1 **Extending service life of household water filters by mixing metallic iron with**

2 **sand**

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13 **Running Title:** Mixing inert additives and reactive Fe^0 is a prerequisite for sustainable filters.

14

15 **Acronym List**

16 CIM   Composite iron matrix

17 KAF   Kanchan™ arsenic filters

18 MDG   Millennium Development Goal

19 SAF   SONO arsenic filters

20 **Keywords**: Drinking water, Filter clogging; Iron/sand filter; Long-term reactivity; Zerovalent iron.

21

21   **Abstract**

22   The use of metallic iron filters (Fe^0 filters) has been discussed as a promising low-cost option for

23   safe drinking water production at household level. Filter clogging due to the volumetric expansive

24   nature of iron corrosion has been identified as the major problem of Fe^0 filters. Mixing Fe^0 and sand

25   (yielding Fe^0/sand filters) has been proposed as a tool to extent filter service life. However, no

26   systematic discussion rationalizing Fe^0:sand mixtures is yet available. This communication

27   theoretically discussed suitable Fe^0:sand proportions for efficient filters. Results suggested that

28   Fe^0/sand filters should not contain more that 50 vol-% Fe^0 (25 wt-% when Fe^0 is mixed with quartz).

29   The actual Fe^0 percentage in a filter will depend on its intrinsic reactivity.

30   **1   Introduction**

31   Safe water provision in emergency situations and in remote communities is a serious global problem

32   [1-12]. In such situations, available raw waters are often polluted by microbial and chemical

33   contaminants. Several filtration systems suitable for rendering raw waters potable have been

34   presented [1, 2, 7, 13-17]. For example, the Josab Aqualite^{TM} System using a natural zeolite was

35   developed and successfully used during the Balkan conflict and after the tsunami [7]. SONO filters

36   based on a composite-iron matrix (CIM) is extensively used for arsenic removal at household level

37   in rural Bangladesh and Nepal [1, 13]. Josab Aqualite^{TM} Systems use ultra filtration and are

38   available as portable and mobile units weighing from 95 to 1800 kg. Although Josab Aqualite^{TM}

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39   filtration systems can be readily transported to the points-of-need, it is clear that simpler efficient

40   systems like SONO filters may be very helpful even in emergency situations. Other more practical

41   systems similar to Josab Aqualite[TM] Systems have been presented [18-22]. These systems mostly

42   use synthetic membranes and are rarely universally affordable.

43   In the developing world the application of low cost household water treatment technology of

44   variable effectiveness (boiling, chlorination, filtration, solar disinfection) has been promoted in

45   recent years in the frame work of efforts to achieve the Millennium Development Goals (MDG) for

46   water [8, 23, 24]. Filtration was identified as the simplest efficient technology [13, 14 ,17]. The

47   main research question is how to design a good stand-alone filter which operates for long time (e.g.

48   12 months or more) demanding neither electricity nor addition of chemicals. Based on recent

49   progresses on understanding mechanistic aspects of aqueous contaminant removal in the presence

50   of Fe^0 (e.g. in Fe^0/H_2O systems) [25-28]. Fe^0 filters have been suggested as low cost, efficient

51   systems for universal safe drinking water production [17, 29, 30]. Previous efforts using Fe^0 filters

52   were limited to individual contaminants, e.g. arsenic [13, 15] or viruses [31].

53   The main concern with Fe^0 filters is the reduction in pore space caused by mineral precipitation

54   (mineral fouling or filter clogging). Fouling of the pore space reduces the porosity and hydraulic

55   conductivity of filter [17, 32, 33]. Likely causes of filter clogging are: (i) adsorption of fouling

56   substances (e.g. colloids), (ii) bio-corrosion, (iii) cake formation, and (iv) pore filling or filter

57　clogging. Bio-corrosion is microbial accelerated iron corrosion likely yielding to rapid clogging.

58　Generally, filter clogging is solved by using mechanical and chemical cleaning. The systems

59　discussed in this communication should not need any cleaning operation apart from some possible

60　intermittent additions of boiled water or lemon juice as recently discussed [17].

61　The aim of this communication is to bring some design of Fe^0/sand filter based on proper theoretical

62　evaluation. A reactive zone containing Fe^0 is incorporated in the fine sand layer of a conventional

63　slow sand filter. The proportion of Fe^0 in this reactive zone varied from 0 to 100 %. The efficiency

64　of the resulting Fe^0/sand filters are discussed. The efficiency of each filter was evaluated by the

65　extent of filling of the initial pore space upon Fe^0 depletion. A range of suitable combination of Fe^0

66　and sand as filter media was deduced. For the sake of clarity, the process of aqueous iron corrosion

67　and its use in water treatment will be recalled.

68　**2　Aqueous iron corrosion and contaminant removal in Fe^0 filters**

69　Filtration through a layer of a suitable medium or a combination of media (anthracite, gravel, sand)

70　is an important process for water treatment [34, 35]. Based on the rate of filtration, filters are

71　divided into slow and rapid types. Rapid filters mostly clarify water by physical and

72　physicochemical processes with less biological mechanisms. In slow filters on the contrary,

73　biological action plays an important role. Household iron filters discussed in this communication

74　are primarily slow filters. Due to improper consideration of the expansive nature of iron oxidative

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75 dissolution (at pH > 4.5) in a porous Fe^0 bed, early attempts to use Fe^0 as filter medium was

76 nonsatisfying in term of acceptable service life [9, 17]. Properly considering the mechanism of iron

77 corrosion will certainly bring some design information for the theoretical evaluation of the filter

78 service life.

79 **2.1 Aqueous iron corrosion and filter clogging**

80 Aqueous Fe^0 oxidation or immersed iron corrosion (without external current) can be regarded as a

81 series of galvanic reactions [36, 37]. At the iron/water interface, ferrous ions (Fe^{2+}) generated from

82 the Fe^0 initially form a Fe^{II} hydroxide intermediate [$Fe(OH)_2$]. In the presence of suitable oxidizing

83 agents (contaminants, MnO_2, O_2), $Fe(OH)_2$ oxidizes to an amorphous Fe^{III} oxide scale [$Fe(OH)_3$].

84 The accumulated corrosion layer restricts diffusion of dissolved oxidizing agents (including O_2),

85 allowing the formation of an inner layer of magnetite (Fe_3O_4). This magnetite layer is less

86 permeable and can effectively inhibit O_2 diffusion from the bulk solution to the bare Fe^0 [38].

87 Within this local anaerobic environment, sulfate-reducing bacteria may impact the corrosion process

88 over time. Under such conditions, a ferric layer of lepidocrocite (γ-FeOOH) can be formed and be

89 transformed to a sulfated green rust [$GR(SO_4^{2-})$]. Other identified iron corrosion products included:

90 goethite (α-FeOOH), akaganeite (β-FeOOH), and maghemite (γ-Fe_2O_3) [39-41]. In general, the

91 solution pH, the abundance of dissolved O_2 (and other oxidizing agents), the dissolved ionic species,

92 and the time-scale of the experiment can influence the nature of iron corrosion products [12].

93 The presentation above clearly shows that aqueous iron corrosion inevitably generates various iron

94 oxides which are larger in volume than iron in the metal lattice [42-45]. This volumetric expansion

95 is the fundamental cause of progressive filter fouling. Therefore, adsorption of fouling substances

96 and cake formation will not be further considered in this work as they are clearly of secondary

97 importance. On the other hand, adsorption of fouling substances can be ruled out as the Fe^0 reactive

98 zone is implemented within a fine sand layer of a conventional Biosand filter [14]. As concerning

99 cake formation, mixing Fe^0 and sand is a tool to retard or even eliminate this process.

2.2 Contaminant removal in an iron filter

101 While intentionally ignoring the interactions between Fe^0 and water, the initial filtration in an iron

102 filter is comparable to slow sand filtration (Fig. 1). As expansive corrosion products are generated,

103 the initial pore space in the filter is progressively filled and the system is gradually transformed to

104 an ultra-filtration one. Accordingly, regardless from physico-chemical interactions between

105 contaminants, iron and corrosion products, contaminant removal by pure size exclusion (or

106 adsorptive size exclusion) will inevitably occur with increasing service life. This fundamental

107 aspect has received little attention to date as the scientific community was focused on interactions

108 between selected contaminants and Fe^0. In this effort a particular attention was paid to chemical

109 transformations of the contaminants [13, 46-50].

110 The next important feature concerns the dynamic nature of iron oxide formation within the filter. It

111   has already been demonstrated that contaminants are fundamentally entrapped within the film of

112   corrosion products in the vicinity of the Fe^0 surface [25-28]. It is essential to note that the formation

113   of corrosion products is a cycle of expansion/contraction [27] occurring in the pore space. During

114   this process native iron (Fe^0: SSA < 1 m^2/g) is first transformed to voluminous iron hydroxides

115   possibly having specific surface area (SSA) > 500 m^2/g before progressively contracted to

116   amorphous and crystalline oxides with SSA ≤ 10 m^2/g. The voluminous colloid which is

117   intermediary formed [51] during an expansion/contraction cycle can be compared to a spider web

118   which traps inflowing contaminants and keeps them adsorbed while the colloid is further

119   transformed. In other words, before the pore space becomes close enough for the Fe^0 filter to act as

120   an ultra-filtration system, the expansion/contraction cycle traps contaminants from the infiltrating

121   water. It is certain, that the kinetics of iron oxidation will decrease as soon as not enough space is

122   available for expansive corrosion. This is a possible explanation for the controversial observation,

123   that TCE removal rates were higher in a system with 85 wt-% Fe^0 than they were in a 100 % Fe^0

124   system (0 % sand) [52]. In the present study substituting a portion of Fe^0 by sand (an inert material)

125   targets at retarding filter clogging.

126   **3   Retarding filter fouling**

127   Previous efforts mixing inert materials (e.g. pumice, sand) and Fe^0 in remediation Fe^0/H_2O systems

128   targeted at: (i) trapping dissolved O_2 to enable anoxic conditions in the subsequent 100 % Fe^0 zone

129   [39], (ii) retarding pore filling as inert materials will not generate new voluminous particles [39, 52],

130   (iii) controlling pH as inert materials will not contribute to pH elevation [53, 54], and (iv)

131   minimizing Fe^0 installation costs by reducing the amount of Fe^0 to be used [52, 55], In previous

132   works, it was commonplace to consider that mixing Fe^0 with sand will diminish the rates of the

133   decontamination process: (i) by decreasing the number of reactive sites, and (ii) by lowering the

134   kinetic of contaminant mass transfer to the Fe^0 surface [52, 56, 57].

135   In the present study it is acknowledged that Fe^0 filters are primarily filtrating systems (contaminant

136   removal by adsorptive size exclusion and co-precipitation). Accordingly, a Fe^0/sand filter is

137   regarded as iron assisted slow sand filtration system (or iron assisted biosand filter) [14, 17, 58, 59].

138   In such a system, contaminants are primarily removed by adsorptive size exclusion. In the Fe^0/sand

139   reactive zone, size exclusion is actively improved by: (i) the cycle of expansion/contraction

140   accompanying iron corrosion as discussed above, and (ii) iron corrosion products progressively

141   filling the pore space possibly yielding to an ultra-filtration system (and at the term to a clogged

142   system when the porosity is zero).

143   Ideally, an optimal proportion of Fe^0 in the Fe^0/sand reactive zone of a filter should exist for which

144   efficient water treatment and long-term reactivity of Fe^0 is achieved. Similarly, (i) a critical minimal

145   proportion of Fe^0 should exist beyond which filters are not significantly more efficient than biosand

146   filters, and (ii) a critical maximal proportion of Fe^0 should exist above which filter clogging will

147 rapidly occur. The possibility of adding calculated amounts of Fe^0 to assist biosand filter will be

148 discussed in the next section on the basis of an hypothetic case. Because size exclusion depends on

149 the available pore in the inter-granular space of the filter bed, the discussion will be based on the

150 volume occupied by initial particles (Fe^0 and sand). That is the volume available for in-situ

151 generated iron corrosion products (ICPs). Clearly, the volumetric proportion of Fe^0 (vol-%) in the

152 initial filter should be preferentially given. Actually the weight percent (wt-%) is routinely given [52,

153 56, 57]. However, given the large difference of specific weight (ρ in kg/m^3) between iron (7,800

154 kg/m^3) and inert materials ($< 2,700$ kg/m^3), the weight percent is not primarily appropriate.

155 **4   Discussing Fe^0:sand ratios in a filter**

156 **4.1 Design**

157 A conventional household biosand filter builds in a cylinder with 90 cm height and 30 cm width

158 (Fig. 1a) will be modified to contain a reactive zone with ≤ 3 kg Fe^0 in the fine sand layer (Fig. 1b).

159 In a conventional biosand filter the fine sand layer is about 40 to 50 cm high. [14] The aim of this

160 investigation is to introduce a reactive zone (rz) of Fe^0 with a thickness $H_{rz} \leq 10$ cm in the fine sand

161 layer.

162 The used iron fillings are spherical in shape with an average diameter of 1.2 mm. Spherical sand

163 particles of comparable particle size are used to "dilute" Fe^0. Calculations show that N = 425,093

164 Fe^0 particles are equivalent to 3 kg according to Eq. 1:

165
$$N = \frac{M}{\rho_{Fe}.4/3\pi.R_0^{3}}$$
(1)

166 where M is the mass of Fe (3 kg), ρ_{Fe} is the specific weight of Fe (7,800 kg/m^3) and R_0 is the initial

167 radius of the Fe particle (6 10^{-4} m).

168 The same number of sand particles (as quartz with specific weight: ρ_{sand} = 2,650 kg/m^3) weights

169 1.02 kg. These calculations demonstrate that mixing sand and Fe^0 in a 1:1 proportion (50 vol-%),

170 yields a weight percent of 74.6 % for Fe^0 and 25.4 % for sand. The relation between the vol-% and

171 the wt-% depends on the relative specific weights of mixed materials. A proper discussion on the

172 evolution of the porosity is necessarily based on the vol-%.

173 It is assumed that Fe_3O_4 is the sole iron corrosion product. This assumption is justified by the fact

174 that a biosand filter ideally works under anoxic conditions. Therefore, the coefficient of volumetric

175 expansion η defined as the ratio between the volume of expansive corrosion products (V_{Fe3O4}) to

176 the volume of iron in the metallic lattice (V_{Fe}) is 2.1.

177 **4.2 Rationale for the used design**

178 The used Fe^0 mass (3 kg) corresponds to the minimum iron amount that was used in the

179 three-pitcher filter in Bangladesh and Nepal [1, 58, 60]. The three-pitcher filter was very effective in

180 arsenic removal for about six weeks, afterwards a decrease in removal efficiency was observed.

181 Accordingly, the Fe^0 containing pitcher would have been replaced up to eight times per year.

182 Moreover, the iron fillings in the filter became clogged and hardened (is transformed to a "cake")

183   while treated water contained high iron concentration. On the other hand, despite effectiveness in

184   arsenic removal, the bacteriological contamination in the effluent water was sometimes high. Based

185   on these limitations, Fe^0 filters were quickly abandoned and replaced by two other devices: (i) Fe^0

186   assisted biosand filters or Kanchan Arsenic Filters (KAF) [15, 16], and (ii) the SONO Arsenic Filter

187   (SAF) [1,13]. In SAF Fe^0 is replaced by a composite iron matrix (CIM) made up of Fe^0 and

188   manganese oxides [1]. CIM has the property to sustain Fe^0 oxidation and thus in-situ produces

189   reactive iron oxides for continuous As removal. Independent researchers have reported on the

190   superiority of SAF on KAF [50]. On the other hand, first attempts to reproduce KAF in Vietnam

191   were not successful [60-62]. One major reason of failure of KAF should be that the As removal unit

192   ideally depletes dissolved oxygen necessary for effectiveness of biosand filters. The present design

193   takes into account the enumerated weakness of three-pitcher filters and KAF filters.

194   **4.3 Improved Fe^0 assisted biosand filters**

195   Contaminant removal in slow sand filters ideally occurs in an in-situ generated biofilm (so-called

196   Schmutzdecke). The Schmutzdecke is a biologically active layer consisting of algae, bacteria,

197   diatoms and zooplankton. A ripening period of 6 to 8 weeks is required for this layer to form, during

198   which time filter performance is sub-optimal [63]. Despite 130 years expertise with biosand filters,

199   complete pathogen removal can not be guaranteed [14, 17]. On the other hand, the removal of

200   chemical contaminants is not their primary target. Therefore, incorporating a Fe^0/sand zone in the

201   fine sand layer (Fig. 1) will increase the filter performance for microbial decontamination while

202   certainly remove chemical contaminants as discussed above. The next section discusses the

203   proportion of sand in the reactive zone.

204   **4.4   Proportion of Fe^0 in the reactive zone**

205   The experience with the three-pitcher filters taught that a reactive layer of 3 kg Fe^0 will be effective

206   only for some six weeks; afterwards the whole layer is transformed to a sort of "cake". As discussed

207   above replacing a portion of Fe^0 by an inert material will delay or even avoid cake formation. To

208   test the suitability of Fe^0:sand mixtures to sustain the long-term reactivity of Fe^0 assisted biosand

209   filters, evolution of the residual porosity as function of the initial percent Fe^0 in the reactive zone is

210   determined from a modeling of the filling of the pore space by iron corrosion products.

211   **4.5   Modeling the evolution of the residual porosity**

212   For granular materials, the compactness C (-) is defined as the ratio of the volume of the particles to

213   the total packing volume (V = volume of the reactive zone). Considering the granular material (sand

214   and Fe^0) as composed of mono-dispersed spheres subjected to soft vibrations, the compactness C is

215   generally considered to be equal to 0.64 for a random close packing. The virtual compactness value

216   (C = $\dfrac{\pi}{3\sqrt{2}}$ = 0.74) for mono-dispersed spheres will not be considered in this study as it is far from

217   reality [64-66]. In laboratory studies, the experimental value for compactness for each system can

218   be determined and used to discuss achieved results.

219   The initial porosity Φ_0 (-) of the reactive zone is then given by:

220         $$\Phi_0 = 1 - C \qquad (2)$$

221   The filling of the porosity by iron corrosion products can be estimated from a simplified modelling

222   (Fig. 2) based on the following assumptions:

223   (i) uniform corrosion: the diameter reduction of the particle is the same for all the Fe particles,

224   (ii) iron corrosion products are fluid enough to progressively fill available pore space,

225   (iii) the compactness C and then the initial porosity Φ_0 remain constant. The volume of the granular

226   material is not modified by the corrosion process: no pressure induced by rust formation around Fe

227   particles and no compaction of the Fe^0/sand mixture during the corrosion process.

228   With these hypotheses the residual porosity Φ can be given as a function of the volume of the iron

229   corrosion products V_{Fe3O4} (m^3) according to:

230         $$\Phi = \Phi_0 - \frac{(V_{Fe3O4} - V_{consumedFe})}{V} \qquad (3)$$

231   where $V_{Fe3O4} - V_{consumed\ Fe}$ (m^3) is the volume of the iron corrosion product without the volume of the

232   consumed Fe^0 and V (m^3) is the volume of the reactive zone.

233   The volume of the iron corrosion product without the volume of the consumed Fe^0

234    $(V_{Fe3O4} - V_{consumed\ Fe})$ is given by:

235         $$V_{Fe3O4} - V_{consumedFe} = N.(\eta-1)4/3.\pi(R_0^3 - R^3) \qquad (4)$$

236   where R (m) is the radius of the consumed iron particle and η the coefficient of volumetric

13

237   expansion.

238   The proportion of consumed iron (% consumed Fe) is given by:

239     $$\%consumedFe = 100.(\frac{R0^3 - R^3}{R0^3})$$ (5)

240   It is possible to determine the proportion of consumed iron when the volume of the iron corrosion

241   product without the volume of the consumed Fe ($V_{Fe3O4} - V_{consumed\ Fe}$) is equal to the initial porosity

242   Φ_0 (m^3) according to:

243     $$\%consumedFe(V_{Fe3O4} - V_{consumedFe} = \Phi_0) = 100.(\frac{\Phi_0}{N.(\eta-1).4/3.\pi R_0^3})$$ (6)

244   **4.6   Discussion of modeling results**

245   Calculations shown that the depth (or thickness) of a reactive zone of 425,093 spherical particles of

246   1.2 mm diameter (equivalent to 3 kg Fe^0 in a three-pitcher-like filter) is 8.5 mm vs. 30 mm (3 cm) in

247   the device discussed by Pokhrel et al. [59]. While Fe^0 is spherical here, the samples used in some

248   three-pitcher filters were commercial iron nails. Currently, granular Fe^0 is mostly available as chips

249   or fillings. Therefore, for real situations the weights (kg) of iron and sand from Table 1 should be

250   used. The calculation results are summarized in Table 1. Figure 3 shows the percent variation of the

251   residual porosity as the proportion of sand in the reactive zone of the filter varies from 0 (biosand

252   filter) to 100 % (three-pitcher filter). The results show progressive uniform decrease of the porosity

253   for all Fe^0-containing systems ($Fe^0 > 0$) yielding at the term to filter clogging for vol-% $Fe^0 > 51$.

254   The kinetics of filter clogging under atmospheric conditions depends on Fe^0 intrinsic reactivity and

255   the water chemistry. Discussing this important issue is over the scope of this communication.

256   However, it can be postulated that using a calculated amount of a Fe^0 material of known reactivity,

257   an initial biosand filter (0 % Fe^0) can be transformed to an ultra-filtration system. For example, if

258   the depletion of used Fe^0 amount could reduce the initial porosity to 70 %. Such a filter should

259   initially contain less than 51 vol-% Fe^0. Contaminant removal is due to two important processes (i)

260   size exclusion and species trapping during expansion/contraction cycles coupled with iron oxidation

261   within the pore space (before Fe^0 depletion) and (ii) pure size exclusion upon Fe^0 depletion.

262   An important feature from Figure 3 (also see Tab. 1) is that Fe^0 depletion could only occur for $0 <$

263   m_{Fe} (kg) ≤ 1.53. This corresponds to the mass of iron which corrosion yields complete fouling of the

264   pore space. This result reveals that in a conventional three-pitcher-like filter up to 1.47 kg (from 3

265   kg) may be superfluous. It can be anticipated that the amount of really superfluous Fe^0 depends on

266   the intrinsic reactivity of the material. In all the cases, bringing more than 70 vol-% Fe^0 in the

267   reactive zone could be regarded as pure resource wastage. Furthermore, too high Fe^0 proportions

268   yield to more rapid clogging (or short filter service live) as uniform corrosion produces more Fe

269   oxides within the pore space (see below).

270   Figure 3 also shows the evolution of consumed and non-consumed Fe^0 as function of the initial

271   volumetric proportion of Fe^0 in the reactive zone. It can be seen that Fe^0 depletion (100 %

272   consumption) can occur only in systems with up to 51 vol-% iron. It appears that there are two types

273   of reactive zone: (i) type 1: Fe^0 > vol-51 %, the pore space is totally filled with iron corrosion

274   products but iron is not depleted, and (ii) type 2: Fe^0 < 51 vol-%, iron is depleted but the pore space

275   is not totally filled by iron corrosion products. Depending on the reactivity of Fe^0, it can be

276   postulated that filters with 30 to 55 vol-% Fe^0 may be very efficient for safe water production. The

277   optimal proportion in filters has to be ascertained in feasibility laboratory studies.

278   Recently, Gottinger [67] reported on a very efficient system containing only 30 vol-% Fe^0 for

279   efficient As removal in a pilot plant for a small community in Saskatchewan/Canada. The results of

280   Gottinger [67] could be regarded as the first validation of the conceptual approach presented here.

281   Gottinger's design was based on primary calculations considered as a rough estimate by the authors

282   [68, 69]. The present work has improved the calculus of Leupin et al. [68] in taking into account the

283   inter-granular voids in the Fe^0/sand bed. The results show that simple Fe^0/sand filters (3-Kolshi

284   filters, KAF) have been abandoned because of lack of system understanding. The theoretical

285   calculations of the present study and the encouraging results of Gottinger [67] suggest that the huge

286   potential of Fe^0/sand filters is yet to be exploited.

287   **4.7   Expanding the reactive zone**

288   The presentation above has shown two important features: (i) a good Fe^0 filter may contain only

289   about 40 vol-% Fe^0 mixed with sand; (ii) with 3 kg Fe^0 a reactive zone is only 8.5 mm high.

290   Realizing such a thin homogeneous reactive zone with a mixture of Fe^0 and sand necessarily

291   requires particular experimental aptitudes and could be a laborious task. On the other hand, a filter

292   with only less than 1.50 kg Fe^0 may be of limited service life. Therefore, one could keep the Fe^0

293   mass constant to 3 kg and vary the mass of sand to achieve various percent of Fe^0 in the reactive

294   zone. In this effort, it should be kept in mind, that the reactive zone should ideally not exceed 10 cm

295   (1/4 to 1/5 of the fine sand layer). On the other hand the calculations above have shown that Fe^0

296   depletion occurs for too low Fe^0 proportions. In such cases, the desired enhancement of the biosand

297   filter performance could remain unsatisfactorily. Based on these considerations this discussion was

298   limited to $Fe^0 > 10\%$.

299   Table 2 shows that the result of dilution 3 kg Fe^0 with various amounts of sand is a thicker reactive

300   zone (up to 8.5 cm). The relative porosity Φ/Φ_0 versus the consumed iron proportion (% consumed

301   Fe) is shown in Table 2, Figure 3, and Figure 4. As discussed above, the trend is the same as for H_{rz}

302   = 8.5 mm. A similar identity is obtained for the evolution of the relative porosity Φ/Φ_0 (Fig 4).

303   Figure 3b gives a more specific image of both systems based on the evolution of the excess Fe^0

304   mass.

305   Figure 3b shows the evolution of a conventional three-pitcher filter (point A) as: (i) the thickness of

306   the reactive zone increases from 0.85 to 8.5 cm (line ABC), and (ii) the proportion of iron varies

307   from 100 to 10 vol-% in 0.85 cm reactive zone (line ADE). The line AFG corresponds to the

308   evolution of the excess Fe^0 (% non consumed) in the reactive zone with 3 kg Fe^0. Point G (0 % Fe^0)

309   corresponds to a biosand filter. The line BDF corresponds to filter clogging (porosity equal to zero).

310   The major difference between both investigated systems (constant and variable reactive zones) is

311   reflected in the segment AB and AD. From D to A no additional Fe^0 consumption occurs whereas

312   from B to A, a progressive decrease of Fe^0 consumption is observed. The most important feature

313   from Figure 3b is that the filter corresponding to point A is the system with the largest iron excess

314   (line AF). Accordingly, the pore space is filled very quickly and the service life is very short.

315   Therefore, the three-pitcher is the worst possible Fe^0 filter. A better Fe^0 consumption could be

316   achieved by reducing the proportion of Fe^0 (ADE) or augmenting the proportion of sand (ABC).

317   While decreasing the Fe^0 proportion at constant reactive zone thickness, Fe^0 depletion was achieved

318   for systems > 51 vol-% Fe^0. A similar result was achieved by extending the thickness of the reactive

319   zone. Extending the reactive zone is a better way to increase filter efficiency because more material

320   is consumed. More Fe^0 consumption is directly related to more iron corrosion products for

321   contaminant removal and improved size exclusion.

322   In the case of extending the reactive zone, the two types of reactive zones described for a constant

323   (8.5 mm) reactive zone are reproduced: (i) Fe^0 > vol-51 %, filter clogging occurs without Fe^0

324   depletion; and (ii) Fe^0 < 51 %, iron depletion occurs and no filter clogging is observed. Thus, a filter

325   concealing long-term reactivity and long term permeability will contain about 50 % Fe^0. In this case,

326   3.0 kg Fe^0 are used compared to 1.50 kg Fe^0 in the other case (see Tab. 1 and Tab.2). Although the

327   actual kinetics of filter clogging depend on several factors, this principle calculation clearly showed

328   that a reactive zone of 100 % Fe^0 (conventional three-pitcher filter) is pure material wastage. In

329   other words, mixing inert material to reactive Fe^0 is a prerequisite for efficient iron filters.

330   The calculations above for two cases ($m_{Fe} \leq 3$ kg and $m_{Fe} = 3$ kg) demonstrate the feasibility of

331   delaying filter clogging by rationally choosing the proportion of Fe^0 in the filter. For each specific

332   couple (Fe^0, contaminated water) researchers will have to identify the optimal Fe^0 proportion in

333   their filters as the kinetics of filter clogging depend both on (i) material intrinsic reactivity

334   (susceptibility to corrosion) and on (ii) the aggressiveness of contaminated water. Additionally, the

335   specific contribution of contaminants and other dissolves species (including molecular size) to pore

336   fouling should be taken into account.

337   **5   Concluding remarks**

338   In this communication, the perspective regarding Fe^0/H_2O systems as assisted slow sand filtration

339   systems [15, 16] is rationalized and theoretically optimized. Accordingly, a Fe^0/H_2O system is a

340   slow sand filtration system which efficiency is improved by addition of a calculated amount of

341   metallic iron (Fe^0). The aqueous corrosion by infiltrating raw water should ideally transform the

342   initial Fe^0/H_2O filter to an ultra-filtration system. Improved efficiency is based on the volumetric

343   expansive nature of iron corrosion which should partly fill initial pore space in the biosand-like

344 filter. Calculations demonstrate improved efficient and extended service life while using about 40

345 vol-% of Fe^0 mixed with sand.

346 To warrant quantitative contaminant removal in the system in the initial stage of the operation, the

347 entrance zone or a separate unit could contain very reactive powdered Fe^0 material disseminated in

348 sand. The major output for this theoretical study is that very efficiency filters could be constructed

349 by very simple and low cost materials (Fe^0 and sand). Intensive research is needed to realize this

350 noble goal. The fate of this concept will depend on the capacity of researchers to properly

351 characterize the reactivity of Fe^0 materials and select appropriate ones for household filters. Results

352 will be up-scalable to field Fe^0 permeable barriers. Two simple methods for material selection were

353 recently introduced [70-72].

354 Finally, it should be explicitly said that the approach considering remediation Fe^0/H_2O systems as

355 primary filtration systems is not negating the complex chemical and physical processes (adsorption,

356 co-precipitation, desorption, oxidation, reduction) occurring in it. On the contrary, this approach

357 takes into account the fundamental fact that volumetric expansive iron corrosion needs free space to

358 occur optimally and explains why so many species have been successfully removed in systems

359 designed for individual compounds [1, 13, 50, 73, 74]. For example, Johnson et al. [74] reported

360 decrease in sulfate, carbonate, and calcium concentration in a barrier designed to remove explosives

361 from contaminated groundwater. Constructing efficient Fe^0/sand filters is an engineering challenge

362   for the water treatment community.

363   **Acknowledgments**

365   **References**

366   [1] S. Ahamed, A.K.M. Munir, A. Hussam, Groundwater arsenic removal technologies based on

367   sorbents: Field applications and sustainability. In Handbook of Water Quality and Water Purity,

368   Elsevier Inc. **2009**, Chapter 16, 379.

369   [2] J.M. Arnal, B. Garcia-Fayos, G. Verdu, J. Lora, Ultrafiltration as an alternative membrane

370   technology to obtain safe drinking water from surface water: 10 years of experience on the

371   scope of the AQUAPOT project *Desalination* **2009**, 248 (1-3), 34.

372   [3] E. Awuah, R.T. Morris, P.A. Owusu, R. Sundell, J. Lindstrom, Evaluation of simple methods of

373   arsenic removal from domestic water supplies in rural communities. *Desalination* **2009**, 248

374   (1-3), 42.

375   [4] L.J. Banasiak, A.I. Schäfer, Removal of inorganic trace contaminants by electrodialysis in a

376   remote Australian community. *Desalination* **2009**, 248 (1-3), 48.

377   [5] A. Gadgil, Drinking water in developing countries. *Annu. Rev. Energy Environ.* **1998**, 23, 253.

378   [6] L. Groendijk, H.E. de Vries, Development of a mobile water maker, a sustainable way to

379   produce safe drinking water in developing countries. *Desalination* **2009**, 248 (1-3), 106.

380 [7] C. Hårleman, G. Jacks, B. Rybeck, The use of a clinoptilolite-based filter in emergency

381   situations. *Desalination* **2009**, 248 (1-3), 629.

382 [8] P.R. Hunter, Household water treatment in developing countries: comparing different

383   intervention types using meta-regression. *Environ. Sci. Technol*. **2009,** 43 (23), 8991.

384 [9] A.H. Malik, Z.M. Khan, S. Mahmood, S. Nasreen, Z.A. Bhatti, Perspectives of low cost arsenic

385   remediation of drinking water in Pakistan and other countries. *J. Hazard. Mater*. **2009,** 168 (1),

386   1.

387 [10] W.-P. Schmidt, S. Cairncross, Household water treatment in poor populations: Is there enough

388   evidence for scaling up now? *Environ. Sci. Technol.* **2009**, 43 (14), 986.

389 [11] W.-P. Schmidt, S. Cairncross, Response to comment on "Household water treatment in poor

390   populations: Is there enough evidence for scaling up now?" *Environ. Sci. Technol*. **2009**, 43

391   (14), 5545.

392 [12] T.C. Timmes, H.-C. Kim, B.A. Dempsey, Electrocoagulation pretreatment of seawater prior to

393   ultrafiltration: Pilot-scale applications for military water purification systems. *Desalination*

394   **2010**, 250 (1), 6.

395 [13] A. Hussam, A.K.M. Munir, A simple and effective arsenic filter based on composite iron matrix:

396   Development and deployment studies for groundwater of Bangladesh. *J. Environ. Sci. Health*

397   *A* **2007**, 42, 1869.

398 [14] Lea, M. Biological sand filters: low-cost bioremediation technique for production of clean

399   drinking water. *Curr. Prot. Microbiol*. **2008,** 1G.1.1-1G.1.28.

400 [15] T.K.K. Ngai, S. Murcott,, R.R. Shrestha, B. Dangol, M. Maharjan, Development and

401   dissemination of Kanchan™ Arsenic Filter in rural Nepal. *Water Sci. Technol. Water Supply*

402   **2006**, 6, 137.

403 [16] T.K.K. Ngai, R.R. Shrestha, B. Dangol, M. Maharjan, S.E. Murcott, Design for sustainable

404   development – Household drinking water filter for arsenic and pathogen treatment in Nepal. *J.*

405   *Environ. Sci. Health A* **2007**, 42, 1879.

406 [17] C. Noubactep, A. Schöner, P. Woafo, Metallic iron filters for universal access to safe drinking

407   water. *Clean* **2009**, 37 (12), 930.

408 [18] S. Madaeni, The application of membrane technology for water disinfection. *Water Res*. **1999**,

409   33 (2), 301.

410 [19] J.G. Minnery, J.G. Jacangelo, L.I. Boden, D.J. Vorhees, W. Heiger-Bernays, Sensitivity analysis

411   of the pressure-based direct integrity test for membranes used in drinking water treatment.

412   *Environ. Sci. Technol*. **2009**, 43 (24), 9419.

413 [20] M. Peter-Varbanets, C. Zurbrügg, C. Swartz, W. Pronk, Decentralized systems for potable

414   water and the potential of membrane technology. *Water Res*. **2009**, 43 (2), 245.

415 [21] X.-m. Wang, T.D. Waite, Role of gelling soluble and colloidal microbial products in membrane

416   fouling. *Environ. Sci. Technol*. **2009**, 43 (24), 9341.

417   [22] W. Yang, N. Cicek, J. Ilg, State-of-the-art of membrane bioreactors: Worldwide research and

418   commercial applications in North America. *J. Membrane Sci*. **2006**, 270, 201.

419   [23] R. Meierhofer, G. Landolt, Factors supporting the sustained use of solar water disinfection –

420   Experiences from a global promotion and dissemination programme. *Desalination* **2009**, 248

421   (1-3), 144.

422   [24] WHO. Guidelines for Drinking-water Quality, Recommendations, 3rd ed.; World Health

423   Organization: Geneva, 2006; Vol. 1.

424   [25] C. Noubactep, Processes of contaminant removal in "Fe^0–H$_2$O" systems revisited. The

425   importance of co-precipitation. *Open Environ. J.* **2007**, 1 (1), 9.

426   [26] C. Noubactep, A critical review on the mechanism of contaminant removal in Fe^0–H$_2$O

427   systems. *Environ. Technol.* **2008**, 29 (8), 909.

428   [27] C. Noubactep, The suitability of metallic iron for environmental remediation. *Environ. Progr*.

429   **2010**, doi: 10.1002/ep.10406.

430   [28] C. Noubactep, A. Schöner, M. Sauter, *Significance of oxide-film in discussing the mechanism of*

431   *contaminant removal by elemental iron materials*. In "Photo-Electrochemistry &

432   Photo-Biology for the Sustainablity"; S. Kaneco, B. Viswanathan, H. Katsumata (Eds.),

433   Bentham Science Publishers, **2010**, 1, 34.

434   [29] C. Noubactep, Better drinking water for rural regions of Africa. (in German) In Afrika &

435   Wissenschaft **2008,** 1, 36.

436   [30] C. Noubactep, P. Woafo, Elemental iron (Fe^0) for better drinking water in rural areas of

437   developing countries. In Merkel B.J., Hasche-Berger A. (Eds.) *Uranium in the Environment*.

438   Springer, Berlin, Heidelberg **2008**, 121.

439   [31] Y. You, J. Han, P.C. Chiu, Y. Jin, Removal and inactivation of waterborne viruses using

440   zerovalent iron. *Environ. Sci. Technol.* **2005**, 39 (23), 9263.

441   [32] L. Li, C.H. Benson, E.M. Lawson, Impact of mineral fouling on hydraulic behavior of

442   permeable reactive barriers. *Ground Water* **2005**, 43 (4), 582.

443   [33] L. Li, C.H. Benson, E.M. Lawson, Modeling porosity reductions caused by mineral fouling in

444   continuous-wall permeable reactive barriers. J. Contam. Hydrol. **2006**, 83 (1-2), 89.

445   [34] R.E. Arndt, E.J. Wagner, Rapid and slow sand filtration techniques and their efficacy at

446   filtering triactinomyxons of Myxobolus Cerebralis from contaminated water. *North Am. J.*

447   *Aquacul.* **2004**, 66, 261.

448   [35] N.C. Thanh, S. Vigneswaran, C. Polprasert, Optimization of dual-media filter performance. *J.*

449   *Am. Water Res. Ass.* **1979**, 15, 1459.

450   [36] H. Klas, H. Steinrath, *Iron corrosion and its abatement*. (in German) Stahleisen, Düsseldorf

451   **1974**, 632 pp.

452 [37] C. Noubactep, A. Schöner, Metallic iron for environmental remediation: Learning from

453 electrocoagulation. *J. Hazard. Mater*. **2010**, 175 (1-3), 1075.

454 [38] M. Stratmann, J. Müller, The mechanism of the oxygen reduction on rust-covered metal

455 substrates. *Corros. Sci*. **1994**, 36 (2), 327.

456 [39] P.D. Mackenzie, P.D. Horney, T.M. Sivavec, Mineral precipitation and porosity losses in

457 granular iron columns. *J. Hazard. Mater*. **1999**, 68 (1-2), 1.

458 [40] D.H. Phillips, D.B. Watson, Y. Roh, B. Gu, Mineralogical characteristics and transformations

459 during long-term operation of a zerovalent iron reactive barrier, *J. Environ. Qual*. **2003**, 32 (6),

460 2033.

461 [41] Y. Roh, S.Y. Lee, M.P. Elless, Characterization of corrosion products in the permeable reactive

462 barriers. *Environ. Geol*. **2000**, 40 (1-2), 184.

463 [42] K. Bhargava, A.K. Ghosh, Y. Mori, S. Ramanujam, Model for cover cracking due to rebar

464 corrosion in RC structures. *Eng. Struct*. **2006**, 28 (8), 1093.

465 [43] S. Caré, Q.T. Nguyen, V. L'Hostis, Y. Berthaud, Mechanical properties of the rust layer induced

466 by impressed current method in reinforced mortar. *Cement Concrete Res*. **2008**, 38 (8-9),

467 1079.

468 [44] S. Caré, Q.T. Nguyen, K. Beddiar, Y. Berthaud, Times to cracking in reinforced mortar beams

469 subjected to accelerated corrosion tests. *Mater. Struct*. **2010**, 43 (1-2), 107.

470   [45] T. El. Maaddawy, K. Soudki, A model for prediction of time from corrosion initiation to

471   corrosion cracking. *Cem. Concr. Compos*. **2007**, 29 (3), 168.

472   [46] P. Bayer, M. Finkel, Life cycle assessment of active and passive groundwater remediation

473   technologies. *J. Contam. Hydrol*. **2006**, 83 (3-4), 171.

474   [47] A.D. Henderson, A.H. Demond, Long-term performance of zero-valent iron permeable reactive

475   barriers: A critical review. *Environ. Eng. Sci*. **2007**, 24 (4), 401.

476   [48] M.R. Higgins, T.M. Olson, Life-cycle case study comparison of permeable reactive barrier

477   versus pump-and-treat remediation. *Environ. Sci. Technol.* **2009**, 43 (24), 9432.

478   [49] M.M. Scherer, S. Richter, R.L. Valentine, P.J.J. Alvarez, Chemistry and microbiology of

479   permeable reactive barriers for in situ groundwater clean up. *Rev. Environ. Sci. Technol*. **2000**,

480   30 (3), 363.

481   [50] Md. Shafiquzzaman, Md.S. Azam, I. Mishima, J. Nakajima, Technical and social evaluation of

482   arsenic mitigation in rural Bangladesh. *J. Health Popul. Nutr*. **2009**, 27, 674.

483   [51] L. Livage, J. Lemerle, Transition metal oxide gels and colloids. *Ann. Rev. Mater. Sci*. **1982**, 12,

484   103.

485   [52] E. Bi, J.F. Devlin, B. Huang, Effects of mixing granular iron with sand on the kinetics of

486   trichloroethylene reduction. *Ground Water Monit. Remed*. **2009**, 29 (2), 56.

487   [53] D.R. Burris, T.J. Campbell, V.S. Manoranjan, Sorption of trichloroethylene and

488   tetrachloroethylene in a batch reactive metallic iron-water system. *Environ. Sci. Technol*. **1995**,

489   29 (11), 2850.

490   [54] Y.X. Wu, L.D. Slater, N. Korte, Effect of precipitation on low frequency electrical properties of

491   zerovalent iron columns. *Environ. Sci. Technol.* **2005**, 39 (23), 9197.

492   [55] R.M. Powell, R.W. Puls, Proton generation by dissolution of intrinsic or augmented

493   aluminosilicate minerals for in situ contaminant remediation by zero-valence-state iron.

494   *Environ. Sci. Technol*. **1997**, 31 (8), 2244.

495   [56] J.F. Devlin, J. Patchen, The effect of diluting granular iron with a non-reactive porous medium

496   on contaminant transformation rates. 5th Joint Conference of the IAH-CNC and the Canadian

497   Geotechnical Society (CGS), October 24–27 (**2004**), Quebec City.

498   [57] S.F. O'Hannesin, R.W. Gillham, Long-term performance of an in situ "iron wall" for

499   remediation of VOCs. *Ground Water* **1998**, 36 (1), 164.

500   [58] A. Neku, N. Tandulkar, An overview of arsenic contamination in groundwater of Nepal and its

501   removal at household level. *J. Phys. IV* **2003**, 107, 941.

502   [59] D. Pokhrel, B.S. Bhandari, T. Viraraghavan, Arsenic contamination of groundwater in the Terai

503   region of Nepal: An overview of health concerns and treatment options. *Environ. Int*. **2009**, 35,

504   157.

505   [60] H. Chiew, M.L. Sampson, S. Huch, S. Ken, B.C. Bostick, Effect of groundwater iron and

phosphate on the efficacy of arsenic removal by iron-amended biosand filters. *Environ. Sci.*

Technol. **2009**, 43 (16), 6295.

[61] C. Noubactep, Comments on "Effect of groundwater iron and phosphate on the efficacy of

arsenic removal by iron-amended BioSand Filters". *Environ. Sci. Technol.* **2009**, 43 (22),

8698.

[62] S. Tuladhar, L.S. Smith, SONO filter: An excellent technology for save water in Nepal.

SOPHEN **2009,** 7, 18.

[63] E.D. Joubert, B. Pillay, Visualisation of the microbial colonisation of a slow sand filter using an

environmental scanning electron microscope. *Elect. J. Biotechnol.* **2008**; April 15, (ISSN:

0717-3458).

[64] D.J. Cumberland, R.J. Crawford, *Handbook of powder technology. The packing of particle* Vol.

6, J.C. Williams & T. Allen, Elsiever, Amsterdam (**1987**).

[65] F. de Larrard, *Granular structure and formulation of concretes* (in French), Etudes et

recherches des Laboratoires des ponts et chaussées 34, Laboratoire central des ponts et

chaussées, Paris (**1999**), 414 pp.

[66] A. Nur, G. Mavko, J. Dvorkin, D. Galmudi, Critical porosity; a key to relating physical

properties to porosity in rocks. *The Leading Edge* **1998**, 17 (March 1998), 357.

[67] A.M. Gottinger *Chemical-free arsenic removal from potable water with a ZVI-amended*

524   *biofilter*. Master thesis, University of Regina (Saskatchewan, Canada) **2010**, 90 pp.

525   [68] O.X. Leupin, S.J. Hug, Oxidation and removal of arsenic (III) from aerated groundwater by

526   filtration through sand and zero-valent iron. *Wat. Res.* **2005**, 39 (9), 1729.

527   [69] O.X. Leupin, S.J. Hug, A.B.M. Badruzzaman, Arsenic removal from Bangladesh tube well

528   water with filter columns containing zerovalent iron filings and sand. *Environ. Sci. Technol.*

529   **2005** 39 (20), 8032.

530   [70] C. Noubactep, G. Meinrath, P. Dietrich, M. Sauter, B. Merkel, Testing the suitability of

531   zerovalent iron materials for reactive walls. *Environ. Chem.* **2005**, 2 (1), 71.

532   [71] C. Noubactep, T. Licha, T.B. Scott, M. Fall, M. Sauter, Exploring the influence of operational

533   parameters on the reactivity of elemental iron materials. *J. Hazard. Mater*. **2009**, 172 (2-3),

534   943.

535   [72] C. Noubactep, Characterizing the reactivity of metallic iron upon methylene blue discoloration

536   in Fe^0/MnO_2/H_2O systems. J. Hazard. Mater. **2009**, 168 (2-3), 1613.

537   [73] R.L. Johnson, R.B. Thoms, R.O'B. Johnson, T. Krug, Field evidence for flow reduction through

538   a zero-valent iron permeable reactive barrier. *Ground Water Monit. Remed*. **2008**, 28 (3), 47.

539   [74] R.L. Johnson, R.B. Thoms, R.O'B. Johnson, J.T. Nurmi, P.G. Tratnyek, Mineral precipitation

540   upgradient from a zero-valent iron permeable reactive barrier. *Ground Water Monit. Remed*.

541   **2008**, 28 (3), 56.

542   30

542   **Table 1**: Variation of the residual porosity (Φ/Φ_0) and Fe^0 consumption ($[Fe]_\infty$) for long-term

543   operation as function of the initial iron proportion ($[Fe]_0$) in a reactive zone with 425,093

544   particles with 1.2 mm diameter. The compactness of the granular material is considered

545   to be C = 0.64 (see text). The thickness of the reactive zone is 0.85 cm (8.5 mm). Ideally,

546   iron depletion occurs for $[Fe]_0 < 51$ vol-% without filter clogging. On the contrary, filter

547   clogging occurs without material depletion for $[Fe]_0 > 51$ vol-%. This results suggest that

548   using filter with $[Fe]_0 > 70$ vol-% is pure material wastage.

549

$[Fe]_0$	$[Fe]_0$	$[sand]_0$	Φ/Φ_0	$[Fe]_\infty$	$[Fe]_\infty$
(vol-%)	(kg)	(kg)	(-)	(wt-%)	(kg)
0.0	0.0	1.02	100	/	/
10.0	0.30	0.92	80.4	100.0	0.30
20.0	0.60	0.82	60.9	100.0	0.60
30.0	0.90	0.71	41.3	100.0	0.90
40.0	1.20	0.61	21.8	100.0	1.20
50.0	1.50	0.51	2.2	100.0	1.50
51.1	1.53	0.50	0.0	100.0	1.53
60.0	1.80	0.41	0.0	85.2	1.53
70.0	2.10	0.31	0.0	73.0	1.53
80.0	2.40	0.20	0.0	63.9	1.53
90.0	2.70	0.10	0.0	56.8	1.53
100.0	3.00	0.00	0.0	51.1	1.53

550

551

551 **Table 2:** Composition and thickness (H_{rz}) of the reactive zone for 3 kg of Fe^0. Fe^0 and sand particles

552 are 1.2 mm in diameter. The value $C = 0.64$ is considered for compactness (see text).

553

$[Fe]_0$	$[Fe]_0$	$[sand]_0$	H_{rz}	Φ/Φ_0	$[Fe]_\infty$	$[Fe]_\infty$
(vol-%)	(kg)	(kg)	(cm)	(-)	(wt-%)	(kg)
10.0	3.00	9.17	8.50	80.4	100.0	3.00
20.0	3.00	4.08	4.25	60.9	100.0	3.00
30.0	3.00	2.38	2.83	41.3	100.0	3.00
40.0	3.00	1.53	2.13	21.8	100.0	3.00
50.0	3.00	1.02	1.70	2.2	100.0	3.00
51.1	3.00	0.97	1.66	0.0	100.0	3.00
60.0	3.00	0.68	1.42	0.0	85.2	2.56
70.0	3.00	0.44	1.21	0.0	73.1	2.19
80.0	3.00	0.25	1.06	0.0	63.9	1.91
90.0	3.00	0.11	0.94	0.0	56.8	1.70
100.0	3.00	0.00	0.85	0.0	51.1	1.53

554

555

555 **Figure captions**

556 **Fig. 1:** Schematic diagrams of (a) a common BioSand filter and (b) an iron-reactive-zone containing

557 BioSand filter. The illustration highlights major principles and generic size dimensions. Modified

558 after ref. [14]. The thickness of the reactive layer containing up to 3 kg Fe^0 discussed in the text

559 varies from 8.5 to 42.5 mm (< 5 cm).

560 **Figure 2**: Reactive zone of volumetric ratio Fe^0:sand = 1:1 at the initial date (t_0) and at $t \gg t_0$. At

561 any time $> t_0$ the pore space is filled of by iron corrosion products (rust). As iron corrosion proceeds

562 the pore space is progressively filled by rust. The net result is a porosity loss yielding filter clogging

563 at the term. Whether and when filter clogging occurs depends on the proportion of Fe^0 in the

564 Fe^0/sand system (see text).

565 **Figure 3**: Variation of the residual porosity (%), the proportion (%) and amount (g) of consumed

566 and non-consumed (excess) Fe^0 as function of the initial volumetric proportion of Fe^0 in the

567 Fe^0/sand filters. In percent (%), the same trend is obtained in both systems for all parameters (a).

568 However, there is a net difference in the mass of consumed Fe^0 (b). It is clear from Fig. 3b that in

569 the three-pitcher filter (point A, see text), iron consumption is minimal (shortest service life). The

570 lines are not fitting functions; they simply connect points to facilitate visualization.

571 **Figure 4:** Evolution of the relative porosity ($\Phi/\Phi o$) of the reactive zone as function of the percent

572 Fe^0 consumption for several Fe^0/sand mixtures. Again the same trend is observed for (i) a 8.5 mm

573 reactive zone initially containing 0 to 3 kg Fe^0 and (ii) a 8.5 to 42.5 mm reactive zone initially

574 containing 3 kg Fe^0. It can be seen that for 51 vol-% iron (3 kg) and 49 vol-% sand (0.98 kg), the

575 pore space is filled at iron depletion. For Fe^0 < 51 vol-%, no filter clogging will be observed, for Fe^0

576 > 51 vol-% no material depletion could occur. This observation attests that a reactive zone of 100 %

577 Fe^0 is pure material wastage (see text).