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### Metallic iron filters for universal access to safe drinking water

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- 10 Abstract

11 The availability of sustainable safe drinking water is one of Millennium Development Goals 12 (MDGs). The world is on schedule to meet the MDG to "halve by 2015 the proportion of people without sustainable access to safe drinking water in 2000". However, present 13 14 technologies may still leave leaves more than 600 million people without access to safe water 15 in 2015. The objective of the present article is to present a concept for universal water filters primarily made of metallic iron (Fe<sup>0</sup>) and sand. The concept of Fe<sup>0</sup>/sand filters is based on the 16 17 combination of: (i) recent development of slow sand filtration and (ii) recent progress in understanding the process of contaminant removal in  $Fe^{0}/H_{2}O$  systems. The filters should be 18 made up of more than 60 % of sand and up to 40 % of Fe<sup>0</sup>. The actual Fe<sup>0</sup> proportion will 19 20 depend on its intrinsic reactivity. The most important question to be answered regards the 21 selection of the material to be used. The design of the filter can be derived from existing filters. It appears that  $Fe^{0}/H_{2}O$  based filters could be a technology for the whole world. 22

- 23 Keywords: Contaminant removal, Point of use, Safe water, Water filtration, Zerovalent iron.
- 24 Acronym List
- 25 AMD Acid mine drainage
- 26 BSF Biosand filter
- 27 CIM Composite iron matrix
- 28 EDTA Ethylenediaminetetraacetate
- 29 KAF Kanchan<sup>TM</sup> arsenic filters

- 30 MDG Millennium Development Goal
- 31 NGO Non-governmental organization
- 32 POU Point-of-use
- 33 ZVI Zerovalent Iron

#### 34 Introduction

Naturally readily available waters (shallow groundwater; surface water, water from boreholes and springs) are the main sources for drinking water production. These waters may become contaminated with organic/inorganic chemical pollutants and pathogenic microorganisms (bacteria, fungi, protozoa, viruses) from various origins: (i) natural (geogenic) hydrogeochemical processes, (ii) artificial recharge with wastewater or water from septic tanks or leaking sewage pipes, (iii) discharges of wastewater and/or manure run-off from agricultural land.

42 To produce safe drinking water from natural waters, chemical pollutants and pathogens need 43 to be removed. One effective natural way is the passage of surface water through soil, as is 44 the case in bank filtration [1-4]. The effectiveness of bank filtration to produce safe drinking 45 water from natural waters depends on four major factors: (i) the extent of water pollution, (ii) 46 the nature of the soil, (iii) the thickness of the soil or the travel distance within the soil, and 47 (iv) the water flow rate or travel time. One can learn from nature and produce drinking water 48 at small scale. For this purpose, it can be assumed that for a sufficient amount of any relevant 49 treatment material in a filter, an adequate water flow rate will yield satisfactory contaminant 50 mitigation. The reactive material can be (i) a natural material, (ii) a synthetic material or 51 composites, or (iii) a mixture of materials. The material amount determines the thickness of 52 reactive layer in the filter. The water flow rate determines travel time.

53 During the past three decades, treating water at the household level has been shown to be one 54 of the most efficient means of preventing waterborne diseases world wide [5-10]. Here are six 55 examples for illustration: the use of (i) commercial filtration devices (e.g. Brita) mostly in developed countries [11-13], (ii) ceramic pot filters or Kosim filters in Ghana [14], (iii) 3-Kolshi filters and SONO filters in Bangladesh [6, 15, 16], (iv) Kanchan<sup>TM</sup> arsenic filters in Nepal [17, 18], (v) Danvor plastic biosand filter [19], and (vi) Potters for Peace Filtron ceramic filter [19,20]. Promoting household water treatment and safe storage helps populations to actively take charge of their own water security. Therefore, providing people with the knowledge and affordable tools to treat their own drinking water at the point of use is a noble objective.

The present study aims at presenting a concept using metallic iron (Fe<sup>0</sup>) as universal filter 63 64 material for water treatment at the point of use (POU) in general and at household level in particular. The secondary objective is to demonstrate the suitability of Fe<sup>0</sup> for water treatment 65 in remote locations of developing countries. In these regions, available water could be 66 67 chemically safe (no geogenic or anthropogenic contamination). In this case, microbial 68 contamination due to lack of sanitation and improper hygiene is the sole source of water 69 contamination. Table 1 summarises some relevant water pathogens and related diseases [21]. 70 The presentation is based on the state-of-the-art knowledge on the mechanism of contaminant removal in the presence of  $Fe^0$  ( $Fe^0/H_2O$  systems) which will be presented first. 71

# 72 Mechanisms of contaminant removal in Fe<sup>0</sup>/H<sub>2</sub>O systems

Passive remediation of water pollution by using Fe<sup>0</sup>/H<sub>2</sub>O systems has been developed for 73 74 about 20 years [22-24]. Applications of these innovative systems included (i) groundwater 75 remediation, (ii) drinking water treatment, and (iii) wastewater treatment. Successful 76 quantitative removal of metals (e.g. Cd, Co, Cr, Cu, Pb, U, Zn), non-metals (e.g. As, Mo, Se, Sn), anions (e.g. AsO43-, F, MoO42-, NO3-, PO43-, SeO42-, SO42-, SO42-), organic dyes, organic 77 compounds (e.g. benzene, chlorinated solvents, phenol, pesticides, toluene), bacteria, 78 79 suspended solids, and viruses has been reported [16, 25-30]. Almost all studies dealing with pollutant removal were limited to proving the viability of Fe<sup>0</sup>/H<sub>2</sub>O systems for a few target 80

pollutants and were not incorporated within a broad-based understanding of Fe<sup>0</sup> remediation
technology.

It is interesting to note that another technology, electrocoagulation [31-33], using  $Fe^0$ , could exhibit similar efficiency for a large spectrum of contaminants. Iron electrocoagulation can be considered as an electrochemically driven accelerated corrosion process. In both cases (passive  $Fe^0/H_2O$  and iron electrocoagulation), pollutants are removed from water by a multitude of mechanisms operating synergistically.

Likely removal mechanisms include: (i) adsorption onto  $Fe^0$  and Fe oxides/hydroxides, (ii) 88 89 co-precipitation with Fe oxides (or co-precipitation on the substrate), (iii) direct reduction by  $Fe^{0}$ , (iv) indirect reduction by  $Fe^{II}$  or  $H/H_{2}$ , (v) indirect oxidation by in situ generated radicals 90 91 HO. The exact sequence of these reactions depend on the system. Because redox-insensitive pollutants have been quantitatively removed in passive  $Fe^{0}/H_{2}O$  systems, any individual redox 92 93 process could not be the fundamental removal mechanism as originally assumed [23, 34]. Therefore, the fundamental mechanisms of contaminant removal in Fe<sup>0</sup>/H<sub>2</sub>O systems are 94 95 adsorption (including surface complexation) and co-precipitation [35, 36].

96 Certainly, for some contaminants, such as  $Cr^{VI}$  which are reducible even in the aqueous phase 97 by Fe<sup>II</sup> (under local anoxic conditions), quantitative reduction may precede adsorption and co-98 precipitation. However, considering the nature of the Fe<sup>0</sup>/H<sub>2</sub>O system, there is no reason to 99 consider quantitative reductive transformation a priori for some species. Moreover, a Fe<sup>0</sup>/H<sub>2</sub>O 100 system should be considered as zone of precipitating iron oxides/hydroxides [37]. During this 101 process expansive amorphous iron hydroxides are generated and further transformed by 102 dehydration to more crystalline oxides (Tab. 2) [38, 39].

# 103 Diversity in the reactivity of iron oxides in Fe<sup>0</sup>/H<sub>2</sub>O systems

In order to better understand the role of metallic iron in  $Fe^{0}$ /sand filters, the dynamic nature of iron corrosion in  $Fe^{0}/H_{2}O$  systems will be discussed. For this purpose, synthetic bulk oxides and oxide-films on  $Fe^{0}$  should be compared in terms of reactivity towards contaminant

107 removal. Synthetic oxides and in situ formed oxide-films are fundamentally different in that 108 the latter are reactive systems ("reactive oxides") by virtue of the continual generation and 109 annihilation of point defects at the interfaces. Furthermore, oxide-films (passive layers), unlike synthetic oxides, continually grow into the metal at the Fe<sup>0</sup>/layer interface while being 110 111 simultaneously destroyed by dissolution or restructuring at the layer/H<sub>2</sub>O interface [40, 41]. 112 Accordingly, synthetic oxides are strictly "coatings" and hence, with respect to the processes that influence Fe<sup>0</sup> reactivity, they are "dead" [40]. Synthetic oxides can only act as 113 114 contaminant adsorbents. Although synthetic oxides may be capable of simulating the 115 properties of the deposited outer layers (at the layer/H<sub>2</sub>O interface), it is difficult to see how they can simulate the barrier layer (at the Fe<sup>0</sup>/layer interface - "reactive oxides"), whose 116 117 defect concentration is normally far in excess of that which can be obtained in bulk oxides. "Reactive oxides" are better adsorbents and may co-precipitate contaminants during their 118 119 formation and transformation to "dead oxides" [25, 35, 36, 42, 43]. "Dead oxides" are good 120 adsorbents of limited and selective affinity to some contaminants. The consequence of this analysis is that Fe<sup>0</sup> can be regarded as a permanent source of highly reactive hydroxides 121 122 ("reactive oxides") in a treatment system (statement 1). Statement 1 is the major argument on which the concept of Fe<sup>0</sup>/sand filters for universal access to safe drinking water is built. 123

124 A schematic illustration of the time-dependant evolution of iron corrosion products is given in Figure 1. The cross-section of a spherical  $Fe^0$  material experiencing uniform corrosion is 125 represented. It is assumed that the initial material ( $Fe^0$  in Fig. 2) is progressively covered by 126 127 concentric layers of iron oxides from which the three most internal layers  $(d_1, d_2, d_3, Fig. 2)$ 128 are "reactive" (see above) and all the external layers  $(d_4)$  are comparative in their reactivity to 129 synthetic bulk oxides ("coatings"). While this assumption is somewhat arbitrarily,  $d_1$  can be regarded as a layer of amorphous ferrous iron hydroxides (Fe(OH)<sub>2</sub>), d<sub>2</sub> a layer of mixed 130 131 amorphous iron hydroxides (Fe(OH)<sub>2</sub> and Fe(OH)<sub>3</sub>),  $d_3$  a layer of mixed amorphous iron 132 hydroxides/oxides (green rust, magnetite) and  $d_4$  a layer of aged corrosion products 133 (crystallized iron oxides).

134 The schematic representations in Fig. 1 and 2 have intentionally neglected the expansive 135 nature of the process of iron oxide/hydroxide production form metallic iron [39, 44]. In fact, the theoretical ratio ( $\alpha_1 = V_{oxide}/V_{Fe}$ ) between the volume of corrosion products and the 136 137 volume of iron in the metallic structure varies between 2.0 for Fe<sub>3</sub>O<sub>4</sub> and 6.4 for 138 Fe(OH)<sub>3</sub>.3H<sub>2</sub>O (Tab. 2). This volume increase is the principal cause of the expansion and ultimately the loss of hydraulic conductivity (permeability loss) for remediation Fe<sup>0</sup>/H<sub>2</sub>O 139 systems. Clearly, Fe<sup>0</sup>/sand filters end of service is not (or should not be) dictated by Fe<sup>0</sup> 140 141 depletion but rather by loss of hydraulic conductivity. The practical expansion of corrosion products can be discussed from a proper kinetic law which correlates the radius loss of Fe<sup>0</sup> 142 143 and the corresponding volume of accumulated iron oxides [39]. In this work, mixing inert sand and  $Fe^0$  is a practical tool to extent filter service life. It is expected that for any 144 appropriate Fe<sup>0</sup> material, an optimal weight ratio sand/Fe<sup>0</sup> should exist for satisfactorily water 145 146 treatment in the medium or long term (e.g.  $\geq 12$  months).

#### 147 Water treatment by sand filtration

Promising strategies for providing people with access to safe water are available, but may not be suitable/affordable for areas of low population density with little perspective for economic growth (e.g. rural areas, [45, 46]). It may further prove challenging to guarantee the quality of drinking water in these populations. Methods that allow the treatment of the water at the place where it is consumed (POU methods) may provide a low-cost, promising, easy and flexible solution for increasing drinking water quality in much of the population in need. Slow sand filtration is one of these methods.

#### 155 **Basic sand filtration**

156 The earliest form of water treatment was slow sand filtration. Slow sand filters were 157 developed in the 1820's in Scotland by Robert Thom and in England by James Simpson. They

158 became successfully established in Europe by the end of the 19th century [47-50]. This 159 technology used sand filter beds through which water is slowly trickled. The natural 160 formation of a biological layer (biofilm, widely termed as Schmutzdecke) and the filtering 161 action of sand removes bacteria, silt and chemical pollutants. The treatment efficiency is 162 affected partly by physical straining but more importantly by biological action within the 163 biofilm that formed on top of the sand [51]. A household-scale intermittent slow sand filter 164 (the Davnor Biosand filter or Davnor BSF) developed by Manz has been successfully tested 165 by several governments, research and health institutions and NGOs in Bangladesh, Brazil, 166 Canada, Haiti, Nicaragua, Vietnam and other countries [52, 53]. Lantagne et al. [5] 167 summarized the drawbacks of the BSF as follows: (i) low rate of virus inactivation, (ii) lack 168 of residual protection and removal of less than 100 percent of the bacteria, which leads to 169 recontamination, (iii) current lack of studies proving health impact, and (iv) difficulty in 170 transport and high initial cost, which make scalability more challenging. Accordingly the 171 major problem with BSF is that complete pathogen removal can not be guaranteed. Therefore, 172 if water can be successfully freed from pathogens within a sand filter, this technology can be 173 used with more confidence for safe drinking water at a household level. The presentation on the mechanisms of contaminant removal in  $Fe^{0}/H_{2}O$  systems suggests that coupling reactive 174  $Fe^{0}$  to sand could solve the problem (statement 1). This work has already been done but  $Fe^{0}$ 175 176 aimed at eliminating arsenic as presented below.

### 177 Improved sand filtration

Recently, the Arsenic Biosand Filter (Kanchan<sup>TM</sup> Arsenic Filter - KAF) was developed and distributed in Nepal by Ngai et al. [17, 18]. The KAF is built on the platform of a slow sand filter, modified to include/increase arsenic removal capability. The KAF combines the concepts of slow sand filtration and an intermittent household-scale system with the innovation of a diffuser basin containing iron nails for arsenic removal. In the KAF, arsenic is certainly quantitatively removed by adsorption onto or by co-precipitation with iron oxyhydroxides from rusting iron nails. Pathogens (e.g. bacteria) have been reported to be removed mostly by physical straining provided by the fine sand layer, by attachment to previously removed particles and, to a lesser degree, by biological predation occurring in the top few centimeters of the sand. The KAF was demonstrated successful for simultaneous arsenic and pathogen removal and is considered as the best among all household arsenic filters available in Nepal [18]. Chiew et al. [54] have recently published primary results on attempts to extent the concept of KAF filtration in Cambodia (Fig. 3).

191 Recent developments in the understanding of the process of contaminant removal in  $Fe^{0}/H_{2}O$ 192 systems demonstrates that pathogens are certainly removed/inactivated by co-precipitation 193 with iron oxyhydroxides from a rusting  $Fe^{0}$ . Therefore, a further development of the 194 Kanchan<sup>TM</sup> Arsenic Filter is to disseminate  $Fe^{0}$  in of the "pathogen removal unit" of the filter 195 (BioSand filter – Fig. 2) or to replace the "arsenic removal unit" by a water saturated  $Fe^{0}$ /sand 196 layer. The second possibility is discussed below. Such a filter will remove all possible 197 contaminants (including arsenic and bacteria) from the aqueous solution.

### 198 **Fe<sup>0</sup>/sand filters**

199 The works of Dr. Manz yielded an efficient intermittent household-scale slow sand filtration 200 system for "safe" drinking water [50]. Ngai et al. [18] further developed this system by 201 adding small amount of metallic iron to optimize arsenic (and pathogen) removal. As a rule 202 the nature and extent of water contamination in rural areas of developing countries is 203 unknown [46]. However, despite the variability of the influents the system outputs have to be 204 satisfactory. Based on the state-of-the-art knowledge on the mechanism of contaminant removal by Fe<sup>0</sup>/H<sub>2</sub>O systems, the used of metallic iron as universal material for safe drinking 205 206 water has been suggested [55, 56]. The works of Ngai et al. [18] suggested that the amount of Fe<sup>0</sup> in the filter is not necessarily high (e.g. 10 % weight). Furthermore, since chemical 207 208 contaminants and pathogens are certainly co-precipitated with precipitating iron corrosion products, the flow rate of contaminated waters through the Fe<sup>0</sup>/sand filter can be enhanced to 209

210 reach the daily demand of individual families within a few tens of minutes. That is the idea 211 behind  $Fe^{0}$ /sand filters.

### 212 The innovation in Fe<sup>0</sup>/sand filters

People familiar with the problematic of arsenic contamination in Bangladesh and Nepal are 213 aware that an effective Fe<sup>0</sup>/sand filtration system (3-Kolshi system) was abandoned because 214 215 of loss of porosity of the system. A close look of the 3-Kolshi filtration system reveals that the 216 top Kolshi was filled with 3 kg of cast iron and 2 kg of sand on top of the iron turnings [15]. In other words the Kolshi with  $Fe^0$  (here cast iron) contains a zone of 100 %  $Fe^0$  overlying the 217 218 sand layer. The 3-Kolshi filtration system was replaced by the SONO filter. Here, the primary 219 active material is a porous composite iron matrix (CIM), a mass made of cast iron turnings 220 through a proprietary process to maintain active CIM integrity for years [6, 16]. Again a 100 221 % layer of composite is sandwiched between two layers of (coarse) sand. The reported loss of porosity is necessarily coupled to the too high proportion of  $Fe^{0}$  in the reactive zone (actually 222 223 100 %). Given the relative low concentration of arsenic in contaminated waters, this huge amount of Fe<sup>0</sup> (and possibly composite) is obviously unnecessary. As discussed above Fe<sup>0</sup> 224 225 depletion should not occur. However, even the KAF of Ngai et al. [18] contains a 100 % layer 226 of iron nails at the top of fine sand, coarse sand and gravel. The relative amount of iron nails 227 in KAF was small compared to that of reactive materials in 3-Kolshi and SONO filtration 228 systems and the KAF are efficient for several years [17]. Therefore, reducing the proportion 229 of  $Fe^{0}$  in the system is a sensible modification to maintain filter permeability for a long time.

The real novelty with the proposed  $Fe^{0}$ /sand filters is that no 100 %  $Fe^{0}$  will be available. Rather, the  $Fe^{0}$  reactive layer will be a mixture of at least 60 % sand, gravel or porous volcanic rocks and up to 40 %  $Fe^{0}$ . The actual proportion of the  $Fe^{0}$  will depend on its intrinsic reactivity and particle size. The advantage of porous volcanic rocks [57-59] is that generated iron hydroxides may fill their porous structure extending service life (or retarding loss of hydraulic conductivity). On the other hand,  $Fe^{0}$  materials could be produced locally 236 [60-63] or selected from available iron products (including production wastes and by-237 products) such as nails rivets, nuts, bolts, barbed wire, packing wire, chicken wire mess, fencing wire, steel wool, construction materials and other Fe<sup>0</sup> products. Using available Fe<sup>0</sup> 238 materials will protect filter users from the market law as increased demand is only indirectly 239 240 coupled to the original use of the material. For example, it can not be expected that the price of packing wire increases just because it is used for water treatment. Another approach will 241 consist of testing potential Fe<sup>0</sup> materials in industrialised countries (including China), that are 242 243 likely exported to developing countries and built a database for suitable materials.

### 244 **Potential beneficiaries of Fe<sup>0</sup>/sand filters**

It is certain that Fe<sup>0</sup>/sand filters will accelerate the health gains associated with improved 245 drinking water until the goal of universal access to piped, treated water is achieved [5]. Even 246 247 after this hypothetical goal is achieved, the world will still have to face critical situations (e.g. 248 accidental contamination, earthquakes, epidemic plagues, wars, tsunamis), in which available 249 water should be rapidly treated. These critical situations are managed worldwide mostly by armies and NGOs (including the Red Cross). Therefore, the Fe<sup>0</sup>/sand filters are not only 250 251 suitable for developing countries but for the whole world. Recently, an uranium 252 contamination was discovered in drinking water production wells in Barlissen in Lower 253 Saxony/Germany and the wells were precautionary put out of service [64]. Barlissen is a 254 typical situation where Fe<sup>0</sup>/sand filters could help in industrialized countries.

On the other hand, working on  $Fe^{0}$ /sand filters will give researchers from developing countries the opportunity to solve a crucial problem with local solutions. The technology is low cost and good results can be achieved with simple equipment. Furthermore, experiments do not involve any stringent reaction conditions nor expensive devices, as the major experiments are to be conducted under atmospheric conditions. It is expected that research groups working on  $Fe^{0}$ /sand filters will achieve results which are helpful for the further development of iron reactive walls. In fact,  $Fe^{0}$ /sand filters can be regarded as a sort of "rapid small scale column test" which could help to bridge the gap between short-term studies in the lab (few weeks) and field  $Fe^0$  reactive barriers (some two decades). In fact, testing  $Fe^0$ /sand filters offers a unique opportunity to test the reactivity of the same  $Fe^0$  material at several sites with natural waters of various characteristics. Clearly, a South/North symbiosis can be expected: a technology developed in the North tested and used in a modified version in the South could contribute to the further development of the original technology (not only for the North).

## 269 Ways to affordable Fe<sup>0</sup>/sand filters

The concept of Fe<sup>0</sup>/sand filters is based on scientific understanding of the complex chemical 270 271 and physical processes involved in an evolving technology (iron remediation technology) that has being successfully applied for almost 20 years. Provided that a relevant reactive Fe<sup>0</sup> is 272 used, the effectiveness of  $Fe^{0}$ /sand filters is not to be demonstrated, except some technology 273 274 verification in the field (monitoring). The sole tasks are: (i) selecting and processing the appropriate  $Fe^0$  materials, (ii) designing the filters, and (iii) avoiding the use of  $Fe^0$ /sand 275 filters for waters of pH  $\leq$  5. In fact, Fe<sup>0</sup>/sand filters are based on the anodic dissolution of iron 276 277 in neutral and close-to-neutral aqueous systems. In this pH range, primary iron dissolution is 278 followed by a continuous build up and transformation of a corrosion product layer in the vicinity of Fe<sup>0</sup> [41, 65, 66]. 279

### 280 Material selection

Practically any available  $Fe^0$  material (mainly low alloyed steel and cast iron) is theoretically applicable for water treatment. Ideally, any newly obtained  $Fe^0$  material should be characterized and tested for water treatment capacity by a standard method. In using activated carbon for wastewater treatment for example, it is generally accepted that a good decolorizing carbon should fulfil at least 200 mg/g removal capacity for methylene blue in batch experiments [67]. Until recently, there was no experimental parameter to characterize the intrinsic reactivity of  $Fe^0$  materials [68-70]. Noubactep [69, 70] has introduced a parameter,

 $k_{EDTA}$ , which could enable purposeful material selection. Per definition,  $k_{EDTA}$  is the slope of 288 the line of the time-dependent oxidative dissolution of ion from a given Fe<sup>0</sup> material in a 2 289 mM EDTA solution.  $k_{\text{EDTA}}$  is determined in batch experiments and characterized material 290 intrinsic reactivity under any given experimental conditions [70]. For example, materials used 291 292 in Fe<sup> $^{0}$ </sup>/sand filters should exhibit a k<sub>EDTA</sub> value above a critical value. This critical value is yet 293 to be determined. The k<sub>EDTA</sub> value of the iron nails used by Ngai et al. [18] could be used as guide to select the Fe<sup>0</sup> loading of the filter. For a less reactive material a larger Fe<sup>0</sup> loading 294 than in the Kanchan<sup>TM</sup> Arsenic Filter will be applied and lesser Fe<sup>0</sup> loadings are needed for 295 more reactive materials. The long term reactivity of available Fe<sup>0</sup> materials has to be tested as 296 well. Initially, iron nails from the KAF filters or iron composites from the SONO filters can 297 298 be used as starting point.

### 299 Filter designs

Basically, Fe<sup>0</sup>/sand filters are slow sand filters for intermittent use [49, 50]. Almost 130 years 300 301 of experience is available for this type of system. The most recent developments are those of Kanchan<sup>TM</sup> Arsenic Filter [18], SONO filters [6, 16] and Kosim filters [14]. These devices 302 can be modified to use a Fe<sup>0</sup>/sand reactive layer as described above. Alternatively, new 303 304 designs may be conceived based on local specifications [71]. Fig. 3 shows a cross-section of 305 the filter design used by Chiew et al. [54]. Arsenic removal and bacterial removal units are 306 illustrated. A potential shortcoming of this design is that the "arsenic removal unit" 307 experiences wet and dry periods. In an improved design, the high of the outlet pipe should enable immersion of the Fe<sup>0</sup> layer. 308

In all cases the filters should be constructed from local materials and the reactive layer should be embedded in sand layers. Available  $Fe^0$  materials (e.g. iron nails, steel wool) should be tested and approved at least in the early stage of technology implementation. The  $Fe^0$ /sand filter should be manufactured locally by trained workers regardless of their scholar education. To sustain the Fe<sup>0</sup> reactivity, intermittent additions (e.g. once a month) of boiled water [16] or lemon juice [72] could be envisaged.

While Hussam and Munir [16] used boiled water to eventually kill pathogens, the present review demonstrated that upon purposeful dimensioning, pathogens will be sequestrated by iron corrosion products. Therefore, boiled water should increase/sustain  $Fe^0$  reactivity by elevating the temperature. Intermittent temperature elevation certainly disturb the process of iron passivation, and thus sustain  $Fe^0$  reactivity as reported by Hussam and Munir [16].

320 The rational for the use of citric acid is the well-known increase iron corrosion with 321 decreasing pH value [65, 66]. Cornejo et al. [72] tested three commercially available lemon 322 species as citric sources. Commercial lemon species may content undesirable preservatives 323 and little parts of fruit. Even though these undesirable contents are non toxic, these additives 324 should be regarded as contaminants and will be removed in the filtering system as well. In 325 rural areas natural lemon juice should be at least seasonally available at low cost or cheaper than commercial lemon juices. While testing the ability of citric acid ( $pK_{a1} = 3.15$ ,  $pK_{a2} =$ 326 4.77,  $pK_{a3} = 6.40$ ) to sustain reactivity, care will be taken to keep the pH > 4.5 to avoid 327 328 dissolved Fe in filtered water. Again, this technology is not applicable for the treatment of 329 waters of initial pH < 5. Such waters, known as from acid mine drainage (AMD) are world 330 wide available at abandoned and active mining sites [73].

## 331 Conclusions

The fact that field  $Fe^{0}/H_{2}O$  systems have successfully removed various contaminants from polluted water is a testament to their potential, which is yet to be fully realized. This paper presents a concept to realize this potential at a household level by a simple water filtration on a Fe<sup>0</sup>/sand column. The Fe<sup>0</sup>/sand filter is an innovation combining two proven water treatment techniques: (i) adsorption and co-precipitation in Fe<sup>0</sup>/H<sub>2</sub>O systems, and (ii) size exclusion by slow sand filtration. There is no doubt that a proper material selection and design of the filtration system will contribute to achieve and even exceed the Millennium Development 339 Goal. The original merit of this concept is that it allows researchers from developing countries 340 to actively work on a crucial problem while results will be useful for the developed world as well. Intensive laboratory and field research is needed to develop Fe<sup>0</sup>/sand filters. The efforts 341 should be accompanied by numerical modeling. This aspect is under investigation in the 342 343 research group of one of the authors (P. Woafo) using mathematical equations describing the 344 spatio-temporal evolution of pollutants concentration and porosity in the filters complemented 345 by experiments. On the other hand, reportedly efficient filters (e.g. Danvor BSF, Filtron ceramic filter, Kosim filter) can be amended with Fe<sup>0</sup> to enhanced pathogen inactivation. 346

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### 353 **Conflict of Interest**

354 The authors have declared no conflict of interest.

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Microbes	Examples	Diseases	
Bacteria	Vibrio cholerae	Cholera	
	Shigella sonnei	Bloody diarrhea	
	Salmonella enterica	Gastrointestinal illness	
	Helicobacter pylori	Chronic ulcers and cancer	
	Escherichia coli	Bloody diarrhea, kidney failure	
	Arcobacter butzleri	Acute gastrointestinal illness	
Viruses	Astro- and Caliciviruses	Gastrointestinal illness	
	Hepatitis A/E virus	Acute liver disease (hepatitis)	
	Rotavirus	Severe diarrhea	
Protozoa	Blastocystis hominis	Diarrhea and abdominal pain	
	Entamoeba histolytica	Gastrointestinal illness	
	Naegleria fowleri	Amebic meningoencephalitis	
Fungi	Aspergillus fumigatus	Respiratory illness	
	Fusarium solani	Skin-related infections	

**Table 1**: Some relevant water pathogens and related diseases. Compiled after ref. [21].

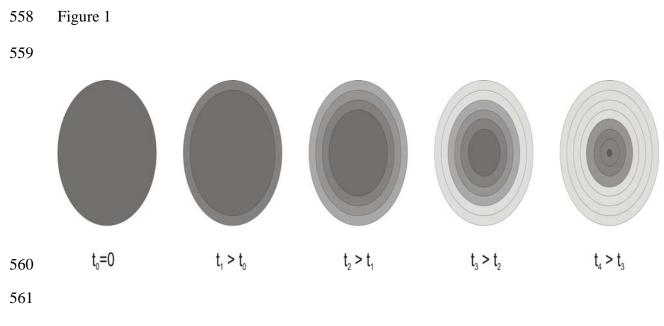
**Table 2**: Some relevant characteristics of metallic iron and its main corrosion products.  $\alpha$  is the molecular weight of iron to the molecular weight of the corrosion products.  $\alpha_1$  is the ratio of volume of expansive corrosion products to the volume of iron in the metallic structure. Compiled from refs. [38] and [39].

555

Name	Formula	Structure	Density	α	α1
			$(kg/m^3)$		
Iron	a-Fe	bcc	7800	_	-
Hematite	1/2 Fe <sub>2</sub> O <sub>3</sub>	Trigonal	5260	0.699	2.12
Magnetite	1/3 Fe <sub>3</sub> O <sub>4</sub>	Cubic	5180	0.724	2.08
Goethite	α-FeOOH	Orthorhombic	4260	0.629	2.91
Akageneite	β-FeOOH	Tetragonal	3560	0.629	3.48
Lepidocrite	γ-FeOOH	Orthorhombic	4090	0.629	3.03
	Fe(OH) <sub>2</sub>	Trigonal	3400	0.622	3.75
	Fe(OH) <sub>3</sub>	n.a.	n.a.	0.523	4.20
	Fe(OH) <sub>3</sub> .3H <sub>2</sub> O	n.a.	n.a.	0.347	6.40

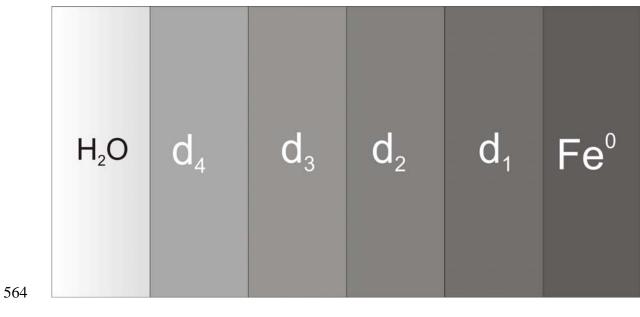
556 n.a. = not available

557

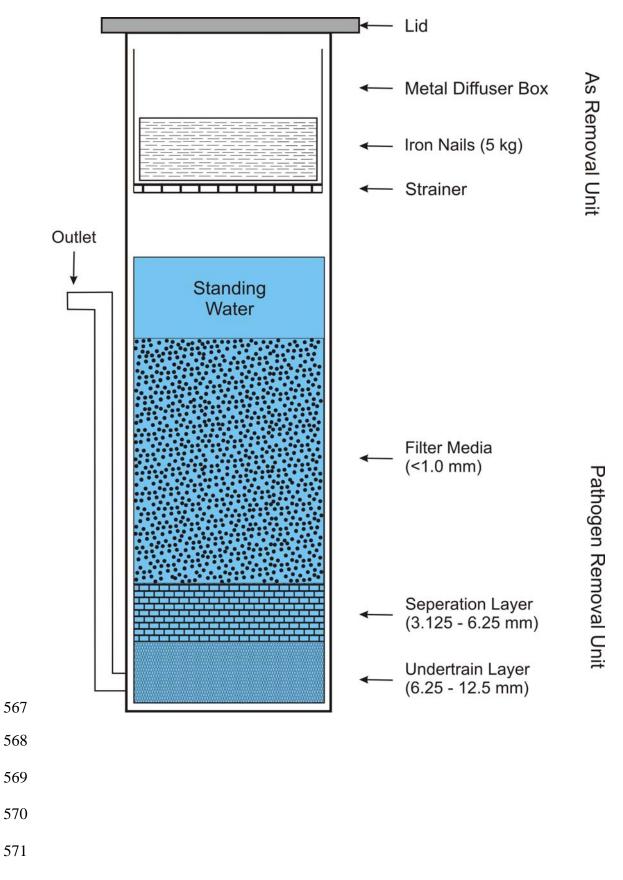


562 Figure 2:

# 







### 571 **Figure captions**

572

**Figure 1**: Time dependence evolution of  $Fe^0$  and Fe corrosion products from a spherical material assuming uniform corrosion.  $Fe^0$  corrosion continuously produces concentric layers of iron hydroxides which are transformed to iron oxides. It is assumed (see text) that the three layers next to  $Fe^0$  surface are reactive (d<sub>1</sub>, d<sub>2</sub>, d<sub>3</sub>) while the outer layers are non reactive (d<sub>4</sub>).

578

579Figure 2: Cross-sectional diagram of a uniformly corroding  $Fe^0$  particle at  $t > t_2$  (Fig. 1). d<sub>1</sub>,580d<sub>2</sub> and d<sub>3</sub> are supposedly the reactive or transforming layers in which contaminants581may be adsorbed, co-precipitated or chemically transformed. d<sub>4</sub> is the growing582layer of aged corrosion products on which contaminants could adsorb. d<sub>4</sub> is less583porous than the inner layers but may be fissured.

584

Figure 3: Design of the filter tested for arsenic and pathogen removal in Cambodia by Chiew
et al. [54]. Rigorously the pathogen removal unit is a conventional BioSand filter
in which suspended solid are removed.