1 **Designing iron-amended biosand filters for decentralized safe drinking water provision**

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9 **Abstract**

10 There are ongoing efforts to render conventional biosand filters (BSF) more efficient for safe

11 drinking water provision. One promising option is to amend BSF with a reactive layer

12 containing metallic iron (Fe^0). The present communication presents some conceptual options

13 for efficient Fe^0-amended BSF in its fourth generation. It is shown that a second fine-sand

14 layer should be placed downwards from the Fe^0-reactive layer to capture dissolved Fe. This

15 second fine-sand layer could advantageously contain adsorbing materials (e.g. activated

16 carbons, wooden charcoals). An approach for sizing the Fe^0-reactive layer is suggested based

17 on 3 kg Fe^0 per filter. Working with the same Fe^0 load will ease comparison of results with

18 different materials and the scaling up of household BSF to large scale community slow sand

19 filters (SSF).

20 **Key words**: Biosand filter, Iron/sand filter; Point of use, Drinking water, Zerovalent iron.

21 **Acronym List**

22 BSF   Biosand Filter

23 RZ   Reactive Zone

24 SSF   Slow Sand Filtration

25 WHO   World Health Organization

26

1   Introduction

Safe drinking water may derive from surface water or groundwater. Surface water is often polluted with pathogens (e.g. bacteria, viruses). Groundwater is mainly contaminated/polluted by inorganic species (e.g. arsenic, iron, nitrate, uranium). Both surface water and groundwater may be turbid (physical pollution) and contain organic and inorganic contaminants from both natural and anthropogenic origins [1-5]. Accordingly, at any location available water may contain biological, chemical and physical contamination. The World Health Organization (WHO) guidelines for drinking water quality [6] outline a preventive management framework for safe drinking water [2,7-9]. The WHO guidelines [6] entail: (i) health based targets, (ii) system assessment from source through treatment to the point of consumption, (iii) operational monitoring of the control measures in the drinking water production, (iv) management plans documenting the system assessment and monitoring plans and (v) a system of independent surveillance that verifies that the above are operating properly.

Whenever a water is polluted, e.g. after the WHO Guidelines [6], it should be rendered safe before consumption. Universal appropriate treatment technologies should be able to efficiently remove all three classes of contamination. For rural and peri-urban areas in the developing world, the treatment system should ideally occur in a single-stage filtration process at household or small community level [2,7,10-23]. From the available technologies, household biosand filters (intermittent slow sand filters) have been tested the most [2,24-36]. Currently, at least 500,000 people are using biosand filters (BSF) to provide safe drinking water [28,30,32]. However, it has been traceably shown that BSF do not remove all of the pathogens from water. Accordingly, several attempts are tested to improve the efficiency of conventional BSF [14,16,20,27,28,37-45]. For example, Baig et al. [16] used locally available biomass in their innovative biosand filter and reported on significant efficiency enhancement. On the other hand, amending BSF with metallic iron (Fe^0) has been proven beneficial for water treatment [27,28,38,39.40,45,46]. However, as with most innovations, the early

52   development of Fe^0-amended BSF is marked by empirical designs [32]. Therefore, the Fe^0-

53   amended BSF technology should now be translated into rational engineering design criteria.

54   The present communication is a part of a series of theoretical works based on the aqueous

55   chemistry of iron corrosion and aiming at easing research on water treatment using Fe^0 [47-

56   51]. The expansive nature of Fe^0 oxidative dissolution followed by precipitation of Fe^{II}/Fe^{III}

57   species (oxides and hydroxides) at pH > 4.5 [52,53] is properly considered. Much of the

58   impetus for this work has come from the work of Noubactep et al. [12], who have proposed (i)

59   Fe^0 as a universal agent for save drinking water provision, and (ii) to amend BSF with a

60   reactive zone made up of Fe^0 and inert materials (e.g. sand). Subsequent works have

61   rationalized the mixture of Fe^0 with (i) inert materials [47,54], and (ii) reactive but non

62   expansive materials [51] for long-term water treatment. A key question that remains is how to

63   avoid that dissolved iron is present in the treated water? An analysis of published data from

64   Khan et al. [37] to Ingram et al. [46] suggests that a fine sand layer must be placed

65   downstream from the reactive zone containing Fe^0. Based on this observation several design

66   options are discussed. For the sake of clarity a conventional BSF will be first presented.

67   **2   Biosand filter (BSF),**

68   The BSF was developed at the University of Calgary in the early 1990s by Dr. Manz [27,28].

69   The BSF is a downscaling of the slow sand filtration (SSF) technology [55] for intermittent

70   water filtration at household level. The BSF has been reported to be efficient at removing

71   chemicals (e.g. iron, manganese, and sulphur), pathogens (e.g. bacteria, viruses) and turbidity

72   from low turbid water [27-30]. Unlike a 'simple' slow sand filter, the BSF has the ability to

73   perform multiple functions as a single unit [4]. In a BSF, settlement, straining and filtration

74   act in synergy to remove biological, chemical and physical contamination/pollution. Ideally,

75   safe drinking water is produced.

76   The most important process in a BSF occurs in a biological layer (biofilm) called the

77   *Shmutzdecke* [4,12,33,34,57]. The *Shmutzdecke* develops in the presence of atmospheric

3

78 oxygen on the surface of the uppermost layer of sand that is responsible for the removal of

79 microorganisms. Ideally, the fine sand layer downwards from the *Shmutzdecke* is almost

80 anoxic (O_2 free) as O_2 is consumed by microbial activity within the *Shmutzdecke*. It should be

81 anticipated here that water exiting such an anoxic system is ideal for a Fe^0 filter (reactive

82 zone) as volumetric expansion is mostly limited to the formation of Fe_3O_4 with an expansion

83 coefficient (η) of 2.08 [52,53]. Under oxic conditions more volumetric expansive iron oxides

84 and hydroxides formed ($\eta \leq 6.40$). As a rule, the larger the η value, the more rapid the loss of

85 hydraulic conductibility (permeability loss) of the reactive zone.

86 **2.1   Design of a BFS**

87 The engineering principles of household BSF is described in several recent communications

88 [2,4,27-30,32,56-59]. All household BSF share five basic design components [56]: (i) a good

89 source of sand and gravel, (ii) a bucket filled with 40 to 75 cm of fine sand (sand bed), (iii) a

90 layer of static standing water (supernatant water), (iv) a maturation time of 14 to 21 days for

91 formation of the *Shmutzdecke*, and (v) a flow control system. Ideally, water should flow freely

92 from the filter (not tap).

93 Intensive research is still targeted at designing smaller, lighter (portable), less expensive but

94 efficient BSF [4,60]. Alternative BSF designs for better efficiency in aqueous contaminant

95 removal have been tested [2,4,45,46,58,59]. Tested design parameters include: (i) sand type,

96 sand size and sand depth [31,32,61], (ii) maturation time or time to formation of the

97 *Shmutzdecke*, (iii) elevation head, (iv) standing water depth, (v) filter pause time or daily

98 water throughput, as well as (vi) amendment of conventional BSF with reactive layers

99 containing metallic iron (Fe^0). The present communication is focused on the design of Fe^0-

100 amended BSF.

101 Each of the six named operational parameters enumerated above has been reported to

102 significantly impact the performance of the BSF for water treatment [2,4,33,45,59]. In

103 particular, despite amendment with Fe^0, resulted BSF are still tested for their capacity for

104 microbial attenuation and the removal mechanism controversially discussed [33,62,63]. This

105 controversial discussion is not in accordance with the contemporary knowledge on the

106 mechanism of contaminant removal in Fe^0 beds [14,15,64-70] as iron corrosion products

107 should be regarded as 'collectors' in BSF [71].

2.2   Operating mode of a BSF

109 The supernatant water layer provides a head of water that is sufficient to drive the water

110 through the filter bed, whilst creating a retention period of several hours for the water

111 [2,55,72]. A BSF is doted with a underdrain system which provides an unobstructed passage

112 for treated water from the filter bed and supports the sand bed. The outlet flow control

113 maintains submergence of the medium during operation to minimise potential air-binding

114 problems.

115 Water percolates slowly through the porous sand medium. Thereby, inert particles and

116 micoorganisms are removed from the aqueous phase. An algal mat forms on the surface of the

117 sand bed and this is termed *Schmutzdecke* (a biolayer or biofilm of living organisms). After

118 several months of operation, the surface of a BSF becomes clogged due to the deposition of

119 suspended solids. At this time, cleaning of the filter bed is required. The BSF is cleaned by

120 scapping off the top 2-3 cm of the sand bed including the *Schmutzdecke* layer. After a

121 scraping of the sand bed, a re-sanding is necessary with the accompanying maturation time of

122 the formation of a new *Schmutzdecke*.

2.3   Limitations of a conventional BSF

124 Discounting any design limitations, the BSF is not destined to treat chemical contaminants in

125 general and inorganic contaminants in particular. In fact, the affinity of sand for metal

126 adsorptive removal is very low [73,74]. Accordingly, whenever chemical contamination is

127 suspected, alternative to conventional BSF should be sought. One such an alternative is the

128 amendment of conventional BSF by a reactive layer containing reactive Fe^0. The suitability of

129 Fe^0/sand filters for water treatment arises from the fact that iron corrosion products act as

130 collectors [71,75] for all classes of contaminants [62,66,76].

131 The suitability of Fe^0-amended BSF for the developing world arises from the fact that the

132 quality of available water is rarely assessed. A perfect illustration is the arsenic crisis in South

133 East Asia where people have consumed As-contaminated waters for decades [41,77]. On the

134 other hand, success of conventional BSF and other water treatment technology are not

135 currently validated by biological and/or chemical analysis but by the decrease of the

136 frequency of water born diseases [4,8,13,21,44]. For all these reasons, the need of a

137 technology able at removing all classes of contaminants for water is obvious.

138 **3 Fe^0-amended BSF on an historical perspective**

139 The present section will consider only stand-alone Fe^0 filters for save drinking water

140 provision.

141 **3.1   First generation Fe^0-amended BSF: removal of chemicals**

142 The efficiency of iron-oxide-coated sand for contaminant removal is well-documented and

143 has been used for water treatment in household filters for decades [20,42,78-80].

144 Conventional BSF have been proven efficient to remove dissolved iron from the aqueous

145 phase. Moreover, BSF has been proven more efficient in removing inorganic contaminants

146 (e.g. As) when the inflowing water was rich in dissolved Fe. Accordingly, Khan et al. [37]

147 amended conventional 3-Kolshi filters with a layer of Fe^0 to achieve better As removal by a

148 BSF-like filter. It is very important to notice that Fe^0 is added by Khan et al. [37] to produce

149 iron oxides for As removal in filters. Accordingly, the Fe^0-amended 3-Kolshi filter can be

150 regarded as first generation Fe^0-amended BSF.

151 The first generation Fe^0-amended BSF was proven very efficient but not sustainable because

152 of too rapid loss of hydraulic conductivity [40,41,77]. As demonstrated in previous works

153 [12,47-51,54], "diluting" Fe^0 with (inert and) non-expansive materials is a pre-requisite for

154 sustainability. In other words, the first generation Fe^0-amended BSF was wrongly designed.

3.2 Second generation Fe^0-amended BSF: removal of pathogens

The second generation Fe^0-amended BSF was born with the work of You et al. [81]. These authors demonstrated that adding Fe^0 to a sand filter, efficiently removed viruses. The removal was ascribed to pathogen adsorption onto iron (hydr)oxides formed via (anaerobic) iron corrosion. Virus removal by Fe^0 was rapid and most of the removed viruses were irreversibly bound. The irreversible bounding of viruses by Fe^0 corresponds to co-precipitation as presented two years later by Noubactep [64-70]. Strictly, the filter of You et al. [81] is not necessarily a biosand filter but a Fe^0 filter for pathogen removal. In fact, the formation a *Schmutzdecke* layer is not explicitly intended. Rather, Fe^0 itself is the pathogen removing agent. However, recent research based on You et al. [81] are testing Fe^0-amended BSF as alternative to conventional BSF [33,45,46,63,72].

3.3 Third generation Fe^0-amended BSF: removal of chemicals and pathogens

The third generation Fe^0-amended BSF was born with the work of Ngai et al. [38]. The filter designed by Ngai et al. [38,39] combines the concept of a BSF with the innovation of a diffuser basin containing reactive Fe^0. In this design, pathogens are removed mostly by physical straining provided by the fine sand layer (sand bed) [23]. Chemical contamination (e.g. As) is removed by adsorption onto iron oxides and hydroxides generated from rusted Fe^0.

A fundamental mistake was recently discovered in this design [12]. In fact, molecular O_2 necessary for the formation of the *Schmutzdecke* is quantitatively consumed by the over-laying Fe^0 layer. In other words, a reactive over-laying Fe^0 makes the BSF inoperative for pathogen removal as no *Schmutzdecke* is formed. Thus, the obtained filter is at best a first generation Fe^0 filter and strictly not a Fe^0-amended BSF because the 'Fe^0 unit' and the 'BSF unit' are two different components of the filter. On the other hand a pure Fe^0 layer (100 % Fe^0) at the entrance of the filter will rapidly clog due to the formation of voluminous corrosion products ($\eta > 2.1$) [52,53].

181 **3.4 Fourth generation Fe^0-amended BSF**

182 The fourth generation Fe^0-amended BSF was born with the work of Gottinger [43]. It is

183 characterized by a reactive zone containing a mixture of Fe^0 and an inert material (e.g. sand).

184 Fe^0 is purposefully mixed with sand to prevent clogging [12]. It should be noticed that mixing

185 Fe^0 and sand is current in the research on Fe^0 for groundwater remediation [74,82-86]. The

186 present communication is limited to household and small community Fe^0 filters. However, the

187 results are up-scalable to larger Fe^0/H_2O systems.

188 The filter designed by Leupin et al. [87,88] fulfilled these criteria but is classed here as the

189 chronology is not an absolute factor. Furthermore this filter was designed mainly for As

190 removal. Gottinger [43] was the first researcher to mix Fe^0 and sand on a volumetric basis and

191 test several Fe^0/sand ratios (e.g. 50/50 and 40/60 and 0/100) in triplicate columns. Rangsivek

192 and Jekel [89-91] tested 10/90, 20/80 and 30/70 Fe/pumice volumetric mixtures for metal

193 removal but the tests were not systematic. They mostly worked with the 10/90 mixture

194 without specifying the rational [89,91]. Independent theoretical calculations [47,54] have

195 shown that the threshold value of the volumetric proportion of Fe^0 for which the column

196 reactive zone is clogged at Fe^0 depletion is 52 %. The calculations were based on the

197 assumption of ideally packed spherical Fe^0 particles for which an initial porosity of 36 % is

198 relevant. Natural sand is never perfectly spherical. Kubare and Haarhoff [32] reported an

199 average porosity of 45 % for sand beds. However, the theoretical threshold value for Fe^0

200 depletion of 52 % (corresponding to 36 % porosity) will be considered in discussing the

201 design of Fe^0-amended BSF. The meaning of this value is that any filter containing more than

202 52 % Fe^0 (volumetric proportion of the solid phase) will clog before Fe^0 completely depletes.

203 The excess Fe^0 amount relative to 52 % should be regarded are pure material wastage with the

204 additional curse of shortening the filter service life [54].

205 **4 Designing Fe^0-amended BSF**

206 **4.1 Strategic planning**

207   The presentation above showed that a Fe^0-amended BSF should be sequenced as follows: (i)

208   supernatant water (e.g. 5 cm), (ii) fine sand (e.g. 40 to 50 cm), (iii) Fe^0/sand (x cm), (iv) fine

209   sand (y cm), (v) sand (e.g. 5 cm) and (vi) gravel (e.g. 5 cm). The thickness of individual

210   layers is from Lea [56]. In the design of Jenkins et al. [72] sand and gravel are replaced by

211   gravel and rock respectively. The major feature for these layers is the difference in particle

212   size. The above sequence suggests that the depths of the Fe^0/sand layer and that of the

213   underlying fine sand are yet to be determined (x and y values). However, the paramount

214   question is which Fe^0 material should be used?

215   Characterizing the intrinsic reactivity of Fe^0 materials [43,92-98] is a key issue for designing

216   Fe^0-amended BSF. The ideal material should be able to efficiently provide clean water for at

217   least 12 months (one year). Therefore, all tested materials should be characterized for their

218   intrinsic chemical reactivity [94,95,98] and their sphericity in order to enable results

219   comparability. Next to the intrinsic reactivity, used devices should be characterized by

220   [32,61]: (i) their dimensions, e.g. internal diameter and depth of reactive layers for cylindrical

221   columns, (ii) the particle size and the surface state of used materials (e.g. Fe^0, gravel, pumice,

222   sand), (iii) the mass of Fe^0, and (iv) the volumetric proportion of Fe^0 in the reactive layer. The

223   next section will discuss the thickness of the reactive layer.

224   **4.2   Thickness of the Fe^0/sand layer**

225   A cylindrical bed is considered; H is the height and D is the internal diameter. The cylinder

226   contains a reactive zone with the height H_{rz} (x value from section 4.1) and the volume V_{rz}. H_{rz}

227   is necessarily lesser than H (x < H or $H_{rz} < H$). Beds are supposed to be filled by spherical

228   granular materials. The compactness (or packing density) C (-) is defined as the ratio of the

229   volume of the particles to the total packing volume (V_{rz}). Considering the granular material as

230   composed of mono-dispersed spheres subjected to soft vibrations, the compactness C is

231   generally considered to be equal to 0.64 for a random close packing [51]. It is assumed that

232   the particles are non porous.

233   The initial porosity Φ_0 (-) of the reactive zone and the thickness H_{rz} of the reactive zone are

234   respectively then given by:

235                              $$\Phi_0 = 1 - C \qquad\qquad (1)$$

236                              $$H_{rz} = \frac{4}{\pi D^2} \cdot V_{rz} \qquad\qquad (2)$$

237   The filling of the bed porosity by iron corrosion products determines the filter service life and

238   has been extensively discussed in previous works [51,54].

239   To completely define the x value (H_{rz}), V_{rz} has to be correlated to the mass of Fe^0. V_{rz} is the

240   volume occupied ideally by Fe^0 and sand of similar grain size and shape (Eq. 3). For

241   simplification V_{Fe} will be expressed as a fraction of V_{rz} (Eq. 4).

242                              $$V_{rz} = V_{pore} + V_{Fe} + V_{sand} \qquad\qquad (3)$$

243                              $$\alpha_{pore} + \alpha_{Fe} + \alpha_{sand} = 1 \qquad\qquad (3a)$$

244                              $$V_{pore} = \alpha_{pore} * V_{rz} \qquad\qquad (4)$$

245                              $$V_{Fe} + V_{sand} = (1 - \alpha_{pore}) * V_{rz} \qquad\qquad (4a)$$

246                              $$V_{Fe} = \tau * (V_{Fe} + V_{sand}) = \tau * (1 - \alpha_{pore}) * V_{rz} \qquad (5)$$

247   Where α_i is the volumetric fraction of each phase, τ (≤ 0.52) is the volumetric proportion of

248   Fe^0 relative to the solid phase (Fe^0 + sand) in the reactive zone [54]. Per definition, $\alpha_{pore} =$

249    $\Phi = 1 - C$ and $C = 1 - \Phi$. Thus Eq. 5 reads:

250                              $$V_{Fe} = \tau * C * V_{rz} \qquad\qquad (5a)$$

251   The mass of Fe^0 necessary to fill the volume V_{Fe} is given by Eq. 6:

252                              $$m_{Fe} = \rho_{Fe} * V_{Fe} \qquad\qquad (6)$$

253   Where ρ_{Fe} is the specific weight of Fe (7800 kg/m^3).

254   Combining Eq. 2, 4, 5 and 6 give the following relationship between H_{rz} (x value) and m_{Fe}

255   (Eq. 7):

256                              $$H_{rz} = 4 * m_{Fe} / [(\pi D^2) * \tau * C * \rho_{Fe}] \qquad (7)$$

257 Eq. 7 is the equation of the reactive zone. Three major issues should be considered: (i) $\tau \leq$

258 0.52 (52 % Fe^0 (v/v)), (ii) $H_{rz} \geq 5$ cm, and (iii) $H_{rz} < H$. The threshold value $H_{rz} = 5$ cm is

259 considered the minimum height for the realization of a homogeneous well-mixed Fe^0/sand

260 reactive zone [99]. Ideally, H_{rz} is only a fraction of H ($H_{rz} << H$) for example, Noubactep et

261 al. [12] considered that H_{rz} should fulfil the condition $H_{rz} \leq 0.1$ H.

262 The mass of sand to be used is deduced from Eq. 8.

$$m_{sand} = \rho_{sand} * V_{sand} = \rho_{sand} * (1 - \tau) * C * V_{rz} \qquad (8)$$

264 Where ρ_{sand} is the specific weight of sand (2650 kg/m^3).

4.2.1 Illustration

266 To discuss the applicability of the established equations, the biosand filter of Lea [56] with a

267 diameter of 30 cm a height of 90 cm is used. The bed volume of this filter is 63.6 L.

268 Accordingly if the reactive layer must occupy at most 1/10 of the bed, its volume should not

269 be larger than 6.4 L.

270 For comparison, at $\tau = 0.52$, 1 kg of Fe^0 is mixed to 0.16 kg of sand and the mixture occupies

271 a volume (V_{rz}) of 0.2 L. When mixed to 8.32 kg of sand, the same mass of Fe^0 (1 kg)

272 corresponds to a τ value of 0.02 (about 6 % w/w) and the mixture occupies a volume $V_{rz} =$

273 5.01 L.

274 Given that water treatment by Fe^0 is a deep bed filtration, the present example illustrates the

275 necessity of using lower values of τ with the additional advantage of reducing the clogging

276 probability. The discussion in the next section will be mostly based on the threshold τ value of

277 0.52.

4.3   Discussion

279 This work attempts to sustain research on Fe^0-amended BSF by optimizing filter design in the

280 perspective to rationalize experimental conditions and enable/ease results comparison.

281 Accordingly, the most important issue regards sizing a filtration bed. Both for laboratory and

282   field works, one of the first task is to decide which column(s) to use. In some cases, available

283   columns are simply used. But even in these cases, they should be properly filled to achieve

284   reliable results. Sizing a filter bed will be discussed here in two different perspectives: (i)

285   selecting the appropriate bed size, and (ii) selecting the appropriate thickness of the reactive

286   layer (H_{rz}).

287   **4.3.1   Rationale selection of the column diameter**

288   Eq. 7 gives the thickness of the reactive zone H_{rz} as a function of m_{Fe}, D, τ and C. From these

289   4 parameters, C can be considered a constant. Accordingly, H_{rz} is a function of the used mass

290   of Fe^0 (m_{Fe}), the volumetric proportion of Fe^0 in the solid phase (τ) and the internal diameter

291   of the cylinder (D). Accordingly, Eq. 7 can allow the calculation of the diameter of the

292   column to be used to achieve a certain H_{rz} at a given τ value.

293   The results of such calculations are summarized in Tab. 1 for $\tau = 0.52$, C = 0.64 cm and the

294   following values of m_{Fe}: 1.0, 2.5, 5.0, 10.0 and 25.0 kg. From Tab. 1, it is obvious that the

295   sole mass of Fe^0 that is applicable for lab experiment (D = 3 cm) is 1 kg. The corresponding

296   H_{rz} value is 55 cm which represents 61 % of the filter height (90 cm). Rigorously, the

297   resulting system is not a BSF but rather a SONO-like filter. Nevertheless, the following

298   sequence could be tested: 20 cm sand bed, 55 cm reactive zone (x = 55 cm) and 15 cm sand

299   bed (y = 15 cm). Whether this design is satisfactory or not should be tested in laboratory

300   investigation. In the case y = 15 cm sand bed is not sufficient to capture dissolved Fe from the

301   reactive zone, the possibility of adding a second sand bed should be tested (Fig. 1). On the

302   other hand, for $H_{rz} > 0.33$ H the perspective of sandwiching an Fe^0/filter between two BSF

303   must be tested (Fig. 2). The first BSF removes dissolved O_2 among others and the second BSF

304   removes dissolved Fe.

305   Fig. 3 summarizes the results of the variation of H_{rz} as a function of D ($\tau = 0.52$). It is clearly

306   shown that the thickness of the reactive layer increases with decreasing diameter. It is shown

307   (Fig. 3a) that H_{rz} values of up to more than 1300 cm (13 m) are obtained for larger m_{Fe} values.

308   However, relevant values must fulfil the condition $H_{rz} \leq H$ ($H = 90$ cm). Fig. 3b is limited to

309    $H_{rz} = 0.67$ H (60 cm) for a better visualization. Table 1 shows that D = 50 cm is the largest

310   value which may enable a 5 cm H_{rz} while using 25 kg Fe^0. this situation is likely to occur in a

311   field small-community-scale water plant. However, because Fe^0 beds are deep bed filtration

312   systems, it is advantageous to use a lower τ value ($\tau \leq 0.52$) to achieve a thicker H_{rz} with the

313   same Fe^0 mass.

4.3.2   Rationale for the thickness of the reactive zone

315   The thickness of the reactive zone is necessarily correlated to the intrinsic reactivity of used

316   Fe^0. It is intuitive to assume that for each Fe^0 material, a range of τ values ($\tau \leq 0.52$) may exist

317   for which filter operation is optimal. A survey of the experimental conditions of available

318   works suggests that they are highly qualitative. For example, Tellen et al. [45] introduced a 5

319   cm reactive zone containing 0.5 kg Fe^0 in 44 cm sand bed without specifying the thickness of

320   the sand layer under-laying the reactive zone.

321   The design of Tellen et al. [45] was slightly modified by Pachocka [33]. She introduced a 3.2

322   cm reactive zone containing a Fe^0 volumetric proportion of 15 % ($\tau \leq 0.15$). The Fe^0 mass is

323   not specified but calculations using Eq. 7 while considering the geometry and the dimensions

324   of her device showed that about 2.2 kg Fe^0 was used. Discounting the fact that building a

325   homogenous 3.2 cm Fe^0/sand layer is difficult, it is certain that such a thin layer is not suitable

326   for a deep bed filtration process. Accordingly, guidelines are urgently needed to assist the

327   laborious work of amending conventional BSF with reactive Fe^0.

328   The suitability of (τ, H_{rz}) values for several Fe^0 materials and material amounts should be

329   tested in laboratory studies before a trend is identified for generalization and scaling up to

330   household and small-community Fe^0-amended BFS.

331   For further illustration, calculations are made using Eq. 7 for D = 30 cm and various Fe^0

332   masses. The results are summarized in Fig. 4 and Tab. 2. Values in Tab. 2 are obtained for $\tau =$

333 0.52. Fig. 4 shows clearly, that H_{rz} decreases with increasing τ values. For example, 25 kg of

334 Fe^0 occupy a 13.6 cm at $\tau = 0.52$, 26.2 cm at $\tau = 0.27$ and   101.2 cm at $\tau = 0.07$. While

335 limiting the scale to $H_{rz} = 0.67$ H ($H_{rz} = 60$ cm), it is shown that 5.0, 10.0 and 25.0 kg are not

336 suitable for $\tau < 7.0$ % (or $\tau < 0.07$). These examples show clearly that there is an infinite

337 number of possible Fe^0-amended BSF. A systematic approach is therefore essential to identify

338 and characterize useful combinations. The next section will discuss the design of a BSF

339 containing 3 kg of a reactive Fe^0.

340 **4.4   Constructing a Filter with 3 kg Fe^0**

341 A conventional concrete intermittent BSF is used. An approach is suggested to purposefully

342 amend it with a reactive zone containing 3 kg Fe^0. First, Eq. 7 is modified to account for the

343 filter geometry. The cross-section (S) is a square with a length (L) of 30 cm (0.3 m). The

344 volume of the filter is $V = S*H = L^2*H$. The modified Eq. 7 reads as (Eq. 7a):

345 $$H_{rz} = m_{Fe} / [(L^2) * \tau * C * \rho_{Fe}] \qquad (7a)$$

346 The results (Tab. 3) show that H_{rz} takes values from 1.28 to 33.39 cm when the volumetric

347 proportion of Fe varies from 0.52 to 0.02 %. The corresponding admixed mass of sand varies

348 between 0.94 and 49.94 kg. Taking 50 cm as the maximal thickness of fine sand (sand bed)

349 the resulting beds represent 2.6 to 66.8 % of the sand bed volume.

350 Considering the practical constrain that $H_{rz} \leq 5$ cm [99], only τ values less than 0.25 are

351 applicable. That is, 3.0 kg Fe^0 should be mixed to at least 6.46 kg sand to obtain a Fe^0-

352 amended BSF. The efficiency of selected possible filters ($\tau \leq 0.25$) should be tested. It is

353 important to notice in this regard that the first permeable reactive barrier (demonstration pilot

354 scale in Borden, Ontario/Canada) contained only 8 % volumetric ratio of Fe^0 ($\tau = 0.08$) and

355 has been properly working for more than five years [82,100]. Accordingly, smaller τ values

356 should be tested in parallel experiments. In this effort the suitability of the thickness of the

357 under-laying sand bed should be tested (y value section 4.1 – Fig. 1).

358 For $P_{rz} > 10$ % ($\tau \leq 0.12$ – Tab. 3) the alternative of a three-column-system should be used

359 (Fig. 2). In the perspective of a Fe^0/sand column in sandwich between two conventional BSF

360 (section 4.3.1) the possibility of using thin columns to save Fe^0 must be tested. For example,

361 while using 1 kg of Fe^0, halving the internal diameter from 9.0 to 4.5 increases the H_{rz} value

362 from 6.1 to 24.2 ($\tau = 0.52$ – Fig. 3). This is a 4 times thicker layer for deep bed filtration in

363 comparison to only 5.6 cm H_{rz} in a conventional BSF (L = 30 cm – Tab. 3). In other words,

364 using thinner columns enhanced efficiency and save Fe^0.

365 The mathematical relation for the variation of H_{rz} values at constant volume ($V_{rz1} = V_{rz2}$) is

366 easy to establish and is given as follows:

367
$$H_{rz1}*D_1^2 = H_{rz2}*D_2^2 \tag{9}$$

368 For $D_2 = D_1/2$, $H_{rz2} = 4* H_{rz1}$. This corresponds to the results obtained while working with

369 masses.

370 **4.5 Ways to efficient Fe^0-amended BSF**

371 The presented concept of Fe^0-amended BSF is based on the profound understanding of the

372 complex chemical and physical processes involved in the 20-years-old Fe^0 remediation

373 technology [64-70,82,100-105]. To mimic the subsurface Fe^0 bed as closely as possible,

374 anoxic conditions must be created before the Fe^0/sand layer (reactive zone). This condition is

375 satisfied the best by a conventional BSF. On the other hand, to avoid dissolved Fe in the

376 effluent, water from the reactive zone must migrate through a thick fine sand bed. Ideally this

377 is another conventional BSF (Fig. 2). Accordingly, the simplest way to efficient Fe^0-amended

378 BSF seems to go through an experimental design with a reactive zone (a small column – Eq.

379 9) sandwiched between two conventional BSF. After such conclusive principle experiments,

380 the next advantageous step could be to test three compartments in the same column (Fig. 1).

381 In this manner, the important requirement of compact systems for less skilled populations

382 (including illiterates) is properly addressed [19]. Progressively, it is conceivable to

383   miniaturize the system down to Brita-type filters for a limited volume of tap water. However

384   the focus of this communication is on household and small-community Fe^0-amended BSF.

385   Provided that a relevant reactive Fe^0 is characterized, selected, and used, the effectiveness of

386   Fe^0-amended BSF does not need to be demonstrated, except some technology verification in

387   the field (monitoring). Beside proper system design, the two sole tasks are: (i) selecting and

388   processing the appropriate Fe^0 materials (including composites), and (ii) avoiding the use of

389   Fe^0-amended BSF for waters of pH < 4.5 [12]. In fact, the Fe^0 remediation technology is

390   based on the anodic dissolution of iron in neutral and close to neutral aqueous systems. In this

391   pH range, primary iron dissolution is followed by a continuous build up and transformation of

392   a corrosion product layer in the vicinity of Fe^0 [106-108].

393   There is strong evidence that the Fe^0-amended BSF will be efficient. For example, Westerhoff

394   and James [109] performed field continuous-flow experiments at $\tau = 0.25$ ($P_{Fe} = 50$ %. w/w)

395   for almost one year for efficient removal of nitrate. During this period, up to 1500 bed

396   volumes of water were treated. Assuming that two bed volumes of a BSF correspond to the

397   daily need of a rural family, the 1500 bed volumes corresponds to the water demand for 2

398   years. However, the τ value (0.25) of Westerhoff and James [109] was not the result of any

399   systematic preliminary work. Considering that a system at $\tau = 0.08$ was efficient for more

400   than 5 years under anoxic conditions [82,100], it is likely that the system of Westerhoff and

401   James [109] was not optimal with regard to long-term permeability.

402   **5   Concluding remarks**

403   Save drinking water provision using Fe^0-amended BSF is already proven an efficient

404   technology [33,37,41,45,46,81]. The estimated huge number of people (884 million) still

405   living without access to improved drinking water [110] is an urgent appeal to the scientific

406   community for (i) a rapid improvement, and (ii) a generalized implementation of this efficient

407   technology. This technology primarily has a single serious limitation: available water must

408   have a pH value ≥ 4.5. Fortunately, natural water with pH ≤ 4.5 is unusual. Therefore, Fe^0-

409   amended BSF should be regarded as a universal technology that can be used at household and

410   small community levels, but in general for decentralized safe drinking water provision.

411   Using a mathematical modelling, the present work has presented several tools to optimise the

412   efficiency of conventional BSF by adding a reactive zone containing a layer of Fe^0 admixed to

413   a non expansive material (e.g. MnO_2, pumice, sand) [47,51,54]. A systematic procedure is

414   introduced to sustain further research on Fe^0-amended BSF. In particular the reactive zone

415   must have a minimal thickness of 5 cm ($H_{rz} \leq 5$ cm), and the volumetric proportion of Fe^0 in

416   the reactive zone must not be larger than 52 % ($\tau \leq 0.52$). As concerning the sequential design

417   of the Fe^0-amended BSF, it is essential that dissolved O_2 is quantitatively removed from water

418   before its reaches the reactive zone. On the other hand, a find sand layer must be placed

419   downwards from the reactive zone and its thickness should be sufficient for scavenging

420   dissolved Fe from the reactive zone. Like for other filters [4,40] layers of other treatment

421   materials (e.g. activated carbon, wooden charcoal, Zeolite) [111,112] can be purposefully

422   added before and/or after the reactive zone to optimise the treatment operation. It is essential

423   to notice in this regard that efficient and low-cost traditional methods for aqueous iron

424   removal in packed columns have been described in the literature [112-116].

425   Further research at several fronts is needed to develop approaches for the proper design of the

426   Fe^0-amended BSF. Relevant research fields include the intrinsic reactivity of Fe^0, the size,

427   surface state and geometry of Fe^0 and non reactive particles, the size of the filter or the

428   column, the configuration the Fe^0/admixture layer, the thickness of individual layers, the

429   nature of the admixing agent, the relative particle size of materials in the reactive layer

430   (uniformity), the size of the resulting devices, and the water flow velocity. Further

431   mathematical and numerical modelling should be applied to design and validate experimental

432   results. Pilot scale installations are needed to validate the practicality of experimental and

433   modelling results.

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737

737 **Figure captions**

738

739 **Figure 1**: Schematic diagram of a Fe^0-amended biosand filter (BSF). The first column is a

740 conventional BSF. The thickness of the reactive zone (x value) and the aptitude of the under-

741 laying sand bed (y value) to free water from the reactive zone from iron will be tested for each

742 material. For larger x values or very reactive Fe^0, a third column could be essential.

743

744 **Figure 2**: Schematic diagram of a three compartments Fe^0-amended biosand filter (BSF). The

745 first and the third columns are conventional BSF. The thickness of column 2 (reactive zone)

746 depends on the intrinsic reactivity of used Fe^0 and can be optimised using Eq. 7 and 9. The

747 dimension of column 3 (second BSF) and also be optimise using Eq. 8 and 9.

748

749 **Figure 3**: Variation of the height of the reactive layer (H_{rz}) as function of the diameter (D 100

750 $\leq$ cm) of used filter: (a) all values, and (b) $H_{rz} \leq 60$ cm. The lines are not fitting functions;

751 they simply connect points to facilitate visualization.

752

753 **Figure 4**: Variation of the height of the reactive layer (H_{rz}) as function of the initial Fe^0

754 volumetric proportion for different used masses of Fe^0. The lines are not fitting functions;

755 they simply connect points to facilitate visualization.

756

757

757 **Table 1**: Variation of the height of the Fe^0/sand reactive layer (H_{rz}) as a function of the

758 internal diameter ($3.0 \leq D$ (cm) ≤ 100) of used columns for a τ value of 0.52. The

759 corresponding masses of sand and Fe^0 are given. Given that $\tau = 0.52$ is the threshold value for

760 sustainable Fe^0 beds, these calculations show clearly that for laboratory studies (D $\leq$ 12 cm), a

761 maximum of 2.5 kg sand should be used to keep the reactive layer lesser than 10 cm. On the

762 other hand, if a field column has an internal diameter of 50 cm 25 kg Fe^0 will build a 5 cm

763 reactive layer.

764

D	Fe^0 mass				
(cm)	1.0 kg	2.5 kg	5.0 kg	10.0 kg	25.0 kg
3	55.0	136.2	272.4	544.8	1362.0
6	13.6	34.1	68.1	136.2	340.5
12	3.4	8.5	17.0	34.1	85.1
30	0.54	1.36	2.72	5.45	13.62
50	0.20	0.50	1.00	2.00	5.00
75	0.09	0.22	0.44	0.87	2.18
100	0.05	0.12	0.24	0.49	1.21

765

766

766   **Table 2**: Variation of the height of the Fe^0/sand reactive layer (H_{rz}) as a function of the mass

767   of Fe^0 for a τ value of 0.52. The corresponding mass of sand is given. Calculations

768   are made for a cylindrical bed with a internal diameter of 30 cm. Given that $\tau = 0.52$

769   is the threshold value for sustainable Fe^0 beds, such a bed should not contain less

770   than 10 kg of Fe^0 to fulfil the condition $H_{rz} \leq 5$ cm. The corresponding V_{rz} value is

771   3.85 L.

772

Fe^0	(kg)	0.5	1.0	2.5	5.0	7.5	10.0	20.0
Sand	(kg)	0.157	0.314	0.785	1.568	2.352	3.136	6.272
H_{rz}	(cm)	0.27	0.54	1.36	2.72	4.09	5.45	10.90
V_{rz}	(L)	0.19	0.39	0.96	1.93	2.89	3.85	7.71

773

774

775

775 **Table 3**: Data for the amendment of a conventional BSF with a reactive layer containing 3 kg

776 Fe^0 mixed with several masses of sand (m_{sand}) to yield various proportion of Fe^0 (P_{Fe}). H_{rz} is

777 the resulting thickness of the reactive layer, V_{rz} its volume and P_{rz} its volumetric ratio relative

778 to a 50 cm reactive layer.

779

780

τ	H_{rz}	V_{rz}	m_{sand}	P_{Fe} (%)		P_{rz}
(-)	(cm)	(L)	(kg)	(w/w)	(v/v)	(%)
0.52	1.28	1.16	0.94	76.13	52.0	2.6
0.47	1.42	1.28	1.15	72.30	47.0	2.8
0.42	1.59	1.43	1.41	68.07	42.0	3.2
0.37	1.80	1.62	1.74	63.35	37.0	3.6
0.32	2.09	1.88	2.17	58.07	32.0	4.2
0.27	2.47	2.23	2.76	52.12	27.0	4.9
0.22	3.04	2.73	3.61	45.36	22.0	6.1
0.17	3.93	3.54	4.98	37.61	17.0	7.9
0.12	5.56	5.01	7.47	28.64	12.0	11.1
0.07	9.54	8.59	13.54	18.14	7.0	19.1
0.02	33.39	30.05	49.94	5.67	2.0	66.8

781