a chelating ligand forming  $[Fe(BPY)_3]^{2+}$  complexes with Fe(II) at pH > 4, resulting in a red color with a maximum absorption at  $\lambda = 522$  nm which can be measured by a UV-Visible Spectrophotometer (Wenk, 2008). In this study, BPY was spiked in water samples in excess (1.0 mM) to measure the maximum amount of Fe(II) produced by iron corrosion in a water sample.

#### *3.1.3.2 Artificial groundwater for column tests*

In order to evaluate the impacts of water compositions on 1) arsenic removal by iron corrosion products and 2) total arsenic removal efficiency of the lab-scale filter, four types of artificial groundwater denoted as GW1, GW1-P, GW2, GW2-P were used to run column tests in the lab-scaled filters until the filters started to clog. Water preparation is described below.

Artificial groundwater was prepared by spiking a small amount of stock solutions of high purity chemicals to tap water. Water compositions of all water types are listed in Table 2. Water components such as calcium, magnesium, phosphate, silicate, arsenic, chloride and alkalinity are defined as Ca, Mg, P, Si, As, Cl and  $HCO_3^-$ , respectively. When preparing artificial groundwater,

Si-stock solution was added first to the tap water when the pH is low to avoid precipitation while As (III)-stock solution was added last, right before dosing water to filters, to minimize pre-oxidation.

Stock solutions were prepared by dissolving corresponding chemicals with DI water in a volumetric flask and filling up to the mark. DI water used in this study was MilliQ DI water with a resistance of 18.2 and total organic carbon (TOC) of 3  $\mu\text{g/L}$ . The hardness stock solution containing 14.48 g/L Ca and  $4.15 \times 10^3$  g/L Mg was prepared in a 1 L volumetric flask by dissolving 79.26 g  $\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$  and 35.13 g  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  with DI water to the mark. The P-stock solution containing 2.0 g/L P was prepared in a 1 L volumetric flask by dissolving 8.8 g  $\text{KH}_2\text{PO}_4$  with DI water to the mark. The Si-stock solution containing 1.0 g/L Si was prepared in a 500 mL volumetric flask by dissolving 5.1 g  $\text{Na}_2\text{SiO}_3 \cdot 9\text{H}_2\text{O}$  with DI water, adjusting pH with 36.5 mL HCl 1M and adding DI to the mark. The final pH of Si-stock solution was 3.88. The As (III)-stock solution containing 100 mg/L As (III) was prepared in a 1 L volumetric flask by dissolving 0.132 g  $\text{As}_2\text{O}_3$  with 10 mL of NaOH 10% solution, diluting with DI water and acidifying the solution with 50 mL of  $\text{HNO}_3$  1M. DI water was then added to the mark. The Cl-stock solution containing 37.9 g/L  $\text{Cl}^-$  was prepared in a 1 L volumetric flask by dissolving 72.5 g KCl with DI water to the mark. Cl-stock solution was added to GW1 and GW1-P (water components denoted in Table 2) so that the amount of chloride in GW1 and GW1-P was equivalent to that of GW2 and GW2-P as a byproduct of the dissolution of hardness stock solution ( $\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$  and  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ ).



Table 2. Water compositions used in experiments (main variables are in bold)

Water type	Composition (chemical concentrations in mg/L)	Preparation for 1L of corresponding solution
RW1	55 Ca + 7.8 Mg + 163.5 HCO <sub>3</sub> <sup>-</sup>	Original tap water, no chemical added
RW1-P	55 Ca + 7.8 Mg + 163.5 HCO <sub>3</sub> <sup>-</sup> + <b>2 P</b>	1 mL of P-stock spiked to RW1 solution
RW2	<b>200 Ca + 50 Mg</b> + 163.5 HCO <sub>3</sub> <sup>-</sup>	10 mL of hardness-stock spiked to RW1 solution
RW2-P	<b>200 Ca + 50 Mg</b> + 163.5 HCO <sub>3</sub> <sup>-</sup> + <b>2 P</b>	10 mL of hardness-stock, 1 mL of P-stock, and RW1 solution filled to the mark
<b>GW1</b>	55 Ca + 7.8 Mg + 380 Cl + 163.5 HCO <sub>3</sub> <sup>-</sup> + 20 Si + 2 As (III)	20 mL of Si-stock, 10 mL of Cl-stock, 20 mL of As (III)-stock and RW1 solution filled to the mark
<b>GW1-P</b>	55 Ca + 7.8 Mg + 380 Cl + 163.5 HCO <sub>3</sub> <sup>-</sup> + 20 Si + 2 As (III) + <b>2 P</b>	20 mL of Si-stock, 10 mL of Cl-stock, 1 mL of P-stock, 20 mL of As (III)-stock, and RW1 solution filled to the mark
<b>GW2</b>	<b>200 Ca + 50 Mg</b> + 380 Cl + 163.5 HCO <sub>3</sub> <sup>-</sup> + 20 Si + 2 As (III)	20 mL of Si-stock, 10 mL of hardness-stock, 20 mL of As (III)-stock and RW1 solution filled to the mark
<b>GW2-P</b>	<b>200 Ca + 50 Mg</b> + 380 Cl + 163.5 HCO <sub>3</sub> <sup>-</sup> + 20 Si + 2 As (III) + <b>2 P</b>	20 mL of Si-stock, 10 mL of hardness-stock, 1 mL of P-stock, 20 mL of As (III)-stock, and RW1 solution filled to the mark

### 3.2 Experimental setup, sample collection and measurement for batch tests

Two sets of batch experiments were conducted to examine the corrosion rate of the iron nails in this laboratory research. The first batch experiments were to compare iron corrosion rates of ZVI-power (Katsoyiannis et al., 2008), iron nails used in a previous study (Wenk, 2008) and iron nails in this study. The second batch experiments were to determine the linearity and maximum corrosion rate of iron nails in different type of water (Wenk, 2008). All batch experiments were run in triplicate. When iron rusts in the presence of exceeding BPY, the newly formed  $\text{Fe}^{2+}$  will react with BPY producing the red colored complex which then be measured by a UV visible spectrophotometer at  $\lambda = 522$  nm to determine the corresponding amount of Fe(II) in the sample.

In the first set of batch experiments, half of a nail (around 225.6 mg) was put into a plastic cuvette containing 3.82 mL artificial groundwater RW2-P (water component and preparation described in section b, Table 2) spiked with 1.0 mM BPY. The cuvette was inserted to the UV-Visible spectrophotometer every 10 minutes to measure Fe(II) amount at different timepoints for a testing duration of 120 minutes (Wenk, 2008).

In the second set of batch experiments, 5 opaque bottles containing 50 mL of artificial ground water spiked with 1mM BPY were added 1, 2, 4, 8, and 16 nails, respectively. The bottles were closed and slowly shaken at 60 rpm. Every 10 minutes, 3 mL aliquots were taken from the bottles to a plastic cuvette and measured by the UV-Vis spectrophotometer. After analysis, the samples were put in to the corresponding bottles to keep the volume of water in the 50mL bottles unchanged (Wenk, 2008).

In this study, Fe(II) measurement of all water samples was performed by the UV-Visible Spectrophotometer Evolution 201 (Thermo Scientific) by measuring maximum absorption of Fe(II)-BPY red complexes at  $\lambda = 522$  nm.

### 3.3 Equipment design and implementation for column tests

Lab-scale KAFs in this study were resized based on the column design by Young-Rojanchi and Madramootoo (2015). The columns were expected to be representative of the vertical dimension of the KAF version 10 introduced by CAWST Biosand filter construction manual in 2012. The columns were made of 5 cm diameter transparent plastic tubing for easy installation and economical purpose. They were then covered in aluminum foil and tape during the study period to 1) prevent algal growth which may create anaerobic conditions and use up dissolved oxygen (DO) in water, 2) avoid direct sunlight in order to inhibit possible photooxidation process of arsenic compounds, and 3) reduce temperature difference between filters caused by the distance between their locations and the laboratory heating/air conditioning systems. Fifteen lab-scale filters underwent operating tests to ensure their fundamental parameters meet the requirements of the KAF version 10 design by CAWST (2012). Three filters were employed as control filters and twelve filters treated artificial groundwater of four types described in Table 3.

Table 3. Information of filters used in the study

Filter	Nails	Role	Water type	Main variations
#12	No nails	reference	RW1	
#3	136.5 g	reference	RW1	
#1	136.5 g	reference	RW2-P	
#8; #11; #6	136.5 g	testing	GW1	low hardness, no phosphate
#13; #4; #7	136.5 g	testing	GW1-P	low hardness, high phosphate
#10; #9; #14	136.5 g	testing	GW2	high hardness, no phosphate
#5; #15; #2	136.5 g	testing	GW2-P	high hardness, high phosphate

Figure 4 demonstrates the experimental set up of lab-scale KAFs in this study. Twelve filters of four water types (three replicates for each set) were numbered and located randomly to minimize the impact of environmental conditions on a filter set of a particular water type.



Figure 4. Experimental setup of lab-scale KAFs. From left to right, the order of filters is #8, #5, #13, #10, #15, #11, #4, #9, #2, #6, #14, #7. A small tube with tubing clamp was inserted in to each filter at about 3 cm above the surface of filter media to collect supernatant water samples. Effluent water was still collected at the outlet tube respecting the siphon effect.

Before using for water treatment, all lab-scale filters were checked for two most critical operating parameters: the filter loading rate and permeability of filter media.

Filter loading rates (also called as “hydraulic loading rate” or “filtration rate”) were checked by hydraulic loading tests. According to CAWST (2012), a standard KAF version 10 should have a filter loading rate of no more than 400 liters per hour per square meter (400 L/h/m<sup>2</sup>) of sand surface area. With a diameter of 5 cm, the sand surface area of a lab-scale filter in this study is  $1.96 \times 10^{-3} \text{ m}^2$ . Therefore, the maximum filter loading rate (vertical flow rate) of the lab-scale filter should be 0.66 cm/min or 0.785 L/h (equivalent to 13 mL/min). The filter loading rate was measured by the stopwatch app in an iPhone 6S and a 25 mL glass graduated cylinder. Raw water was fed to the 17 cm mark on the loading head of a filter while the outlet tube was stopped by a researcher’s finger. The stopwatch was started when the 17 cm loading head was reached, and the outlet tube was freed. If the filter loading rate was lower than 13 mL/min, the filter passed this test and underwent the tracer test. If the filter loading rate were higher than 13 mL/min, the filter media (fine sand, coarse sand) would be discharged and reloaded with a smaller size and lower uniformity.

The permeability of filter media and behavior of each filter were determined by tracer tests. The testing methods followed the methodology of Young-Rojanschi (2015). Two sets of tracer tests were conducted using sodium chloride (NaCl) solution of 200 mg/L as the tracer. Filters were rinsed several times with deionized water before testing. For each test, a dose of 450 mL of tracer was added, followed by three doses of deionized water. Each new dose was added after at least 425 mL of water of the last dose was filtered. Samples were collected from effluent tube of a filter for each 25 mL and then measured for electrical conductivity (EC) to calculate the Morrill Dispersion Index (MDI). Elliot et al. (2015) described that “the MDI is the ratio of  $T_{90}$  and  $T_{10}$ , where  $T_{90}$  is the time to reach 90% and  $T_{10}$  is time to reach 10% of the feed concentration in the

exit stream. A reactor that exhibits ideal plug flow would have an MDI of 1.0. An MDI of less than 2.0 is classified as “effective plug flow” by the US EPA”.

In the first set of tracer tests, lab-scale filters were tested for short-circuiting by dosing tracer after removing the standing water on the surface of the sand. In the second set, the filters worked under normal operating conditions to be tested for their behavior.

Fifteen filters with a filter loading rate of less than 400 L/h/m<sup>2</sup> of sand surface area and MDI less than 2.0 were employed for the column tests. Based on the pore volume of filter media and the amount of supernatant water (water resting on top of the sand surface after contacting with iron nails) taken in sampling, 500 mL of artificial groundwater were poured into the filter in one dose. However, due to the small volume of the filter reservoir, a full dose could not be fed all at once but were filled up to a 17 cm mark on the loading head. The remaining water was added gradually when the water level lowered by approximately 3 cm (Young-Rojanschi et al., 2015). The filters were fed one dose a day over a period of thirty-five consecutive days (until the filters started to clog, with filter loading rate of less than 6 mL/min). No maintenance was done for the filters during the duration of water treatment to avoid disturbing experimental conditions.

### 3.3 Methods of sample collection and analysis for the column experiments

#### 3.3.1 Sample collection

Water sampling was done once a day in the first week, twice a week in the second week and then once a week for the rest of the treatment over thirty-five consecutive days. On the first day, all filters were flushed with three doses of corresponding artificial water. Influent water samples were collected in the fourth dose for measuring physical parameters. Supernatant water samples were collected to the mark of 15 mL plastic tubes containing 0.75 mL BPY 20 mM for Fe(II) measurement and 50 mL plastic tubes preserved with 1 mL of concentrated HNO<sub>3</sub> added for

instrumental analysis of other water components. Effluent water samples of the corresponding day were collected the day after to that of the influent and supernatant water samples. Effluent water from outlet tubes was collected in clean 500 mL glass flasks in about two hours so that the filtration was close to be finished before being mixing well and collected to 50 ML plastic tubes. Supernatant water was sampled after effluent water sampling to avoid disturbing water treatment process. Water samples were placed in a cool and dark place to prevent further oxidation of water components.

### 3.3.2 Sample measurements and analysis

#### 3.3.2.1 *Physical parameter measurements*

pH and temperature of influent and filtered water samples were measured by the Oakton Electronic pH Meter right after sampling. Calibration was done every day before field tests by using 3 standard solutions of pH 4.0, 7.0 and 10.0.

#### 3.3.2.2 *Instrumental analysis*

The analysis of Fe(II) levels in supernatant water spiked with exceeded BPY were performed by the UV-Vis Spectrophotometer Evolution 201 (Thermo Scientific) at  $\lambda = 522$  nm within an hour after sample collection (Wenk, 2008).

Total iron, calcium and magnesium concentrations in all water samples were analyzed by the Flame Atomic Absorption Spectrometer (F-AAS), PinAAcle 900F model (Perkin Elmer). Liquid samples were filtered through a 0.43  $\mu$ m filter and diluted with an acid solution of 4% HNO<sub>3</sub> for AAS analysis.

Inductively coupled plasma mass spectrometer (ICP-MS) Agilent 7700 (Tokyo, Japan) was used to analyze arsenic, silicate and phosphate concentrations of water samples. Liquid samples were filtered through a 0.43  $\mu\text{m}$  filter and acidified with an acid mixture of 1%  $\text{HNO}_3$  and 0.5%  $\text{HCl}$  for the ICP analysis.

The images and chemical elemental compositions of the surface of a clean nail and four contaminated nails representing four artificial groundwater types were analyzed by the Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM/EDS) Hitachi S-3400N. Iron corrosion powder of these samples was analyzed by X-ray Diffraction (XRD) using the Philips X'Pert Pro Multipurpose X-ray Diffractometer. The Pearson's Crystal Database was used to analyze XRD patterns for phase identification and crystal structure of contaminants.

Transmission Electron Microscopy (TEM) was done at Facility for Electron Microscopy Research (FEMR), McGill University. A few  $\mu\text{L}$  of a supernatant water sample were filtered through a nanomembrane in which suspended solids were retained and undisturbed for a minute before TEM analysis. The colloids were analyzed by TEM for the phase, image and structure of iron corrosion products in the samples.

## B. Field research in the Nawalparasi district, Nepal

### 3.4 Research methodology

Literature review, laboratory-scale experiments, data analysis, and project plans took place from June to September 2018 to determine 1) representative KAFs to study in Nawalparasi, 2) significant factors affecting removal efficiency of the KAF, and 3) the hypothetical influence of those factors on KAF performance. Based on data analysis of an on-going water quality assessment in Nawalparasi for 3 years, CAWST did some modifications for a set of chosen KAFs. Those



KAFs were employed in the field research in late September to early October 2018 for testing filter parameters, water quality, and filter performance. During the sampling and measuring time, KAF users were invited to answer interview questions as well as demonstrate their methods of operating and maintaining the KAF.

In February 2018 (seven months before the trip), the Research and Learning team at CAWST collaborated with a Japanese researcher and the Environment and Public Health Organization (ENPHO) to conduct a set of experiments on 12 KAFs of 12 volunteer families. The studied households were chosen by their geographic location using the same water source or having similar input water quality. The chosen KAFs were divided into three main groups:

Group A (new nails + embedded nails): 4 filters using new nails in the diffusion basin and newly changed filter media with embedded nails in the sand.

Group B (new nails): 4 filters using new nails in the diffusion basin and newly changed filter media.

Group C (old nails): 4 filters using old nails with newly changed filter media (sand and gravels with the standard sizes). Those KAFs were monitoring filters.

The modifications were operated to study the effects of iron nail type and its location on the performance of KAF. Therefore, the results of water quality in three filter groups were compared in two pairs as follows.

- 1) Raw water (RW), supernatant water (SW), and filtered water (FW) of group B compared to those of group C to analyze the impact of nail types on filter performance, and
- 2) RW and FW of group A compared to those of group B to analyze the influence of extra nails embedded in filter media on arsenic removal.

However, 2 of the 12 households in 2 groups of the experiments stopped using their KAFs for some personal reasons. Hence, the study was continued as a set of 9 filters with 3 filters in each group with sampling and testing processes for water quality of water source and filter water. KAF users of those 9 households were also invited to participate in an interview at the beginning of the field study.

The interview of KAF users consisted of numerous open-ended questions for the people mostly in charge of using and maintaining the KAF in a family. Interviewees were asked about health conditions of the family members, their estimated daily amount of water fed to the KAFs, equipment to feed and collect water, purposes of filtered water usage, the satisfaction level of the KAFs regarding water quality, filter appearance, and the convenience of operation and maintenance the KAFs.

Water quality assessment in Nawalparasi was done by on-field tests for physical parameters and laboratory tests for chemical parameters of water samples. Water samples consisted of source water and household water from KAFs (including supernatant water and filtered water).

### 3.5 Sampling methods

Water sampling and filter testing were done over five consecutive days. All households in the scope of the study were asked to use their KAFs every day during the testing period without any disruption for any reasons, maintenance included. On the first day, source water from different tube wells (from now on called raw water) was taken for sampling and flushing the corresponding KAFs. Meanwhile, flow rates of KAFs were checked. Raw water collecting from tube wells and fed to KAFs were done by the users for the researchers to observe using habit of each household. Interviews of KAF users were conducted while filters were working. The environment and public

health organization (ENPHO) staff was in charge of translation for the interview and communication between the English-speaking researchers and Nepali KAF users.

On the second day, supernatant water (resting on the top of the sand surface after contacting with iron nails) and filtered water were collected from a KAF and tested immediately for physical parameters before a new dose of raw water was fed. Raw water was collected for measuring its conditions and parameters, then feeding the KAF. Supernatant water and filtered water samples were tested and collected later on the same day. The same procedure of sampling and testing was done until the study was finished.

If a tube well was the source of groundwater of more than one KAFs, the daily raw water from that source would be tested only once, the corresponding supernatant water in the corresponding filter would be tested once, but the filtered water of all KAFs would be tested. The sampling method was applied due to an assumption that the raw water conditions would not change dramatically in a couple of hours, and the oxidation-reduction process occurring when contaminated water contacting with iron nails in a reservoir would be the same for every KAF using the same water source.

### 3.5.1 Filter check

#### *3.5.1.1 Checking requirements*

The studied KAFs were checked based on some criteria established by CAWST (2012) for suitable working conditions:

- i. Good location. Standards: clean; stable; away from animals; not directly exposed to sunlight and bad weather conditions such as rain, storm, thunder, etc.

- ii. Sustainable appearance. Standards: filter parts (such as the lid, diffuser, filter body, and outlet tube) with no damage and fitting perfectly together.
- iii. Acceptable operation. Standards: KAFs are used every day; input water is not very cloudy or dirty (turbidity is less than 50 NTU); iron nails rust and are distributed evenly in the diffuser; the surface of the sand is flat and level; the supernatant water depth is from 4 to 6 cm; the flow rate of the filter is between 300 to 400 mL/minute; the filtered water is clean without a bad taste or smell; no tap or hose was attached to the outlet tube of the filters.
- iv. Sufficient maintenance. Standards: filters are clean; no leakage, clogging or broken parts observed in the filters; the outlet tubes do not have dirt or algae.
- v. Safe storage. Standards: each household has separate containers for raw and filtered water, the filtered water storage container has a lid and tap or hose for users to collect water easily, the storage container is free from dirt, algae or stains, and is frequently cleaned.

#### *3.5.1.2 Filter checking methods*

Most of the above mentioning factors were obtained by interviewing KAF users and observations. Two filter parameters requiring measurement were filter flow rate and the standing water depth on the surface of the sand.

The filter flow rate was measured by a stopwatch and a 500 mL plastic graduated cylinder. A full dose of 12 L of raw water was fed to the filter while the outlet tube was stopped by a researcher's finger. The stopwatch was started when the maximum loading head was reached, and the outlet tube was freed. If the flow rate were less than 300 mL/min, a swirl and dump maintenance would be done. If the flow rate was more than 400 mL/min, the filter did not work properly.

Supernatant water depth was measured by a ruler. If the water depth were less than 4 cm, sand would be scooped out from the filter, and the sand surface was then flattened and leveled. If the water depth were more than 6 cm, sand would be added to the filter.

As a significant factor expected to affect the KAF performance strongly, corrosiveness of iron nails in each filter was tested by two categories of criteria:

- Cohesion: how strongly the iron nails stick to each other, graded by alphabet characters as follows.
  - A. Completely loose, no nails stick together
  - B. Two or three nails stick together loosely
  - C. Two to three small clusters of nails are observed
  - D. Larger clumps
  - E. All one solid piece
- Corrosive states: how rusty the iron nails are, indicated by the colour of nails which was graded by numbers as follows.
  1. Shiny, chrome original black
  2. Black rust
  3. Slight, some spots of orange
  4. 50% orange
  5. Completely orange/brown (visible corrosion)

### 3.5.2 Raw water sampling and testing

Raw water of KAFs was collected from pump tube wells nearby the households by the KAF users. Some tube wells required a small amount of water poured into the pump to initiate the pumping mechanism. In this case, the pumped groundwater was mixed with the triggering water and

changed its original conditions. Hence, the raw water was collected after at least first 30 seconds of pumping to ensure its natural characteristics and freshness.

Once a day in the morning, 12 L of raw water were collected in a 15 L bucket to feed the KAF and the water was tested immediately for dissolved oxygen (DO) concentration, followed by other physical parameters such as pH, temperature, turbidity, oxidation-reduction potential (ORP), and electrical conductivity (EC). In addition, all raw water samples were stored in clean 250 mL opaque plastic sample bottles containing some drops of concentrated nitric acid to reserve for total arsenic measurement, and in 500 mL clean opaque plastic bottles for measurements of the main water components (iron, phosphate, manganese, calcium hardness, and total alkalinity). Sample bottles were then sent to the ENPHO laboratory (Kathmandu, Nepal) for instrumental analysis. KAF users were asked not to use the filters during the testing time until all water samples were collected.

### 3.5.3 Household water sampling and testing

Household water consisting of supernatant water and filtered water was collected from the KAFs about 2.5 hours after dosing when the filtered water from the outlet tube stopped running or was just dripping very slowly.

The DO level of supernatant water was tested first, followed by other physicochemical parameters (pH, temperature, turbidity, EC, and ORP) without stirring the standing water and disturbing the sand surface. Next, supernatant water samples of KAFs (except for filters number 2, number 11, and number 13) were collected stored in clean 250 mL opaque plastic sample bottles containing some drops of concentrated nitric acid to reserve for total arsenic measurement, and in 500 mL clean opaque plastic bottles for measurements of iron, phosphate, and calcium hardness

concentrations in the ENPHO laboratory. Those water components were believed to have an impact on arsenic removal and/or the corrosion of iron nails.

It was requested that the filtered water from every KAF remain untouched in a 15 L clean bucket in each household until the research group returned for measuring and sampling. After the filtered water was tested for the DO level, it was stirred vigorously in the bucket to be homogeneous. Other physical parameters were then tested. In the meantime, all filtered water samples were collected for arsenic, iron, manganese, and phosphate concentration measurements in the ENPHO laboratory using the same procedure mentioned in raw water sampling.

Alum stones locally purchased with a weight of approximately 50 g per each stone were added to raw water with high turbidity to remove suspended solids in the water. Water samples after flocculation were collected and tested for their composition to see if it was a good pretreatment step for future use.

Water sample data and individual notes were recorded promptly on a CAWST iPad using a Fulcrum data collection platform built by Candice Young-Rojanchi. Samples of raw water, supernatant water and filtered were collected, labeled and numbered in a specific way denoted by the researchers of this study. Water samples were then separated into different groups of testing requirements and their identification was hidden from ENPHO lab technicians for a fair analysis.

### 3.6 Testing methods and apparatus

#### 3.6.1 In-situ tests

During the research trip in Nawalparasi 2018, field tests were conducted for physicochemical analysis of all water samples. Water parameters such as dissolved oxygen (DO), temperature, turbidity, electrical conductivity (EC), pH, oxidation-reduction potential (ORP) were measured

before water samples were taken for laboratory tests. After DO measurements, the water samples were homogenized before other tests by a clean ladle.

Dissolved oxygen level in the water was the first factor to be measured in any water sample. The measurement took place in the field using a dissolved oxygen pen, model 850045 from Sper Scientific LTD. DO probe was calibrated every day before field tests. The temperature of water samples was recorded at the same time the DO level was read as the DO probe including a temperature sensor. The temperature displayed on the screen of the DO pen during the DO measurement took place.

Water samples were collected in a sample cell and measured for turbidity by the Portable Turbidimeter 2100 from the HACH Company. The electrical conductivity of water samples was measured in the field by the Traceable Conductivity/TDS Meter purchased from VWR. The pH of water samples was measured in the field by the Potatest pH Meter by Wagtech. Calibration was done every day before field tests by 3 standard solutions of pH 4.0, 7.0 and 10.0. The oxydation-reduction potential (ORP) of water samples was measured by the Pocket Pro+ ORP tester by HACH company. Calibration and measurements were done every day according to the user manual.

### 3.6.2 Laboratory tests

Chemical analysis for total arsenic, iron, calcium hardness, manganese, phosphate and total alkalinity in the collected water samples took place in the EPHO laboratory accredited by the Nepal Bureau of Standards and Metrology (NBSM).

The total arsenic level of Nawalparari water samples was measured by Atomic Absorption Spectroscopy (AAS) applying the Manual Hydride Generation/Atomic Absorption Spectrometric



method (3114 B) of the Standard Methods for the examination of Water and Wastewater (APHA, AWWA & WEF, 2012). Water samples for arsenic measurements were preserved by a few drops of concentrated nitric acid to maintain an acidic environment of about 2% HNO<sub>3</sub>. Arsenic concentrations were measured for raw water and filtered water samples.

Samples of the following water compositions did not need to be preserved. Iron and manganese concentrations were both determined by Flame Atomic Absorption Spectroscopy (F-AAS) using the Direct Air-Acetylene Flame method (3111 B) of APHA, AWWA & WEF (2012). Phosphate levels in water samples were measured by the AAS using the Manual Digestion and Flow Injection Analysis for Total Phosphorus (4500-P H) of APHA, AWWA & WEF (2012). Calcium hardness concentrations were analyzed by the ethylenediaminetetraacetic acid (EDTA) titrimetric method (3500-Ca B) and total alkalinity as CaCO<sub>3</sub> levels were quantified by Titration Method (2320 B) of APHA, AWWA & WEF (2012).

## CHAPTER 4. Results and Discussion

### A. Laboratory research at Concordia University

#### 4.1 Results from the lab research

##### 4.1.1 Representativeness of lab-scale filters as a vertical-cross section unit of the KAF

The lab-scale filters were built to represent the vertically cross-sectional design of the full-scaled KAF. Therefore, the originally designed factors regarding volume the KAF could not be kept. For example, the diffuser reservoir was designed to have a height of 17 cm (hydraulic head) and a volume of 12 L (which was equal to the void volume of the filter media). Although the lab-scale filters had the same hydraulic heads of 17 cm, the volume of the reservoirs was about 333 mL, less than the volume of the pore water in the filter media of 500 mL. Hence, a dose of water could not be fed to a lab-scale filter all at once. The remaining water of a dose was poured gradually as water was filtered. Consequently, the filtration rate might be slightly affected.

Data collected from the hydraulic loading tests showed that all filters in this study had a filter loading rate of 400 L/h/m<sup>2</sup> (equivalent to a vertical flow rate of less than 0.66 cm/min or 13 mL/min with a filter of 5 cm in diameter, as calculated in section 3.3) which is the CAWST standard for a KAF version 10. The range of flow rate of fifteen filters is  $12.5 \pm 0.3$  ml/min at the hydraulic loading head of 17 cm.

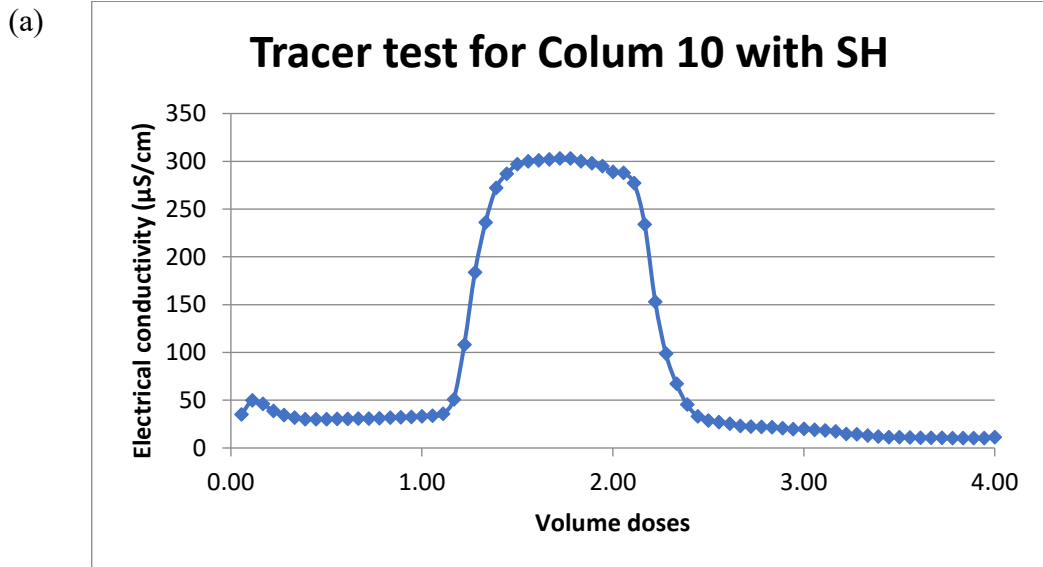
The plug flow behavior of filters in this study was determined by calculating their Morill Dispersion Index (MDI) in tracer tests. MDI of 1.0 represents perfect plug flow, MDI < 2.0 corresponds to effective plug flow (Mark et. al, 2008). Table 4 gives the MDIs of all fifteen filters for short-circuiting and under operational conditions. The tracer tests with standing head (SH) (A)

were to characterize the function of the filters in the operational conditions of the experiment. The tracer tests without SH (**B**) were for short-circuiting.

Table 4. Morrill Dispersion Index for tracer tests of lab-scale filters with and without SH.

#	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
<b>A</b>	1.5	1.5	1.5	1.5	1.5	1.4	1.4	1.4	1.4	1.4	1.3	1.3	1.5	1.5	1.4
<b>B</b>	1.2	1.2	1.3	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.3	1.3

The short-circuiting test results (**B**) prove that the lab-scale filters were plug flow reactors as full-scale BSFs, with an MDI of 1.2. Under experimental conditions, with the standing head (**A**), MDIs of the filters were higher, in the range of  $1.4 \pm 0.1$ . The small deviation in the MDIs of the filters suggests that the set of fifteen filters was under the same operating conditions and functioned similarly to each other.



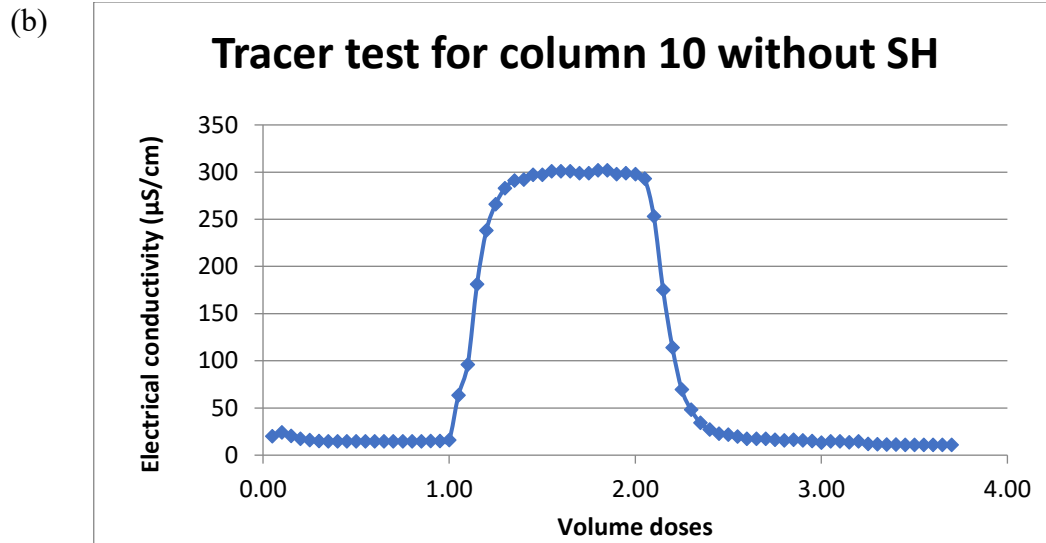


Figure 5. Tracer test results of a representative filter (a) with the standing head (SH) and (b) without the SH

The tracer test results of filter 10 with and without the standing head are used as a representative for the set of lab-scaled filters (Figure 5). Although the tracer was introduced to the filter in the first dose of influent water, no tracer was detected in the effluent. Almost all of the tracer was collected in the second dose and the beginning of the third dose. Therefore, in this study, influent and supernatant water samples were collected on the dosing day and effluent water samples of the corresponding dose were collected the day after.

#### 4.1.2 Influent water characterization

The temperature of artificial groundwater was at room temperature, which was  $20.9 \pm 0.8$  °C. pH of each water type was not adjusted to a fixed level and was measured every day before feeding the corresponding filters. Average pH over the water treatment period of RW1, RW2-P, GW1, GW1-P, GW2, and GW2-P were  $7.45 \pm 0.12$ ,  $7.58 \pm 0.15$ ,  $7.68 \pm 0.17$ ,  $7.71 \pm 0.16$ ,  $7.54 \pm 0.26$ , and  $7.53 \pm 0.29$ , of which, RW1 and RW2-P were influent water of control filters.

The possibility of calcium carbonate scale in GW1 and GW2-P (related to the corrosivity of water) was predicted by their Langelier Saturation Index (LSI). The LSI was calculated by the difference of actual pH of the solution (measured by a pH meter) and pH<sub>s</sub> (the pH at saturation in calcium carbonate calculated from temperature, TDS, concentrations of calcium hardness and total alkalinity) as follows (Dorner, 2015):

$$\text{LSI} = \text{pH}_{\text{actual}} - \text{pH}_s \quad (9)$$

$$\text{pH}_s = (9.3 + A + B) - (C + D) \quad (10)$$

Where:

$$A = (\log_{10} [\text{TDS}] - 1) / 10 \quad (11)$$

$$B = -13.12 \times \log_{10} (t \text{ } ^\circ\text{C} + 273) + 34.55 \quad (12)$$

$$C = \log_{10} [\text{Ca}^{2+} \text{ as CaCO}_3] \quad (13)$$

$$D = \log_{10} [\text{alkalinity as CaCO}_3] \quad (14)$$

LSI < 0: the water will dissolve CaCO<sub>3</sub>, no potential to scale

LSI = 0: borderline scale potential

LSI > 0: CaCO<sub>3</sub> precipitation may occur, scales can form

The LSI of GW1 and GW2-P were 0.7 and 1.5, respectively. This means the artificial groundwater was nonaggressive and CaCO<sub>3</sub> may deposit.

#### 4.1.3 Batch experiments

##### 4.1.3.1 Comparison of iron corrosion rates of materials used in different studies

The corrosion rate of iron nails used in the lab experiments of this study was compared to those of iron nails and zero-valent iron (ZVI) powder in previous studies done by Wenk (2008) and Katsoyiannis et al. (2008), respectively. Fe (II) formation in the presence of exceeded bipyridine (BPY) was the indicator of the iron corrosion rates.

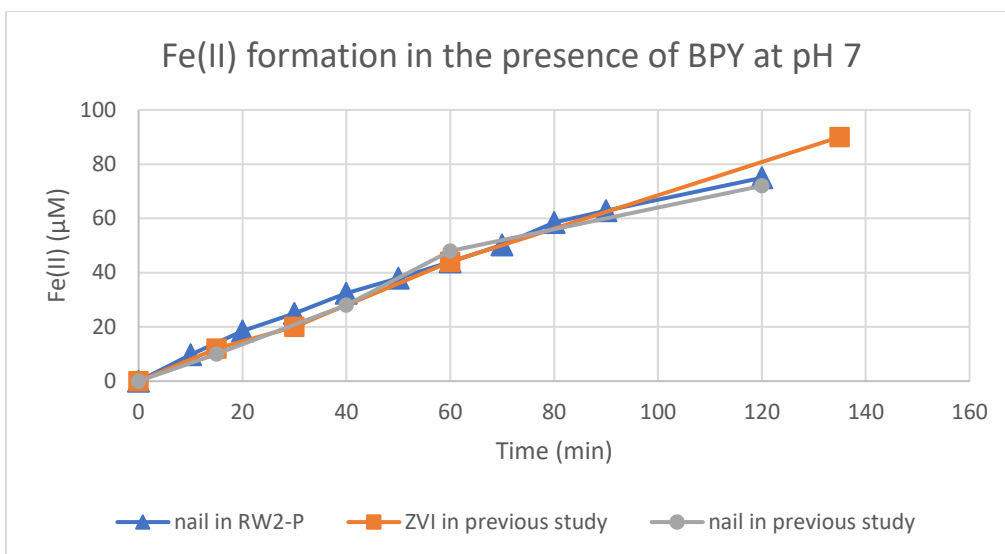


Figure 6. Similar Fe(II) formation rates of different iron products in the presence of BPY at pH 7. Previous study data were retrieved from Katsoyannis (2008) and Wenk (2008), respectively.

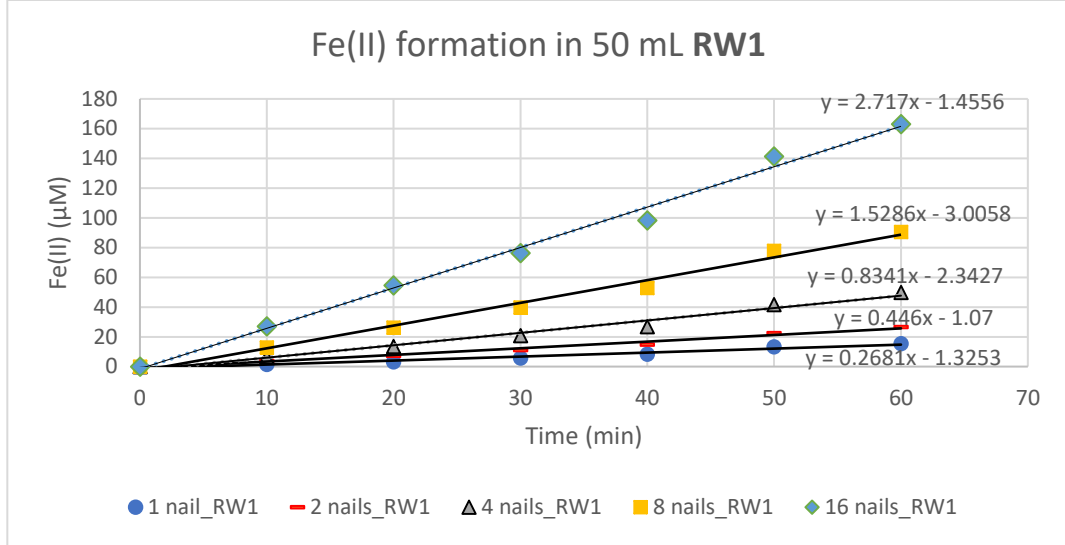
Katsoyiannis et al. (2008) used ZVI with the specific surface area of approximately  $0.1 \text{ m}^2/\text{g}$ , ZVI level in the contaminated water containing  $1 - 3 \text{ mM}$  BPY was  $150 \text{ mg/L}$ , resulting in an activate iron surface of  $15 \times 10^{-3} \text{ m}^2/\text{L}$  or  $15 \text{ mm}^2/\text{mL}$ . The same surface area of iron nails per water volume was used in this experiment, as in Wenk (2008). The result of this batch experiment indicated that all tested iron materials had corroded at a similar rate in the corresponding environment (Figure 6). However, after 90 min, corrosion rates of two types of iron nails started to reduce slightly while that of ZVI kept unchanged. This may explain the fact that ZVI still had more active sites to react with water and BPY at that moment, even though all materials had the same theoretical exposed surface area per volume.

#### 4.1.3.2 Corrosion rates of iron nails in contaminated water without the presence of arsenic

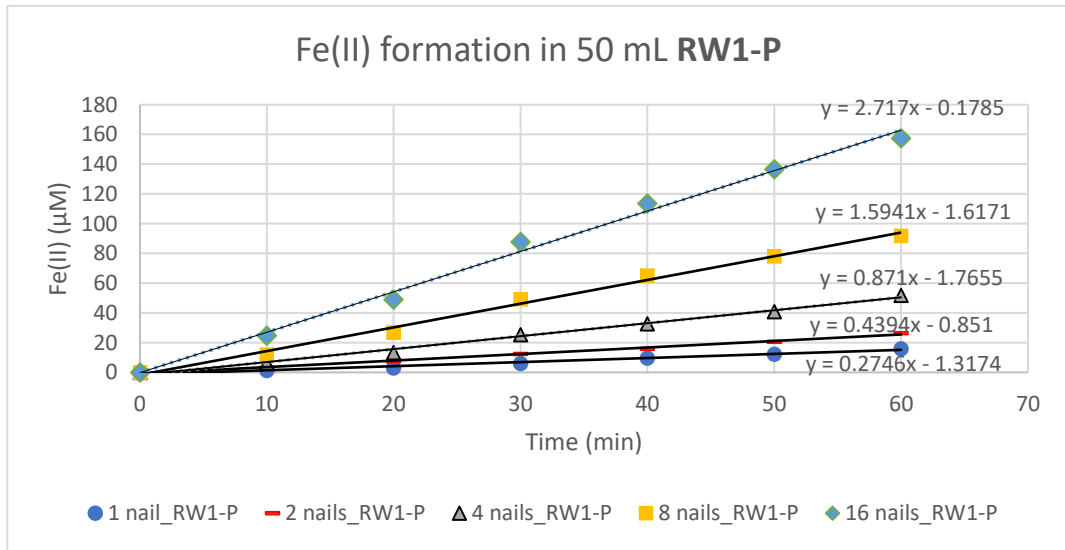
The corrosion rate of iron nails in contaminated water with variable levels of hardness and phosphate in the absence of arsenic was analyzed based on the formation of Fe(II) in 50 mL of RW1, RW1-P, RW2, and RW2-P spiked with  $1 \text{ mM}$  BPY. Figure 7 demonstrates corrosion rates

of 1, 2, 4, 8, and 16 nails in four water samples as a function of Fe(II) levels and time, the amount of Fe(II) produced in the same water volume increases as the number of nails increases (proportional to an increase in the surface area of iron), but does not follow a linear relationship. Corrosion rates of iron nails slowed down over time as a result of the formation of a passive corrosion layer (Katsoyiannis et al., 2008). The average corrosion rate of nails was defined as the average of surface-normalized rates of Fe(II) formation over time in the studied water sample. The purpose of these experiments is to examine the effect of hardness and phosphate per se on Fe(II) formation without the presence of other contaminants such as arsenic and silicate. RW1 and RW1-P have a low total hardness of approximately 55 mg/L Ca and 8 mg/L Mg with respectively 0 mg/L and 2 mg/L of P, while RW2 and RW2-P have high total hardness of approximately 200 mg/L Ca and 50 mg/L Mg with 0 mg/L and 2 mg/L of phosphate, respectively. Of the four water types, iron nails in RW2 had the highest value of average corrosion rate ( $1.22 \pm 0.30 \mu\text{mol/s}\cdot\text{m}^2$ ). Nails in RW1 and RW1-P had similar average corrosion rates of  $0.96 \pm 0.15$  and  $1.00 \pm 0.17 \mu\text{mol/s}\cdot\text{m}^2$ , respectively. RW2-P had the lowest value of average corrosion rates ( $0.89 \pm 0.15 \mu\text{mol/s}\cdot\text{m}^2$ ). This illustrates that hardness plays a significant role in facilitating iron corrosion while phosphate does not show a noticeable impact at low hardness level but inhibits corrosion rate at high hardness.

(a)

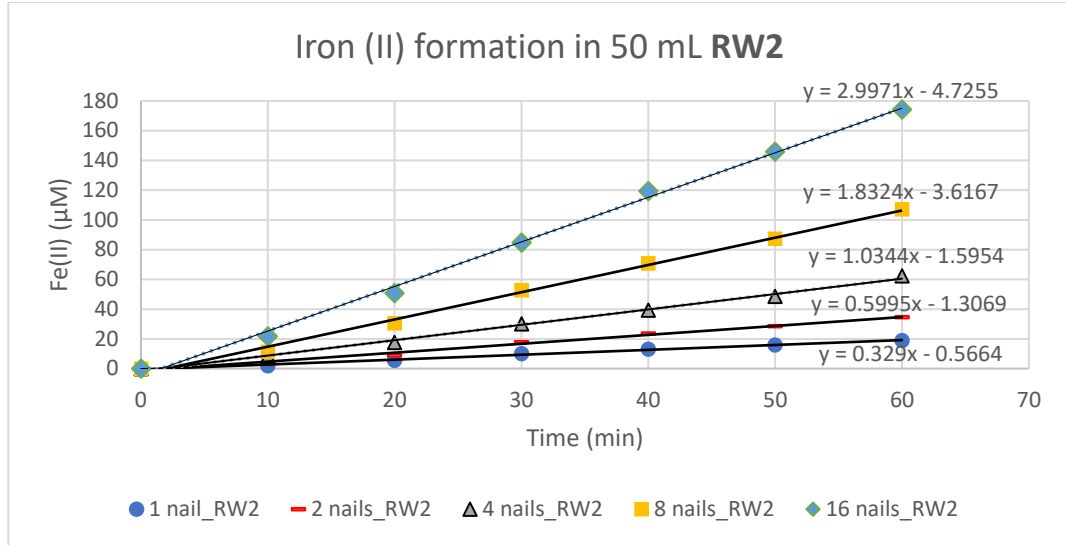


(b)

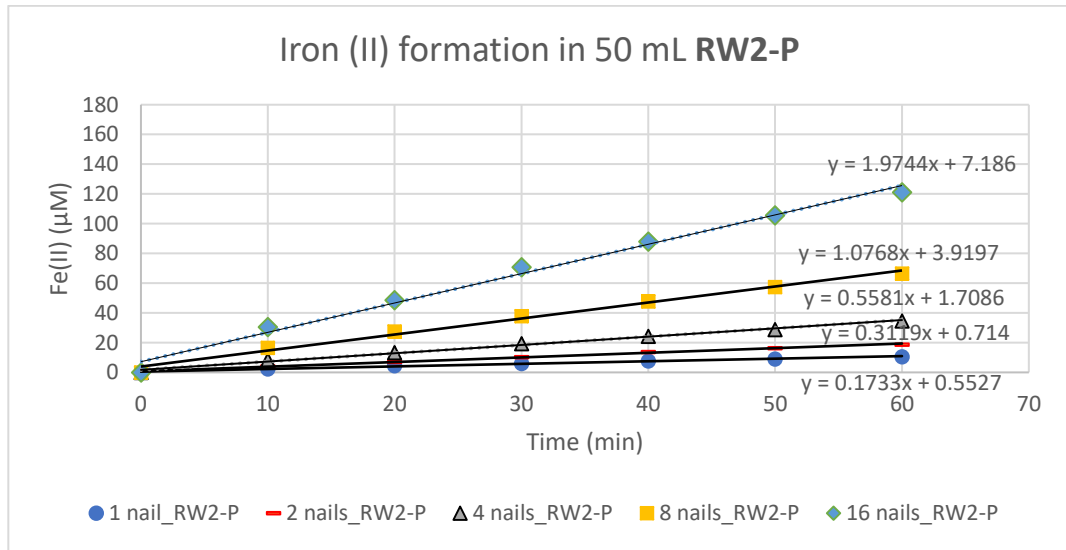




(c)



(d)



(e)

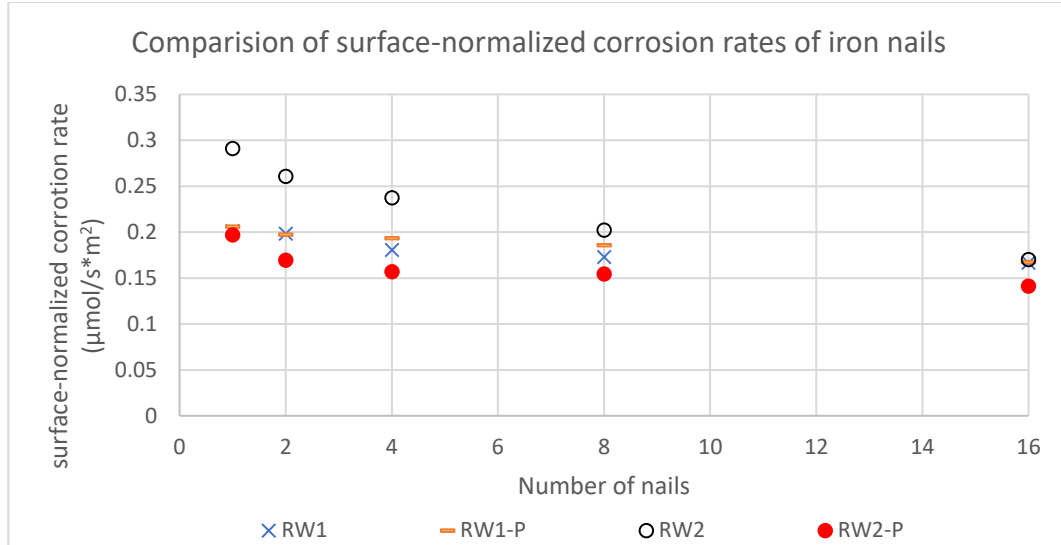


Figure 7. Fe(II) released from 1, 2, 4, 8, 16 nails in (a) **RW1**, average corrosion rate of  $0.96 \pm 0.15 \mu\text{mol/s}\cdot\text{m}^2$ , (b) **RW1-P**, average corrosion rate of  $1.00 \pm 0.17 \mu\text{mol/s}\cdot\text{m}^2$ , (c) **RW2**, average corrosion rate of  $1.22 \pm 0.30 \mu\text{mol/s}\cdot\text{m}^2$ , (d) **RW2-P**, average corrosion rate of  $0.89 \pm 0.15 \mu\text{mol/s}\cdot\text{m}^2$ ; and (e) comparison of surface-normalized corrosion rate of iron nails ( $\mu\text{mol/s}\cdot\text{m}^2$ ) in different water types.

#### 4.1.2 Column experiments

##### 4.1.2.1 Levels of iron corrosion products in supernatant water

The measured concentration of Fe(II) in supernatant water is an amount of the soluble oxidation product of iron after water contacting with iron nails which had not interacted with other water components. Dissolved Fe(II) formed complexes with BPY and were measured by a UV-Vis spectrophotometer. There was no strong correlation between concentrations of Fe(II) and total Fe in supernatant water (Figure 8). Total dissolved Fe levels in the low hardness water SW1-P and SW1 were higher than those of high hardness water SW2-P and SW2 while the reverse was true for the levels of Fe(II). This means that after the interaction between arsenic and iron corrosion

products on the nail surface, further oxidation of Fe(II) can occur in the standing water to form large compounds with arsenic leading to additional arsenic removal by filter straining.

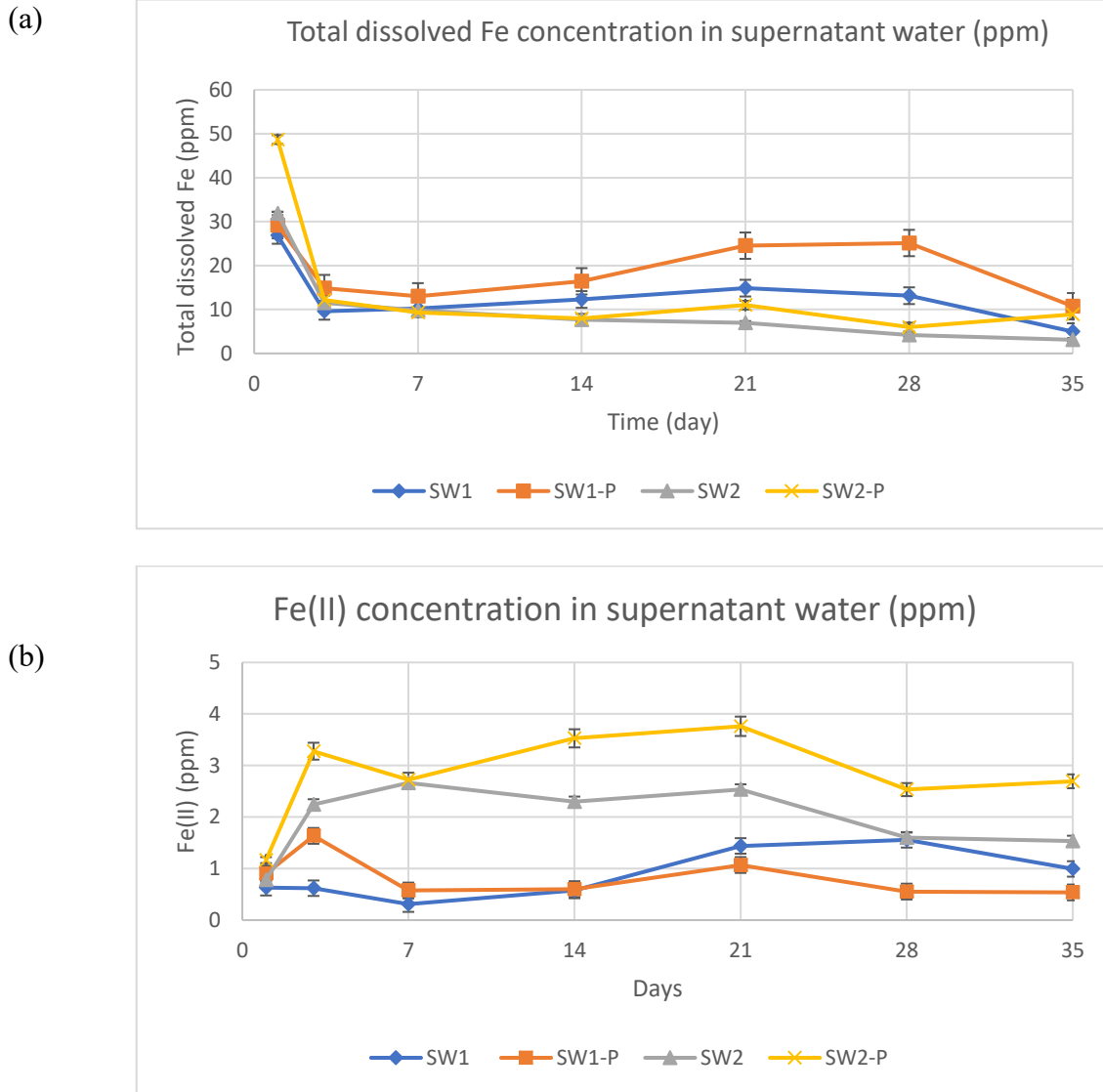
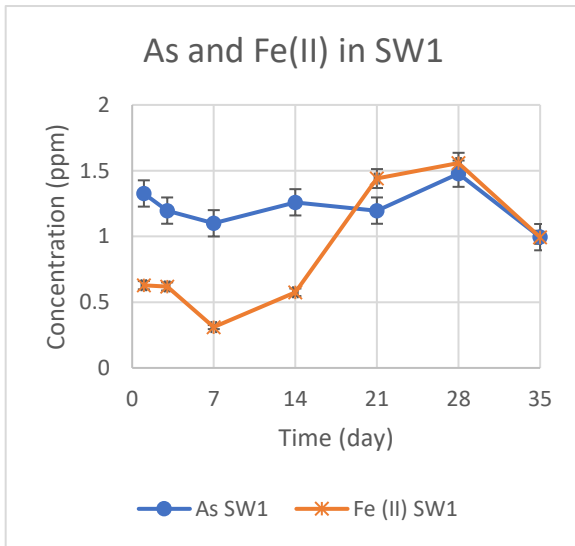


Figure 8. Concentrations of (a) Total dissolved Fe and (b) Fe (II) in supernatant water (SW) of corresponding groundwater GW1, GW1-P, GW2, and GW2-P; denoted as SW1, SW1-P, SW2, and SW2-P.

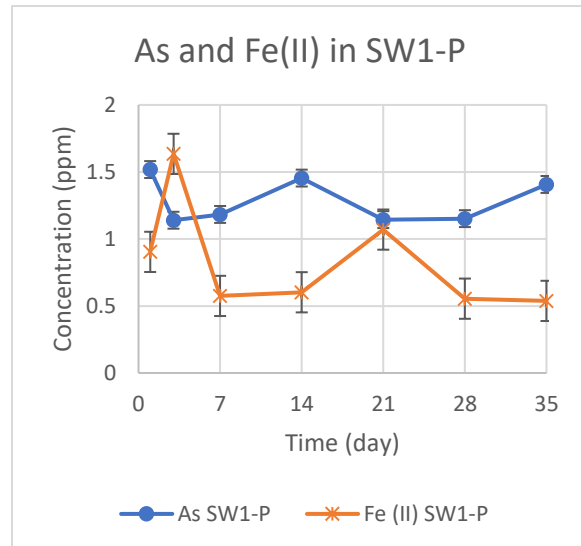
4.1.2.2 The relationship between iron corrosion products and arsenic after contacting with iron nails

i. Correlation of As and Fe(II) in supernatant water

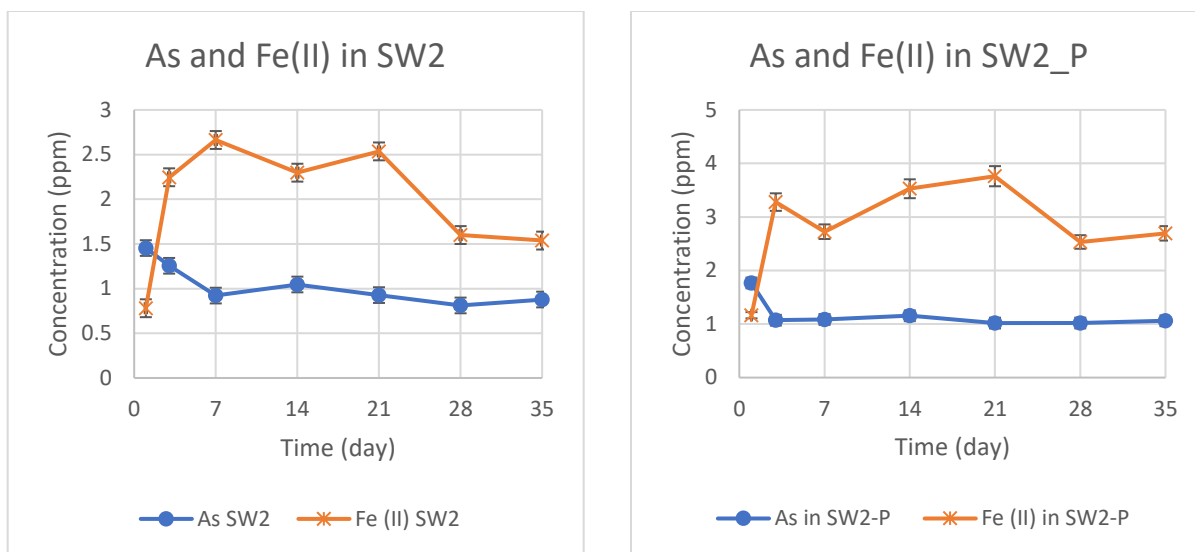
As concentration in supernatant water showed a negative correlation with Fe(II), except for that in SW1 (Figure 9). Fe(II) and As concentration patterns in high hardness water SW2 and SW2-P were very similar, Fe(II) levels were higher than that of As in both case, with the As concentrations of approximately 1 ppm even though the Fe(II) concentration in SW2-P was much higher than that of SW2. On the contrary, in low hardness water SW1 and SW1-P, Fe(II) concentrations were lower or approximately equal to those of As.



(a) SW1



(b) SW1-P



(c) SW2

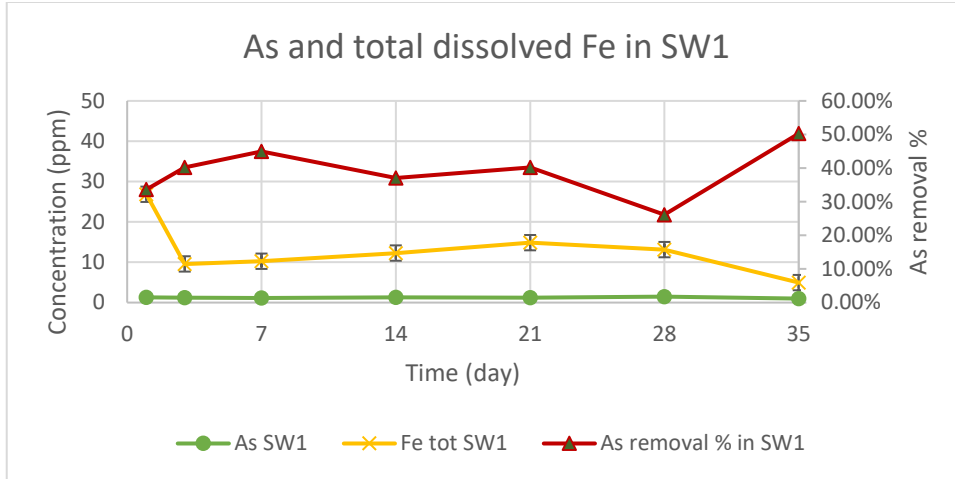
(d) SW2-P

Figure 9. Relationships between Fe(II) and As in (a) SW1, (b) SW1-P, (c) SW2 and (d) SW2-P

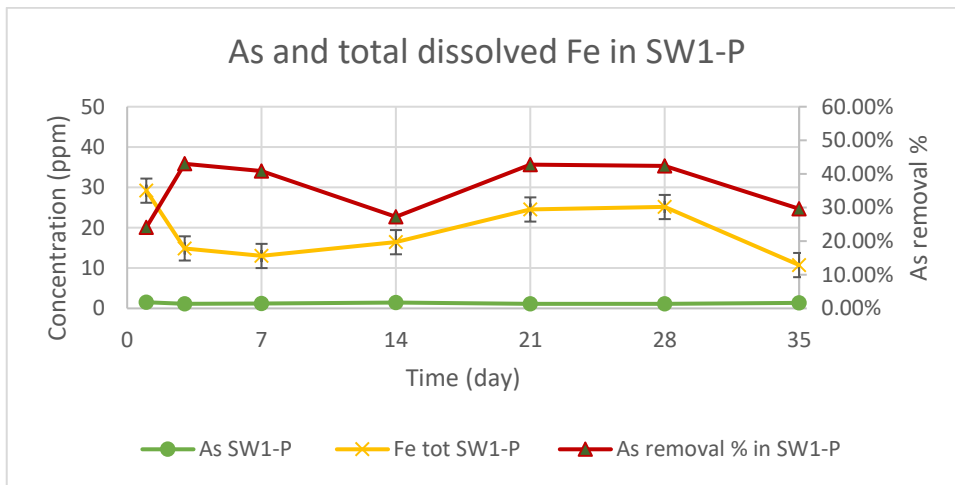
ii. As and total dissolved Fe levels in supernatant water

Except for the water type SW1-P, the percentage of arsenic removal after contacting with nails and total iron levels in all other supernatant water (SW1, SW2, and SW2-P) showed a remarkably negative relationship (Figure 10). It is worth noticing that the total Fe levels in supernatant water of filters treating high hardness groundwater (SW2 and SW2-P) were significantly lower than those of low hardness groundwater (SW1 and SW1-P). Of the groundwater with the same level of hardness, the water with the presence of phosphate had levels of total iron in supernatant water slightly higher than those without phosphate. In general, the descending order of total iron in supernatant water was SW1-P > SW1 > SW2-P > SW2. The descending order of average arsenic level presenting in supernatant water of each filter set over 35 days of treatment was also SW1-P > SW1 > SW2-P > SW2 (Figure 10 (e)). This illustrates a positive relationship between total dissolved iron and arsenic in supernatant water. The correlations between total iron and arsenic presenting in SW1, SW1-P, SW2, and SW2-P were 0.574, 0.077, 0.895 and 0.973, respectively.

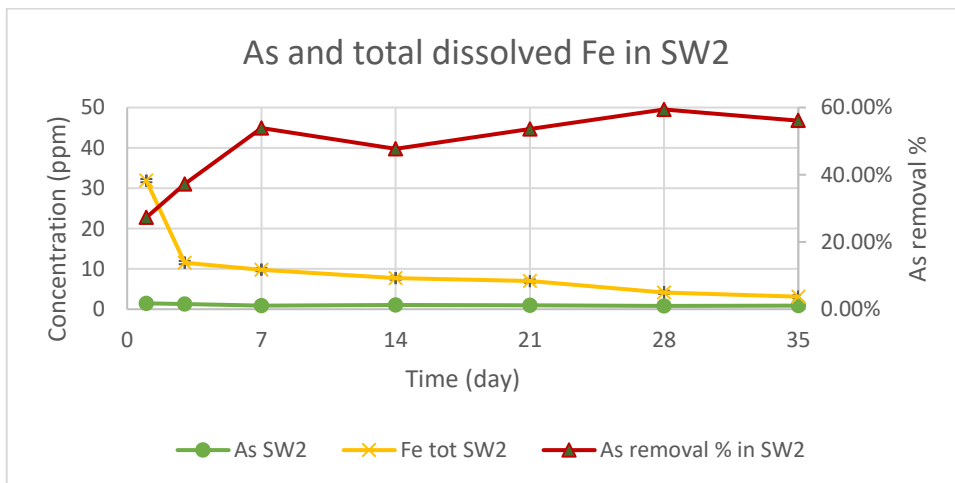
(a)



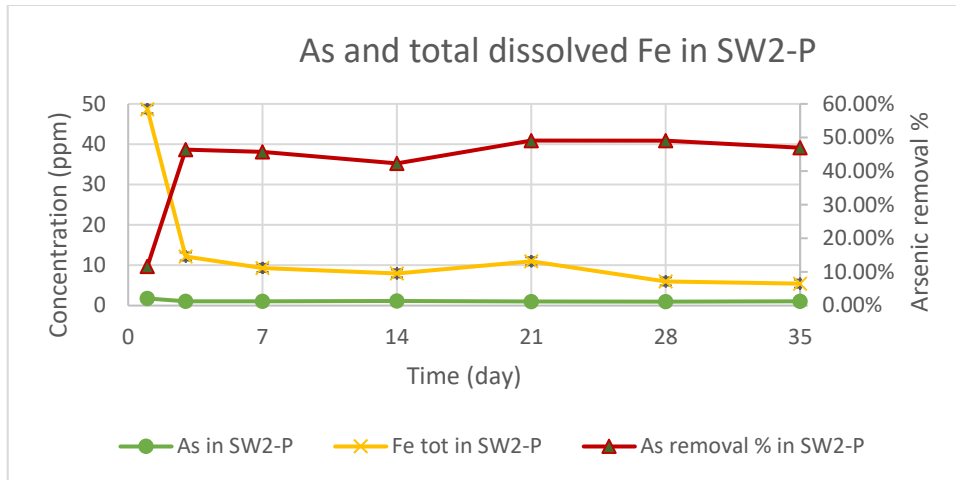
(b)



(c)



(d)



(e)

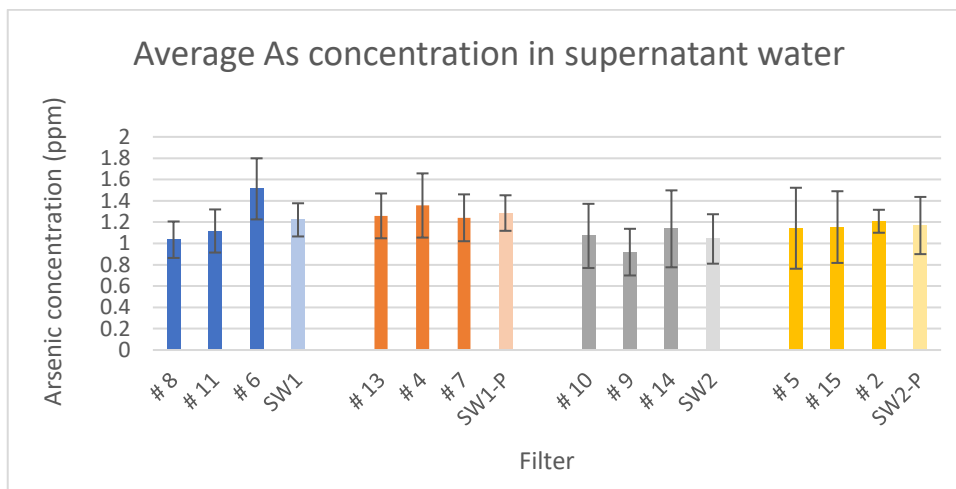


Figure 10. Relationships between As and total Fe levels in (a) SW1, (b) SW1-P, (c) SW2, and (d) SW2-P. Figure 10 (e) average As levels in supernatant water over time within each filter set.

#### 4.1.2.3 Arsenic removal

Figure 11 displays the log As removal by filter media of four types of artificial groundwater. The log As removal was a log of the ratio of As levels in supernatant water and effluent water. Generally, filters treating GW2 (high hardness, no phosphate) had the best As removal, followed by GW2-P (high hardness, high phosphate), GW1 (low hardness, no phosphate) and GW1-P (low hardness, high phosphate). Over 35 days of water treatment with an influent arsenic level of 2000  $\mu\text{g/L}$ , two filter sets treating groundwater with high hardness GW2 and GW2-P demonstrate

consistently outstanding performance with the lowest arsenic removal efficiency of 97.8% and 95.9%, respectively. Arsenic level in effluent water of GW2 and GW2-P were lower than 50 µg/L (Nepal’s drinking water standard) during the whole water treatment duration and up to day 21, respectively. In comparison, the performance of filters treating low hardness water GW1 and GW1-P were noticeably lower and varied dramatically with time, with the lowest arsenic removal efficiencies of 85.0% and 65.9%, respectively (Figure 12). Arsenic removal in these two filter sets started decreased on day 7 and kept reducing before a slight rise in the last few days of the treatment period which may be resulted from a high retention time of water as the filters started clogging.

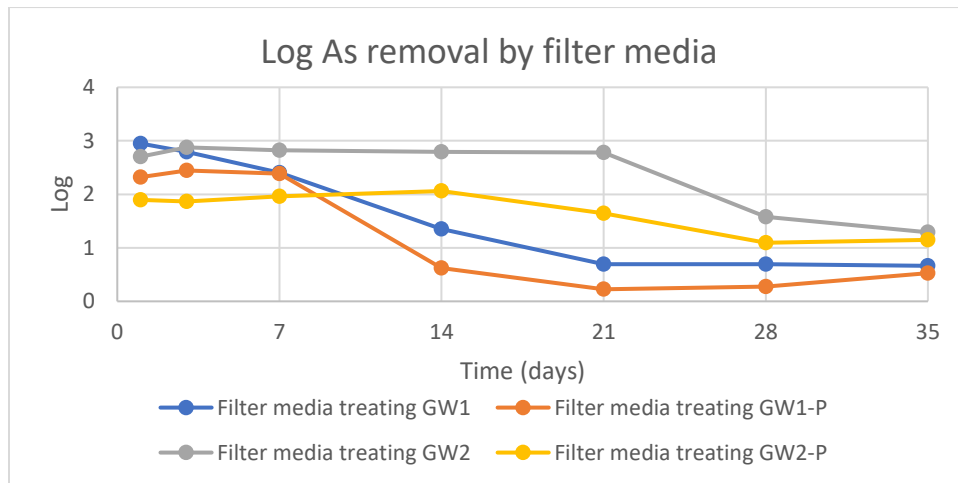
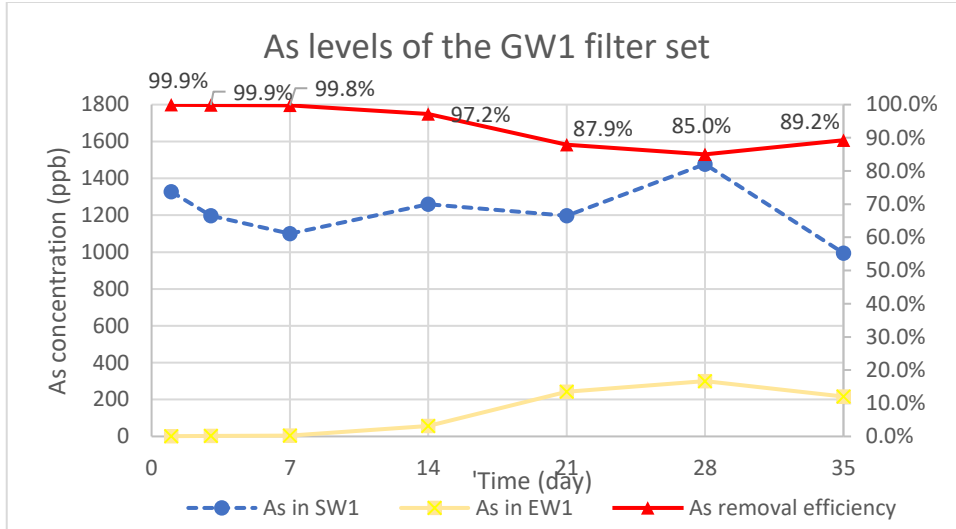


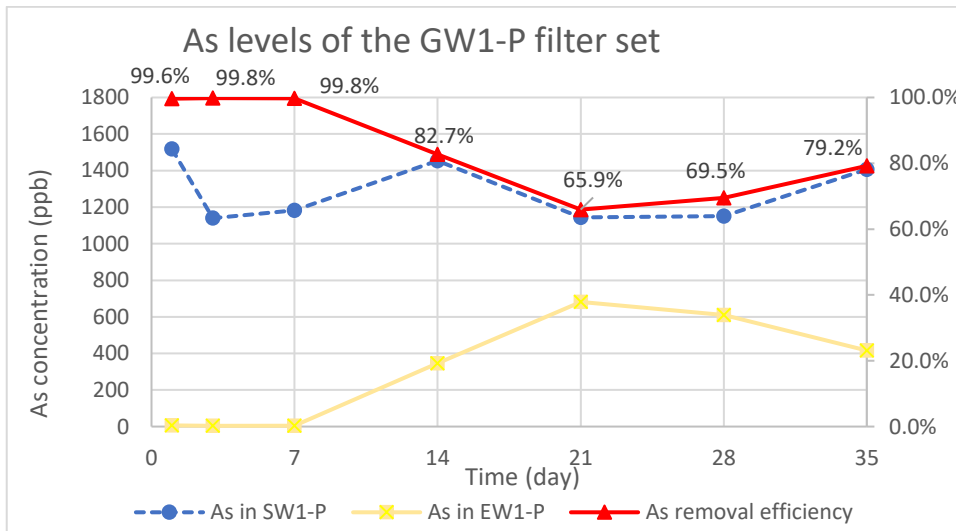
Figure 11. Log<sub>10</sub> As removal by filter media of all artificial groundwater types



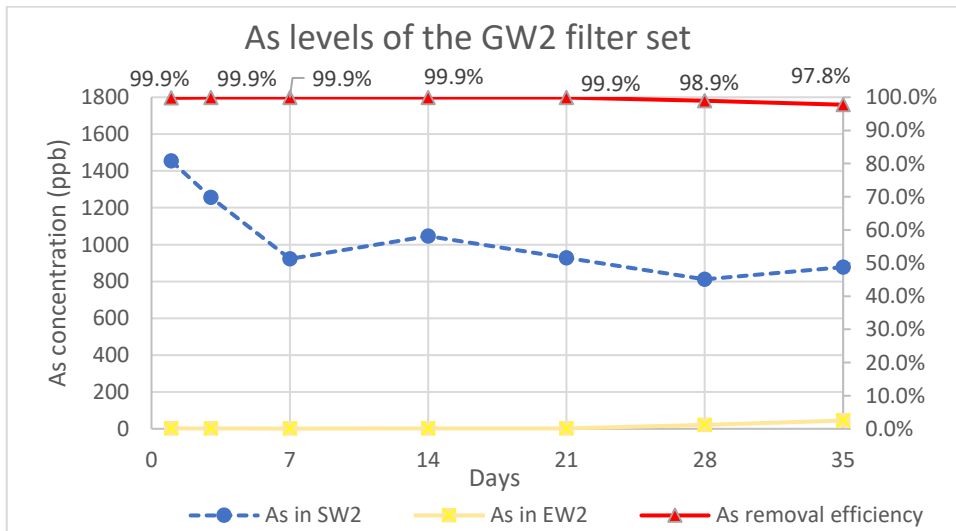
(a)



(b)



(c)



(d)

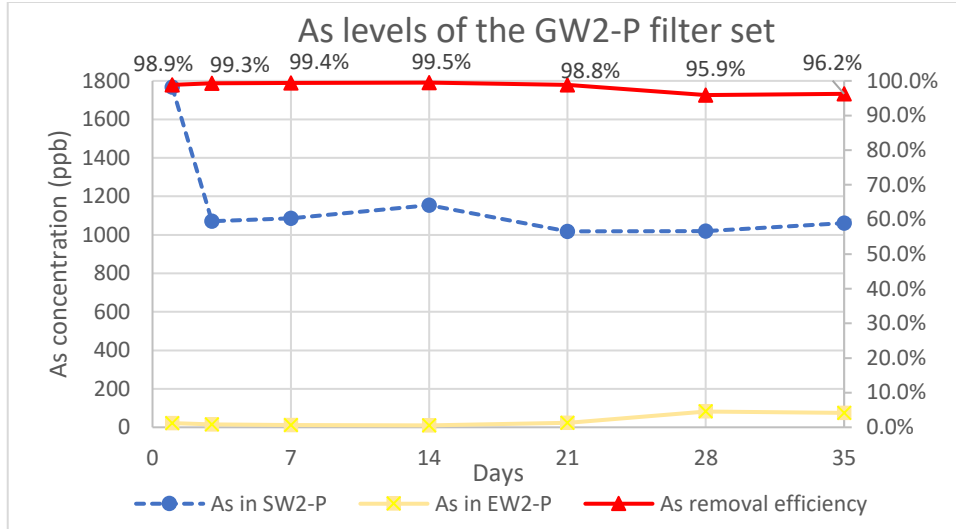


Figure 12. Arsenic levels and removal efficiency in filters using groundwater (a) GW1, (b) GW1-P, (c) GW2, and (d) GW2-P

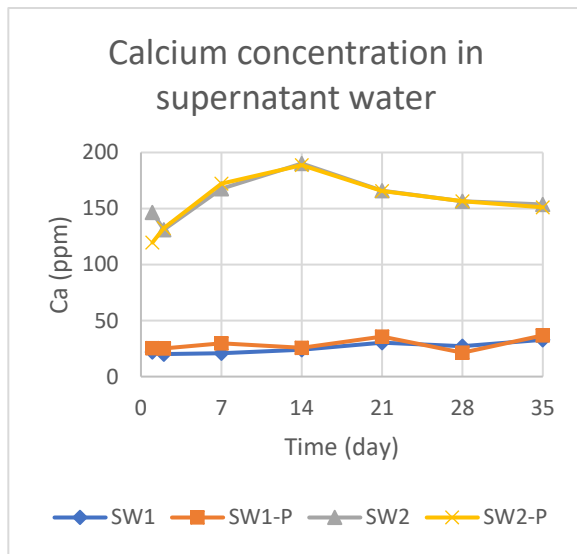
#### 4.1.2.4 Comparison of levels of contaminants in supernatant water and effluent water

The hardness levels in influent water of GW1 and GW1-P were  $55.05 \pm 0.94$  ppm Ca and  $7.85 \pm 0.58$  ppm Mg; that of GW2 and GW2-P were approximately 200 ppm Ca and 50 ppm Mg. In the supernatant and effluent water, there is a high similarity between the patterns of Ca and Mg concentrations of the pair of groundwater having the same level of hardness while phosphate levels of all water types varied tremendously (Figure 13). Of all pairs of water compositions in the supernatant water and effluent water in Figure 13, only the y-axis in (e) differs from (f).

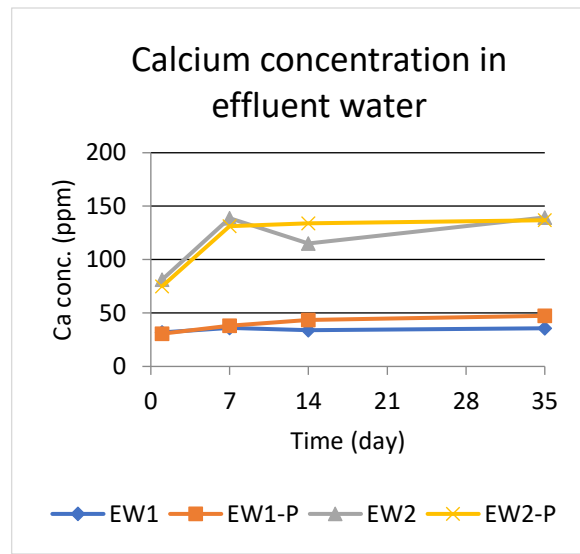
Taking hardness into account, Ca in all supernatant waters reduced to a slight extent compared to the initial levels. At a high concentration of hardness, the lab-scale filters could not remove more than 50% of influent Ca after treatment. Mg levels in the supernatant of GW2 and GW2-P in the first 7 days, and of GW1 and GW1-P in the first 21 days were higher than those in influent water, means that it could be released from iron nails as a by-product of iron corrosion. Surprisingly, at some testing points of filters treating low hardness water, levels of Ca and Mg in the effluent were

higher than in the supernatant water. This means that desorption of contaminants in filter media might occur.

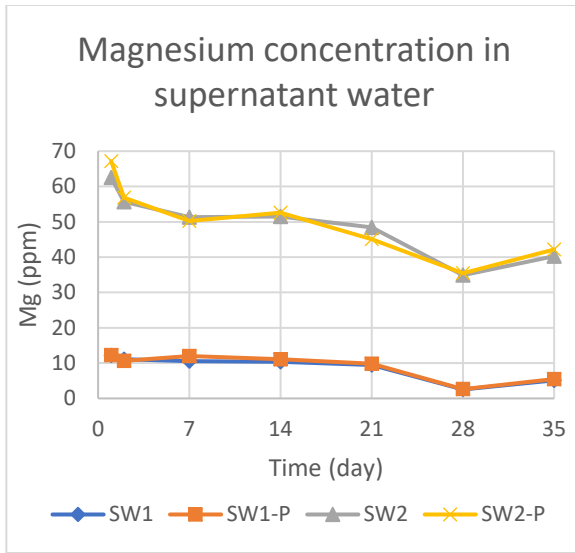
When it comes to phosphate, although no phosphate was added to GW1 and presented in SW1, there was a noticeable amount of phosphate in EW1 which may be evidence of phosphate leaching from the sand. This phenomenon was applicable to phosphate in GW2 to a smaller extent. More than 50% of the initial phosphate amount in GW2-P (2 ppm) present in SW2-P was effectively removed by filter media to approximately a negligible concentration of phosphate in EW2-P. This was not the case for phosphate in GW1-P which may be because of a lack of adsorption sites in the low hardness condition, resulting in a high dissolved phosphate level in EW1-P.



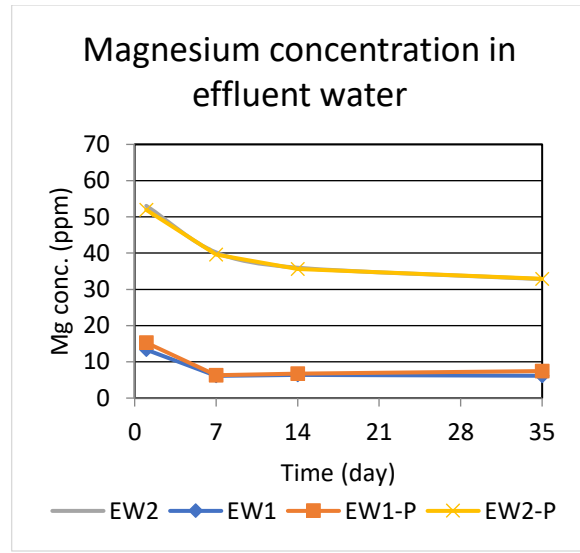
(a) Ca in SW



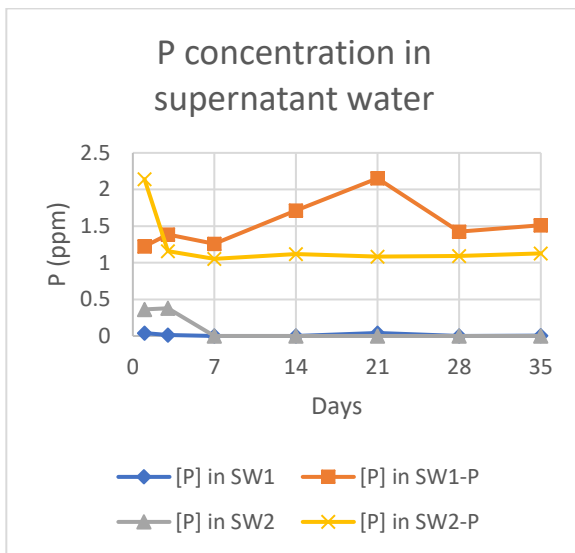
(b) Ca in EW



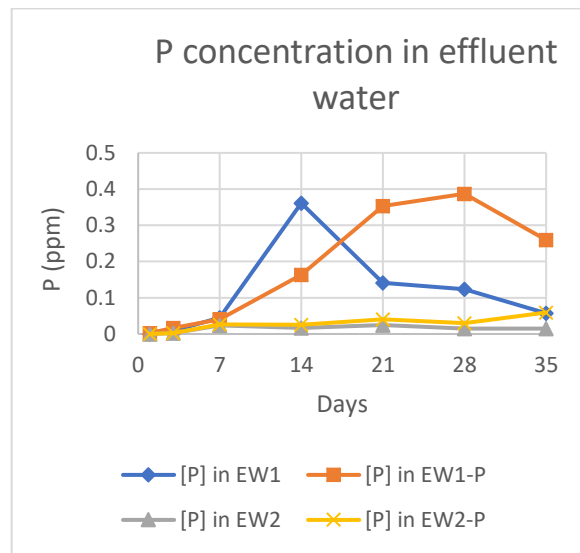
(c) Mg in SW



(d) Mg in EW



(e) P in SW



(f) P in EW

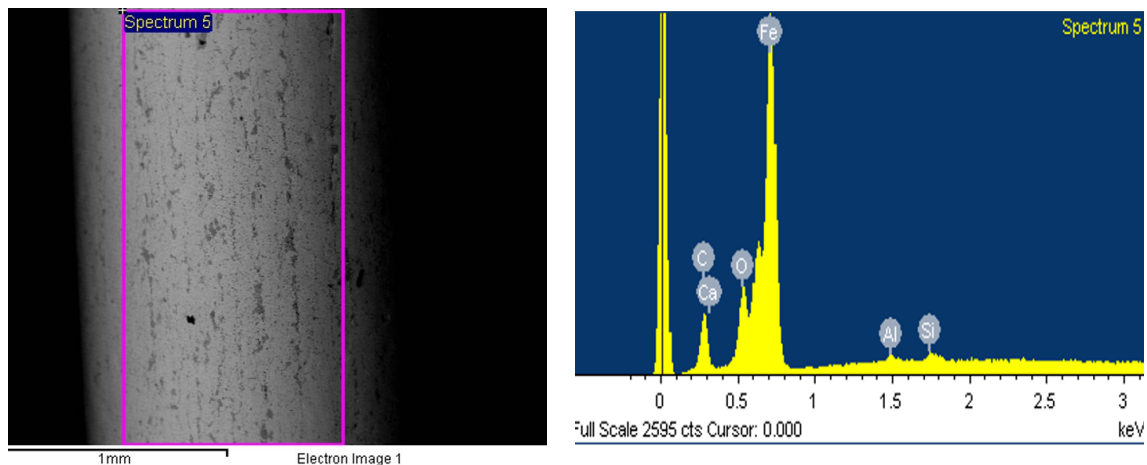
Figure 13. Levels of contaminants in water samples during the water treatment period: calcium in (a) supernatant and (b) effluent water; magnesium in (c) supernatant and (d) effluent water; and phosphate in (e) supernatant and (f) effluent water

#### 4.1.2.5 Characterization of iron corrosion products

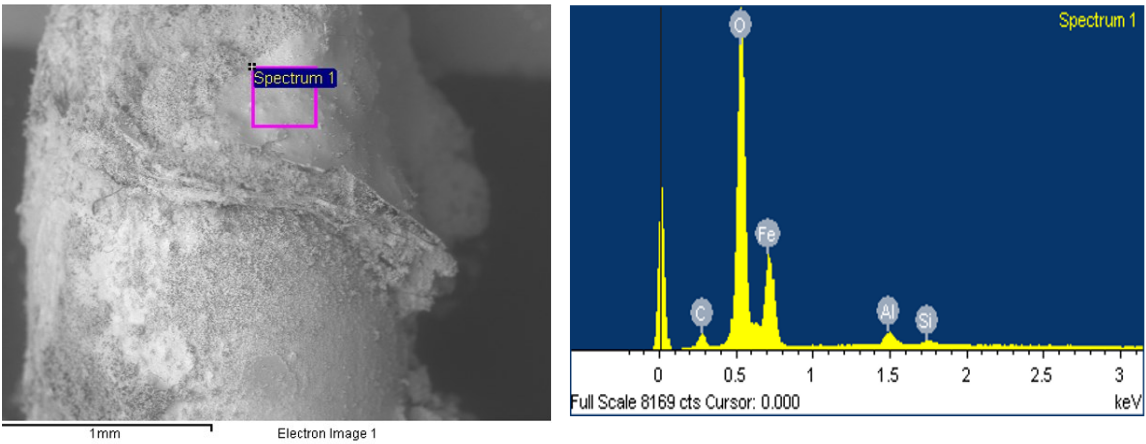
- i. Investigation of iron nails surface by SEM/EDS and XRD

SEM/EDS analysis was applied to characterize the morphology and compositions of the surface of a clean nail before treatment and a representative nail from each filter of two controlling filters and four filter sets running artificial groundwater (Figure 14). The changes in the compositions of elements and structures of the nails' surface were clearly displayed. The clean nail had a homogeneous surface with more than 60 atomic% of iron and trace amounts of Si, Al and Ca. The nails after treatment had heterogeneous surfaces due to the accumulation of contaminants. SEM/EDS results of nails in RW1 and RW2-P demonstrated a deposit of corresponding water components on the nail surfaces. Cl in samples RW2-P, GW2, GW2-P was the by-product of the preparation of hardness-rich artificial groundwater from  $\text{CaCl}_2$  and  $\text{MgCl}_2$ . Although the equivalent amount of Cl was added to GW1 and GW1-P to produce the same background of water, Cl peaks were not shown on SEM/EDS results of the nail samples from those water types.

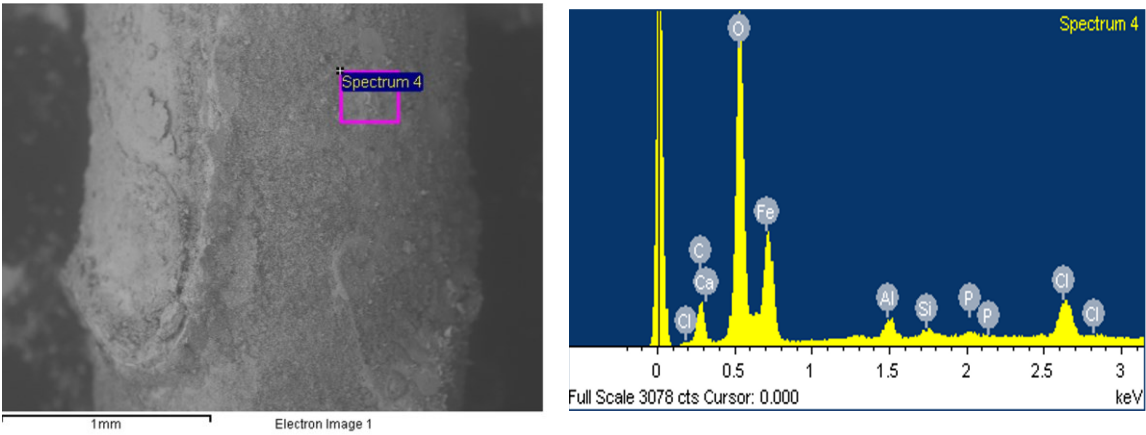
As peaks were shown on all iron nail samples from filters treating GW1, GW1-P, GW2, and GW2-P which means As adsorbed onto iron corrosion products or attached to the absorption site of hardness compounds depositing on the nail surface.



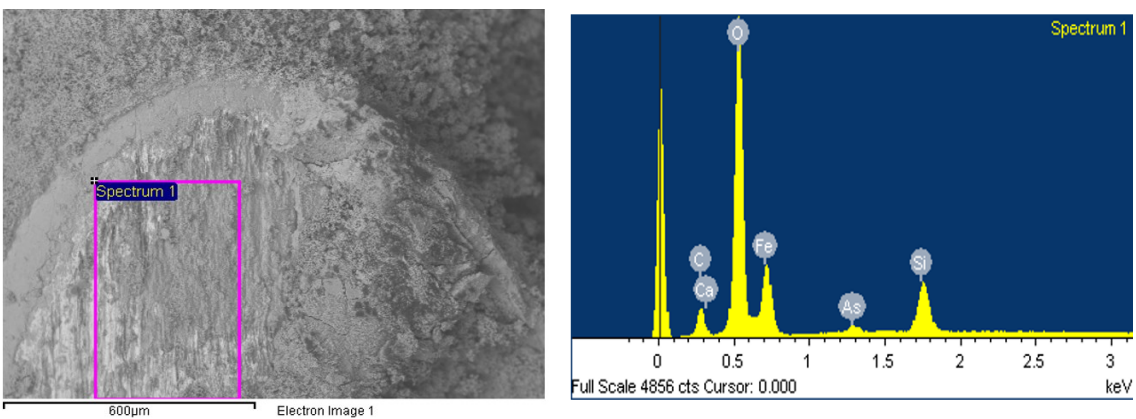
(a) SEM/EDS images of a clean nail



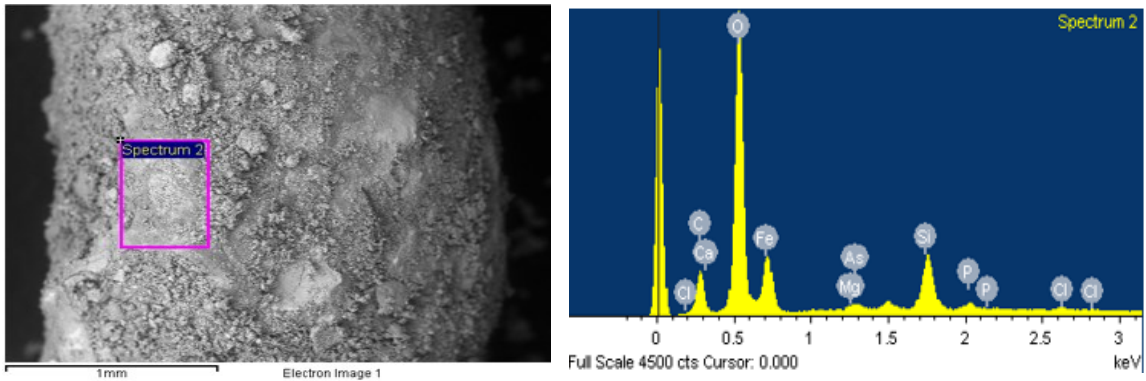
(b) SEM/EDS images of a nail from a controlling filter running RW1



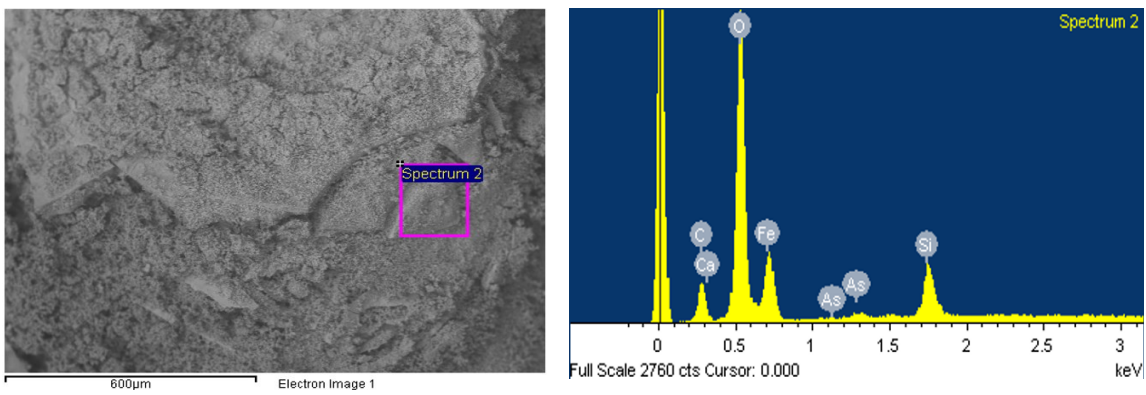
(c) SEM/EDS images of a nail from a controlling filter running RW2-P



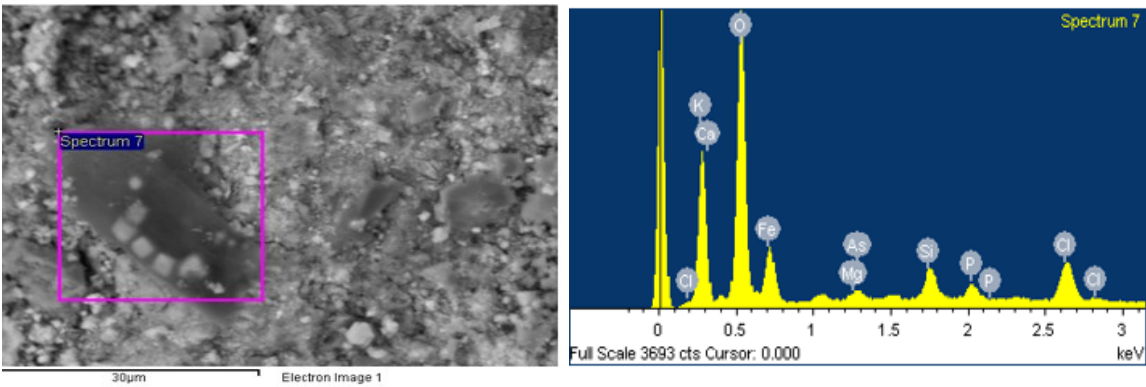
(d) SEM/EDS images of a nail from a testing filter running GW1



(e) SEM/EDS images of a nail from a testing filter running GW1-P



(f) SEM/EDS images of a nail from a testing filter running GW2



(g) SEM/EDS images of a nail from a testing filter running GW2-P

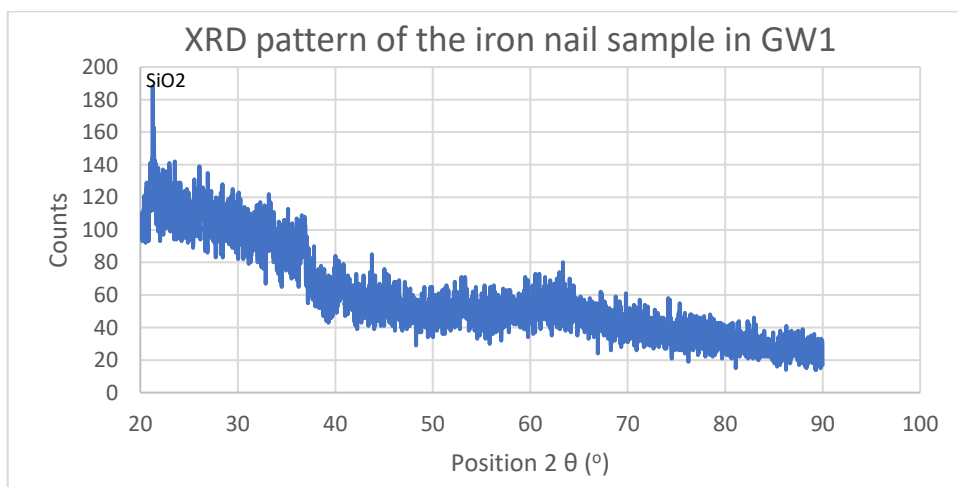
Figure 14. SEM/EDS images (left and right) of the surface of iron nail samples (one selected representative area for each nail): (a) clean nail, (b) nail in RW1, (c) nail in RW2-P, (d) nail in GW1, (e) nail in GW1-P, (f) nail in GW2, and (g) nail in GW2-P

XRD analysis was done for homogeneous dried powder samples of iron nails in all water types as well as suspended solids in the supernatant water of these waters. However, XRD results of all suspended solids had no peaks which can be explained by that either the sample amount was not enough, or the structures of those samples were not crystalline. Therefore, TEM analysis was conducted to address the morphology and composition of the colloids.

XRD patterns of samples from GW1 and GW1-P demonstrated only silicate  $\text{SiO}_2$  peaks while the identity peaks of  $\text{Fe}_2\text{O}_3$  and  $\text{CaCO}_3$  were clearly reported in the results of samples from hardness-rich water GW2 and GW2-P. This implies that in the presence of Ca, iron corrosion performed remarkably which is in harmony with the XRD result of the iron sample from RW2-P in Figure 15 (e).

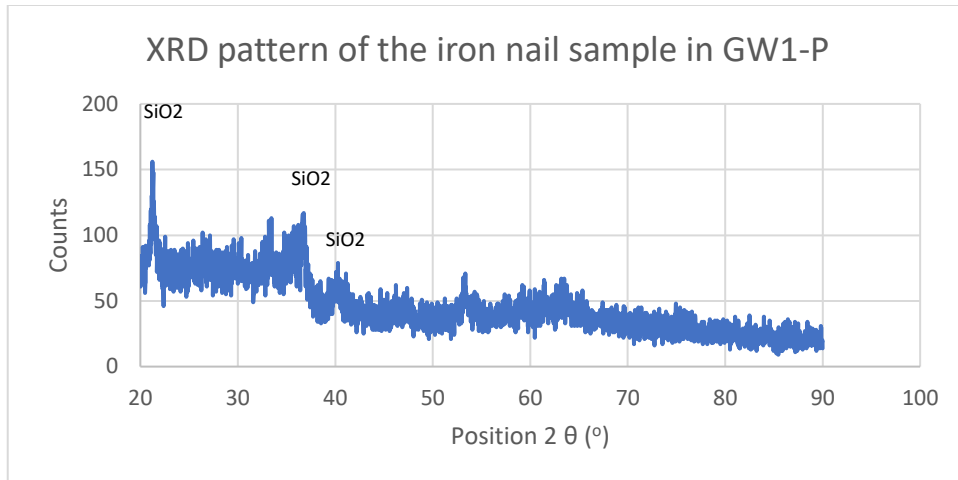
Interestingly, EDS and XRD results of iron nail samples from GW2 and GW2-P both indicate a pronounced amount of Ca depositing on the nail of GW2-P compared to GW2. This suggests a correlation between the presence of phosphate and the accumulation of Ca on the iron surface which may have a direct impact on arsenic removal.

(a)

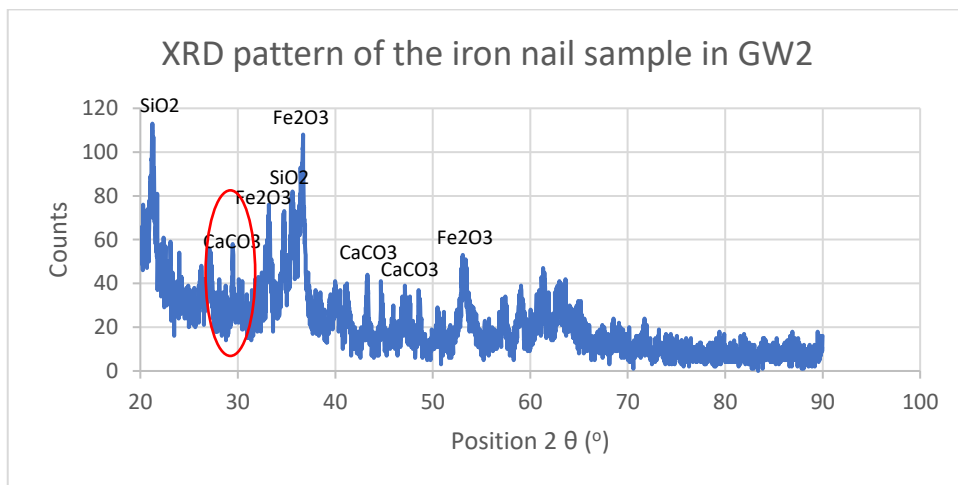




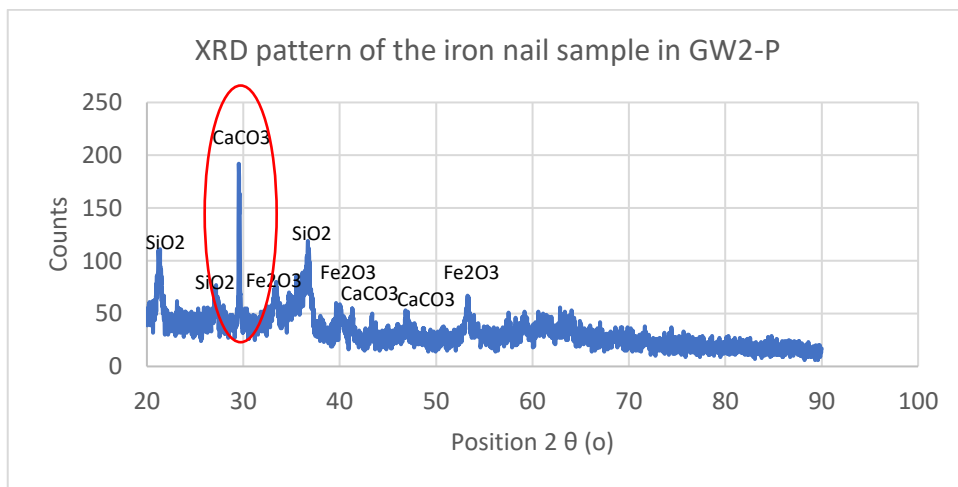
(b)



(c)



(d)



(e)

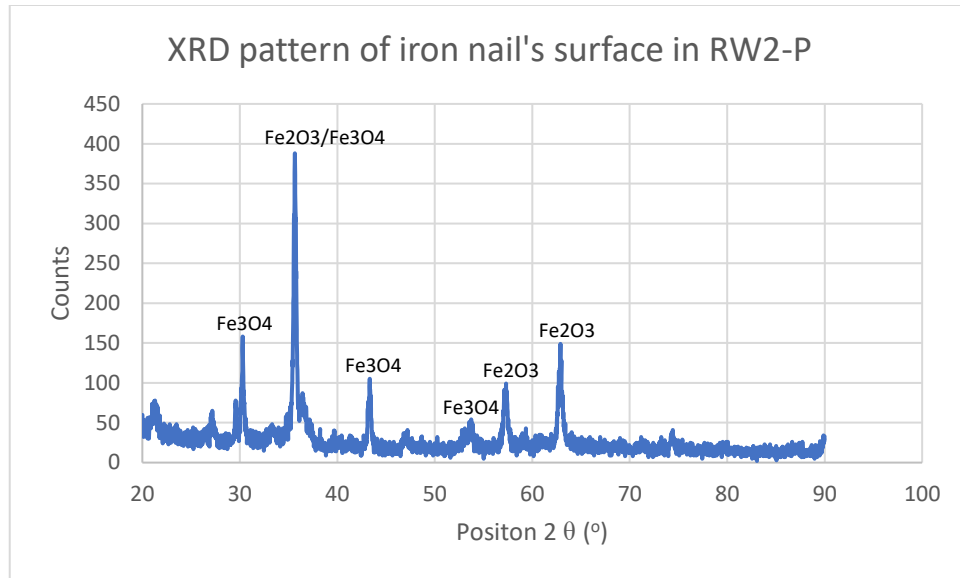


Figure 15. XRD patterns of an iron nail sample from a filter treating (a) GW1, (b) GW1-P, (c) GW2, (d) GW2-P, and (e) RW2-P in 35 days

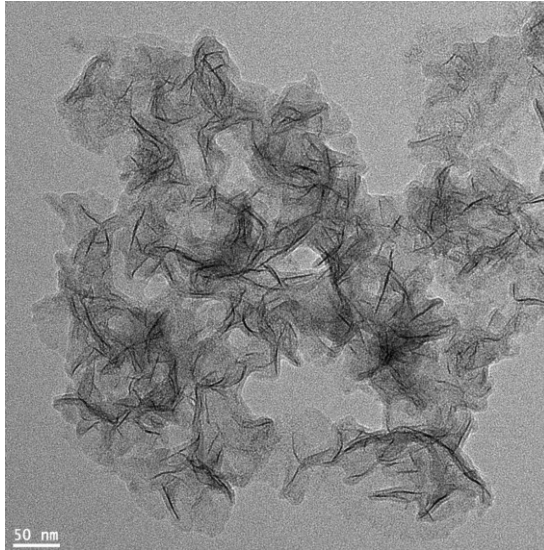
ii. Examination of suspended solids in supernatant water by TEM

TEM analysis was conducted for two samples collected on day 35 of suspended solids in supernatant water of GW1 and GW2-P (Figure 16). The TEM and Selected Area Electron Diffraction (SAED) results exhibited that both samples were amorphous, agreeing with XRD results in part 4.1.2.5 i. Therefore, no TEM analysis was needed for suspended solids in SW1-P and SW2.

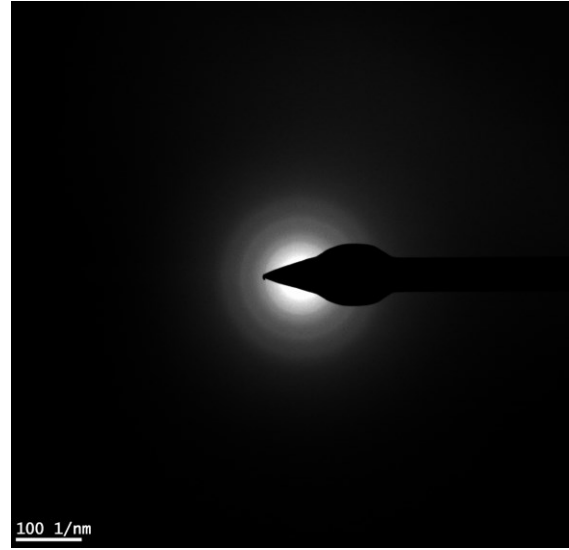
Compositions of the colloids in SW1 and SW2-P were analyzed by the Energy-dispersive X-ray spectroscopy (EDX). The gel-like materials were shown to contain Fe-Ca-Si-As or Fe-Ca-Mg-Si-P-As which were expected to be amorphous ferric arsenate (Fe(III)-As(V)) complexes formed through transformation of the arsenate ions adsorption on the surface of iron(hydr)oxides (Yang et al., 2015; Mak et al., 2008). TEM images demonstrated that the size of the amorphous colloids was very small (less than 1µm). If no coagulation occurred, these tiny colloids carrying arsenic

could easily pass through the filter media carrying all contaminants. This may be one of the reasons for higher arsenic levels in the effluent water of filters running low hardness groundwater.

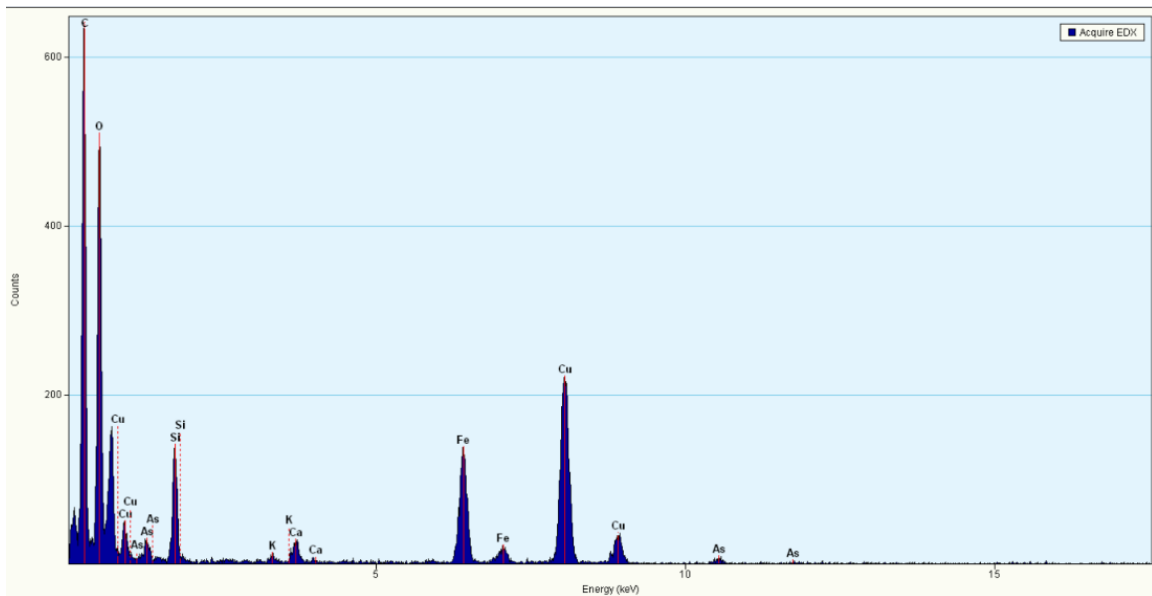
(a)



TEM of suspended solids in SW1

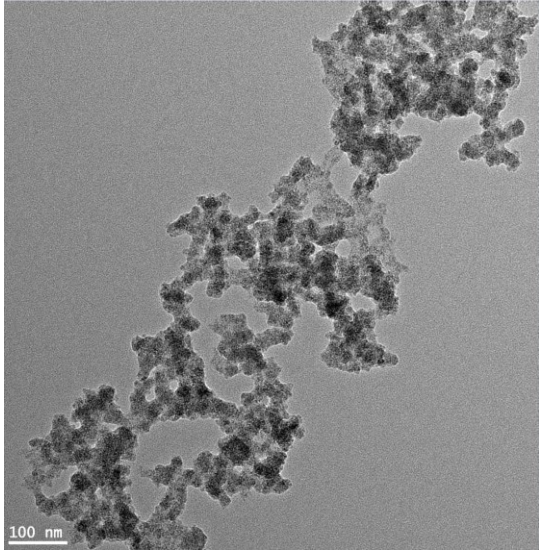


SAED of suspended solids in SW1

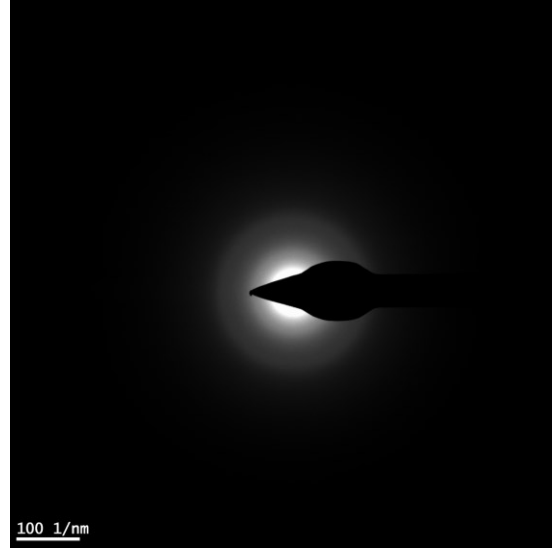


EDX of suspended solids in SW1

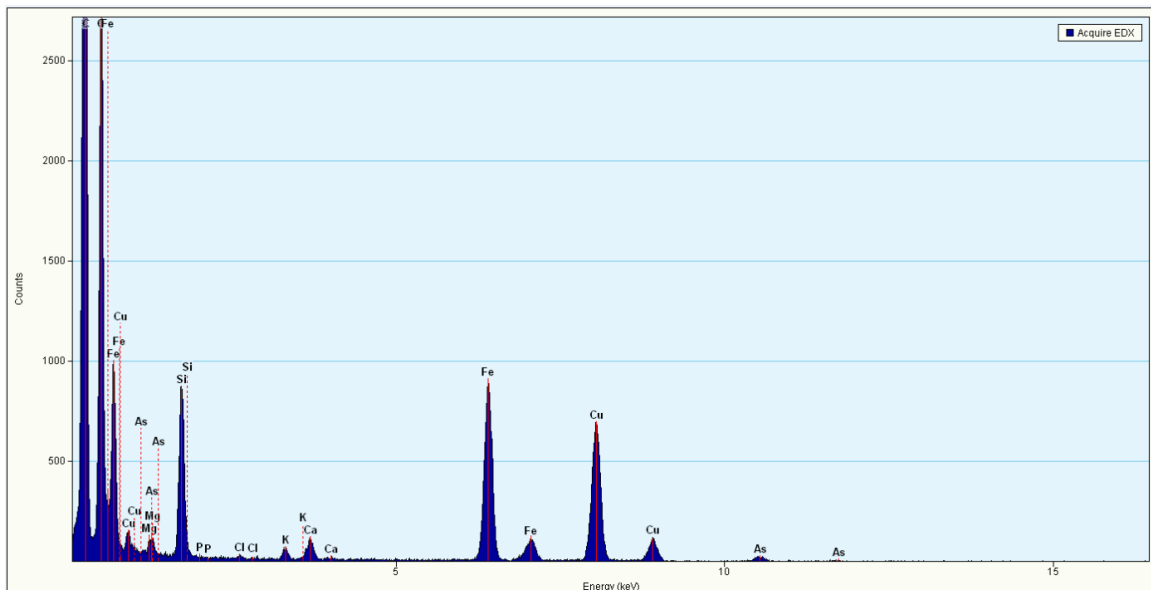
(b)



TEM of suspended solids in SW2-P



SAED of suspended solids in SW2-P



EDX of suspended solids in SW2-P

Figure 16. Characterization of compounds in suspended solids in supernatant water by TEM, SAED and EDX for water samples in day 35 of water treatment process: (a) suspended solids in GW1 and (b) suspended solids in GW2-P

## 4.2 Discussion of lab-experimental results

### 4.2.1 Effects of hardness and phosphate of water chemistry on iron corrosion

The artificial groundwater in this study was indicated as nonaggressive with a positive Langelier Saturation Index. This means  $\text{CaCO}_3$  precipitation may occur (Donner, 2015). Figures 14 and 15 (c) and (d) supported that hypothesis by demonstrating the deposit of Ca compounds on the iron nail surface.

Data collected from quantitative measurements (UV-Vis and ICP-MS) and qualitative analysis (XRD) of iron corrosion products from iron nails immersed in groundwater (GW) and water without the presence of Si and As (RW) were very different from each other. This shows that the corrosion of iron was highly dependent on the water conditions. Solution chemistry, including pH, redox state, and chemical compositions, also had sophisticated impacts on precipitate formation, adsorption and desorption of contaminants (Yadav et al., 2014; Diwakar et al., 2014).

There were obvious discrepancies between levels of total Fe and Fe(II) presenting in supernatant water of a pair of groundwater having the same level of hardness (Figure 14). To be more specific, after 7 days of water treatment, total Fe levels in four types of supernatant water differed considerably. Despite having the lowest total iron concentrations, supernatant water of filter treating GW2-P and GW2 obtained the highest concentrations of Fe(II). This not only showed the promoting role hardness played on iron corrosion by producing more Fe(II) but also indicated the ability of forming precipitates and coagulation bound to iron nail surface and removed approximately 50% influent As before further removal by filter media. Mak et al. (2008) reported that in the presence of bicarbonate,  $\text{CaCO}_3$  could be formed and could act as a nucleation seed for the growth of mineral crystals, such as iron (hydr)oxides. Large particles of iron corrosion products could be formed by the growth of iron(hydr)oxides outward from the seed and big enough to be

removed by a 0.45  $\mu\text{m}$  filter. Therefore, total dissolved iron as a combination of tiny iron rust particles (passing through the 0.45  $\mu\text{m}$  filter) and dissolved iron in SW2-P and SW2 reduced dramatically. In contrast, because of the low level of hardness in GW1 and GW1-P,  $\text{CaCO}_3$  formation in these filters was insufficient for coagulation or formation of big particle. As a result, iron (hydr)oxide colloids could easily pass through the 0.45  $\mu\text{m}$  filter, contributing to the level of total dissolved iron.

#### 4.2.2 Impacts of water chemistry on arsenic removal

Farrell and Chaudhary (2013) indicated that arsenic adsorption onto iron and aluminum oxides has a two-phase kinetics of fast and slowly adsorbing fractions, in which the latter is the rate-controlling step and is a possible consequence of “heterogeneous adsorption sites (Zhang & Stanforth, 2005), the formation of surface precipitates (Hongshao & Stanforth, 2001), and conversion of monodentate to bidentate complexes (Grossl et al., 1997)”. It is comprehensively presented in the literature review that As(V) is less mobile than As(III) and more easily attached to the sorption site on iron surfaces than As(III). Therefore, the colloidal materials collected in supernatant water as products of iron corrosion and interaction with As were expected to be Fe(III)-As(V) complex. The SEM/EDS and XRD results in this study clearly demonstrated the surfaces of iron nails in four filter sets were heterogeneous with co-existing iron phases consisting of Fe(III)-As(V) amorphous complexes, which agreed with previous studies on arsenic removal by iron corrosion products (Manning et al., 2002; Wang et al., 2008; Yang et al., 2015). The heterogeneity of iron nail surface may explain the reason that both cations (calcium, magnesium) and anions (such as negatively-charged arsenate compounds, phosphate, silicate) can be bound to it.

Phosphate did show its high affinity for adsorption sites as phosphate peaks occurred in all EDX and EDS patterns of iron nails in the filter using phosphate-containing water in Figure 14 and Figure 16). The preventing role of phosphate on arsenic removal illustrated in Figure 12 as arsenic removal performance of filters treating high phosphate water was lower than those treating no phosphate water under the same hardness conditions. But as reported in all quantitative analysis results, the enhancing role of hardness had outweighed the inhibiting role of hardness on arsenic removal. Hardness-rich groundwater may promote arsenic removal efficiency by elevating the oxidation of As(III) to As(V) (Katsoyiannis et al., 2008) and supporting As adsorption onto the iron surface (Hug et al., 2008).

The high levels of Fe(II) in SW2 and SW2-P (Figure 9) were an enhancing factor for arsenic removal because more As(V) was formed by a co-oxidation of Fe(II) with As(III) leading to a better adsorption of As to Fe(III)-colloids (Hug and Leupin, 2003). This was proven by a high removal rate of filters treating GW2 and GW2-P (Figure 10). Katsoyiannis et al. (2008) claimed that As(III) oxidation was significantly prevented in the presence of BPY at all tested pH values. This implied that Fe(II) together with other oxidizing substances in water played an exceptional role on As(III) oxidation that Fe(0) and O<sub>2</sub> alone could not compensate. Corrosion rates of iron nails slowed down over time as a result of a formation of a passive corrosion layer.

Adsorption of As into Fe(III)-precipitates may also be facilitated by the electrostatic interactions of Ca by enhancing Fe(III)-colloid coagulation and providing additional adsorption sites, or forming Ca-P complexes which reduce the competitive ability of phosphate for sorption sites on iron corrosion surface (Meng et al., 2001; Tanboonchuy et al., 2012). This explains the elevated amount of CaCO<sub>3</sub> deposited on the surface of iron nail sample in GW2-P shown in Figure 15 (d) and Figure 14 (g), as well as the noticeably higher arsenic removal efficiency of filters treating

hardness-rich groundwater GW2 and GW2-P compared to those of filters treating low hardness groundwater GW1 and GW1-P. Moreover, Tanboonchuy et al. (2012) stated that Ca cations could promote As removal by increasing the positive charge of the iron surface and attracting more As anions but this hypothesis was not proven by any tests in this study.

TEM images demonstrated that the size of amorphous colloids containing Fe-As-Ca-Si or Fe-As-Ca-Si-P in the supernatant water was very small which is in line with an earlier study by Voegelin et al. (2010) that showed in the presence of bicarbonate, small colloidally stable Fe(III)-As precipitates formed, which can pass through the 0.2  $\mu\text{m}$  filters. The effective diameter of the filter media in this study was 0.18 mm, which means coagulation of the Fe(III)-As colloids needed to occur for efficient removal by the filters. In hardness-poor groundwater, coagulation of Fe(III) colloids was less likely to be formed due to the absence of Ca, which results in a low removal performance of the filters running these groundwater. Arsenic released from ferric-arsenate colloids to water was another of the possible reasons for the decrease in arsenic removal efficiency (more remarkable in the GW1 and GW1-P filter sets than the others) illustrated in Figure 10. The desorption of As(V)-Fe(III) precipitates may due to 1) a transformation of Fe(III)-arsenate to hydrous ferric oxides or ferrihydrite over periods of days to weeks (Voegelin et al., 2010); 2) an increase in pH values leading to a negative net surface charge of iron rust layer which may repel negatively charged As compounds (Farrell & Chaudhary, 2013), an occurrence of redox reactions (Mueller, 2016), and the competition with P and Si. Further study needs to be done to address the exact reason for As desorption.

#### 4.3 Contribution to the field research in Nepal in part B

The results and conclusions mentioned from this chapter, together with the knowledge obtained from the literature review, were applied to plan for the field research in Nawalparasi district, Nepal



by determining important parameters to test in each water type and number of samples to be collected.

## B. Field research in Nawalparasi district, Nepal

### 4.4 Observations from the field research

All studied KAFs had a stable location in the house that was away from direct exposure of weather, with some KAFs were better placed than others. The main body parts of KAFs were sustainable in all households. No leakage or severe damage was detected. Figure 17 shows a representative KAF used in the Nawalparasi district.



Figure 17. A representative KAF version 10 in the studied area. Photo taken by the author (2018)

Local organizations and retailers did some small modifications on the exterior of KAF which did not affect technical parameters and filter performance but increased user's interest and pleasure.

For example, a decorated filter with tiles for a more appealing appearance, applying a larger and more durable plastic tube over the small default outlet tube to protect it from damage, and changing the type of tap for the outlet of storage containers to one that was more sustainable and more convenient for KAF users (Figure 17).

Regarding technical acceptance, 100% of the interviewed KAF users were satisfied with the filter design (i.e., the height of the filter, the amount of water load per time, and filtration rate). They had no difficulty in feeding and cleaning the filter. All studied households used their KAFs every day for water treatment.

There was a significant improvement of water quality after treatment by appearance, taste, and smell. All interviewed KAF users preferred filtered water over raw water taken from tube-wells. Source water was grey, cloudy and tasted bad while filtered water was clear, transparent and tasted fresh. The households used filtered water for eating, cooking and food preparing, and sometimes bathing children.

The KAF maintenance durations varied from one household to another, generally on a ten to fifteen day basis, depending on the daily workload of a filter and its filtration rate. People normally cleaned their filters when they observed a slow filtration rate.

On the other hand, the KAFs were not working under optimum conditions. Of the nine KAFs in the scope of this study, two filters had slow flow rates because it was close to their maintenance date, and yet users were asked to postpone the maintenance until the study finished. Four filters had an uneven sand surface with deep water holes on it although iron nails were evenly distributed. It was shown that neither the diffuser plate nor bricks covering the iron nails showed any effects on sand surface conditions. Four filters had supernatant water depths out of the 4 to 6 cm range, three of those had water levels lower than 4 cm then sand was scooped out while one filter had a

water level higher than 6 cm and more sand was added at the last day of the study when all sample collecting was done.

Moreover, all households did not follow the recommended operation and maintenance for KAF in one way or another. First and most commonly, due to their high demand for clean filtered water, the daily amount of water fed to the KAF in all households exceeded a few times that of CAWST recommendation.

In addition, most of the users utilized any available bucket for water dosing or used the same bucket for raw water collection and filtered water storage. This may result in recontamination of the treated water.

It is worth mentioning that, for their own convenience, some users might not always use the same water source as previously listed in the ongoing research. This caused bias in the past results and data analysis by CAWST and negatively affected the filter performance by not following the guidance in CAWST (2012) of using only one water source for better water treatment.

Last but not least, some of the users did not maintain KAFs properly. Common mistakes were poor conditions of cleanliness (a few filters were dirty, iron nail reservoirs had dried leaves and insects, the outlet tube and storage container had stains) and inaccurate maintenance (users did not practice the swirl and dump method properly). The significance of proper maintenance was illustrated by an unexpected claim. A household not in the scope of this study complained about a KAF malfunction. The filtered water from that KAF was black and smelly. After the KAF was flushed and cleaned appropriately, filtered water quality met the user's expectation.

Regarding iron nails observations, the conditions of nail corrosions described by the appearance of nails in filters of different groups were presented in Table 5. In general, iron nails did not rust

much in all filters with no orange rust flakes observed regardless of the nail types. This may be a result of the silt coverage on nail passivating the iron surface.

Table 5 Observation of nail corrosion states

Filter group	Filter #	Grade	Description
A (New nails + embedded nails)	2	3B	Silt covers on nails, no visible orange
	11	3.5B	Slightly orange, very slight rust on sand
	13	3B	Silt covers on nails, very slight orange rust on filter sides
B (New nails)	7	1.5A	Silt covers on sand, no rust
	9	3E	Iron nails had black rust, started to become more orange when removed from the filter, black rust came off easily.  Very slight rust on sand
	10	2A	Some spots of orange, no visible rust on sand
C (Old nails)	3	3A	Silt covers on nails, no visible orange rust on nails
	19	4C	Visible orange rust on nails, some visible areas of orange on sand surface
	20	3B	Silt covers on nails, no visible orange rust on nails

#### 4.5 Field test results and discussion

Groundwater in different tube wells in Nawalparasi had a slightly above neutral pH (Table 6) with a high level of average turbidity of  $47.2 \pm 45.6$  NTU, a high level of average TDS of  $702.7 \pm 10.3$   $\mu$ S, an average alkalinity of  $386.3 \pm 10.7$  ppm, an average Ca hardness level of  $111.06 \pm 11.78$

ppm, an average Fe level of  $2.31 \pm 0.96$  ppm, an average DO level of  $7.09 \pm 0.45$  ppm and an average temperature of  $26.9 \pm 0.7$  °C. With a positive LSI, the groundwater was non-aggressive and favored CaCO<sub>3</sub> precipitation.

Table 6 Average pH of water samples collected from three filter groups

	Filter group A	Filter group B	Filter group C
pH in RW	$7.6 \pm 0.1$	$7.5 \pm 0.1$	$7.5 \pm 0.1$
pH in SW	$7.8 \pm 0.1$	$7.8 \pm 0.0$	$7.8 \pm 0.1$
pH in FW	$7.9 \pm 0.1$	$7.9 \pm 0.1$	$7.9 \pm 0.0$

The pH values of water samples in three filter groups were similar to each other and showed a gradual rise from RW to FW, and the temperature of water samples was also observed as the same increasing trend. These phenomena can explain an exothermic reaction of iron corrosion (McNeill & Edwards, 2002) as follows and other reactions listed in the literature review part.



The reaction releases heat and produces hydroxide ions which increases the pH of the solution. However, pH did not increase significantly due to the buffer effect of alkalinity which was high in the water samples.

As previously mentioned in the limitations of this research, since the measured raw water was only fed once while the household treated several water doses a day, the collected filtered water was not from the same dose as the raw water. Therefore, removal efficiency cannot be calculated, only comparisons of contaminant levels in each water type of the same day can be conducted. Levels of As and P in raw water (RW), supernatant water (SW), and filtered water (FW) of all filter groups

showed a consistent trend with the descending order of filter performance is Filter group A > Filter group C > Filter group B (Table 7). This means, in terms of nail types, filters using old nails performed better than those using new nails. And filters with extra nails embedded in sand performed better than those without them. This conclusion agreed with findings in the data analysis of previous water quality assessment conducted for the same filter set.

Filters without embedded nails in group B (new nails) and group C (old nails) had concentration of As and P in FW higher than that in SW (which means negative removal) while filters in group A showed a positive removal. This means desorption of contaminants had occurred in the filters without embedded nails. The As level in FW of filter group A was 3.5 times lower than that of filter group B. This means embedded nails significantly facilitated removal performance. The DO levels in the FW filter groups A, B, and C were  $7.11 \pm 0.39$ ,  $7.03 \pm 0.44$ , and  $7.12 \pm 0.52$  ppm, respectively. This shows there was an adequate amount of DO for the embedded nails in the filter media to rust.

Table 7. Average levels of As and P in water samples of three filter groups

	Filter group A (New + embedded nails)	Filter group B (New nails)	Filter group C (Old nails)
Average As in RW (ppm)	$0.28 \pm 0.06$	$0.39 \pm 0.24$	$0.26 \pm 0.15$
Average As in SW (ppm)	$0.18 \pm 0.08$	$0.17 \pm 0.08$	$0.12 \pm 0.05$
Average As in FW (ppm)	$0.08 \pm 0.05$	$0.28 \pm 0.20$	$0.15 \pm 0.08$
Average P in RW (ppm)	$0.42 \pm 0.11$	$0.45 \pm 0.12$	$0.29 \pm 0.36$
Average P in SW (ppm)	$0.44 \pm 0.08$	$0.40 \pm 0.18$	$0.37 \pm 0.10$
Average P in FW (ppm)	$0.19 \pm 0.00$	$0.50 \pm 0.18$	$0.43 \pm 0.06$

On the third day of sampling, raw water (RW) samples from a public tube well supplying for three households (number 7, 13 and 20) were tested during 3 different time points: in the morning, at noon and in the afternoon. Raw water test results showed that the water quality of the same source changed considerably at different time points in a day. Regardless of the close distance between the groundwater sources, groundwater conditions were inconsistent. Those two facts may be explained as a consequence of shallow tube wells in Nawalparasi.

High turbidity in groundwater in Nawalparasi is one of the primary concerns because 1) it requires more frequent filter cleaning resulting in the loss of the biolayer on top of the sand, 2) it causes oxygen depletion in water and in the filtration process which leads to a poor chemical removal of KAF, and 3) silt covers the iron nail surface inhibiting iron corrosion and co-precipitation with arsenic. CAWST researchers suggested KAF users to use alum stones to pretreat their raw water before feeding their filters to lower water turbidity and facilitate water treatment efficacy of the KAF. Even though KAF users accepted to try using the alum stones, they did not really support this idea for various reasons. Firstly, they did not like the taste of water after treatment with alum. Secondly, the flocculation with alum took time and they did not want to wait for dosing because of a high demand on daily treated water. Finally, purchasing alum stones would add cost to the water treatment process.

5.1     Findings of the lab research

The results from this study demonstrated that iron corrosion and arsenic removal by the lab-scaled KAFs were notably dependent on water composition. Both quantitative data from UV-Vis and ICP-MS analysis of dissolved iron corrosion products and qualitative data from SEM/EDS and XRD analysis of iron nail surface illustrated remarkable differences between iron corrosion products from different water types. Total dissolved iron in supernatant water of low hardness was higher than of high hardness while the reverse was true for the level of Fe(II). This means further oxidation of Fe(II) and can occur in the supernatant water leading to additional arsenic removal in filter treating hardness-rich water.

The roles of hardness and phosphate played on arsenic removal process observed in this laboratory research was in accordance with previous studies, in which hardness was a strong enhancing factor and phosphate was an inhibiting one. The effect of hardness surpassed that of phosphate. Calcium hardness profoundly contributed to the impact of hardness rather than magnesium. Filters running hardness-rich water (200 ppm Ca) removed arsenic (initial concentration of 2000 ppb) to under the limit of Nepali water standard (50 ppb) while filter running low hardness water removed much less arsenic, especially in the presence of phosphate. On average, 35.7 to 47.9% of arsenic was removed after contacting with iron nails while total arsenic removal was 85.2 to 99.5% over the duration of this study.

XRD and TEM analysis of iron corrosion products as suspended solids in supernatant water demonstrated an amorphous structure which was expected to be ferric arsenate (Fe(III)-As(V)) complexes formed through a transformation of the arsenate ions adsorption on the surface of iron(hydr)oxides (Yang et al., 2015; Mak et al., 2009). TEM images demonstrated that the size of



the amorphous colloids was very small. If no coagulation occurred, these tiny colloids carrying arsenic could easily pass through the filter media carrying all the contaminants and resulting in a low removal efficiency of the KAF.

## 5.2 Conclusions from the field research

The physical conditions of the studied KAFs in Nawalparasi district were acceptable. The inconsistencies in performance of those filters were mostly caused by variations of groundwater quality, improper methods of maintenance and operation of the filter users and unfavorable conditions of the groundwater.

It was unexpectedly discovered that some households used alternative tube wells at their own convenience for many reasons. A direct consequence of source water switches was biased results in research, especially regarding past data of water compositions. It was impossible to identify if the fluctuations in water quality of those households were caused by low performance of KAFs, or groundwater conditions, or the random switches of groundwater source itself.

The excessive use of KAFs and the elevated turbidity with fine dust in the groundwater in Nawalparasi led to several negative impacts on the performance of the KAFs. First and foremost, iron corrosion was prevented as a result of dust coating. Secondly, the rest time of the KAFs was jeopardized. As a result, there was less time for the microbes living in the biolayer to digest the pathogens in the water, for the natural death of pathogens due to a lack of food or air (CAWST, 2012) and for further corrosion of wet iron nails by exposing to air. Thirdly, filter clogging was enhanced, which required more frequent maintenance than the suggestions by CAWST of every one month to six months and disturbed the biolayer which takes some months to develop.

The observations of iron nails in KAFs of the studied area showed a poor corrosion of iron with a layer of silts and fine particles covering the nail surface. This agreed with the above-mentioned impacts of the excess operation of the device and the high turbidity of the groundwater in Nawalparasi.

In agreement with the results from previous water assessments of CAWST on the same KAFs, embedded nails improved the removal efficiency of KAFs remarkably and new nails worked worse than old nails. Even though both types of iron nails did not rust well in the three KAF sets, they still showed a noticeable impact on the filter performance.

### 5.3 Recommendations for future work

As shown in this study, contaminants were desorbed from the filter media of the studied KAFs and lab-scale filtered after a treatment period, leading to a decrease in filtration efficacy. Further study should be done on the desorption mechanism of contaminants, particularly arsenic desorption, and how to prevent that phenomenon.

To date, research studies on arsenic removal efficiency of the KAF mostly focus on chemical and physical aspects. Therefore, in the near future, the influence of natural organic matter and biological processes (as a result of the metabolism, retention and transport of microorganisms within the KAF) on iron corrosion and arsenic removal should be addressed for a comprehensive understanding of the performance of the KAF.

The field research in Nawalparasi district demonstrated a serious impact of high turbidity on filter performance yet the filter user did not want to use an affordable and simple chemical pretreatment like alum due to some reasons. Therefore, a simple equipment for physical straining should be design and install in the reservoir above the layer of iron nails so that fine dust and silt are removed

before contacting with nails without retard treatment time. It is suggested for this purpose is to use a plastic container with holes at the bottom or a mesh sieve, which is covered in cloth and fits in the reservoir a few centimeters above the nail layer. The cloth is locally available, easy to clean and reusable while it helps remove solid particles without any chemical treatment.

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