

Metallic Iron (Fe<sup>0</sup>) Provide Possible Solution to Universal Safe Drinking Water Provision

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**Abstract**

The use of metallic iron (Fe<sup>0</sup>) for environmental remediation is applied industrially with a great degree of empiricism. Rules of thumb seem to be the only guide followed by the greater part of the Fe<sup>0</sup> remediation community. The present communication demonstrates the validity of such a harsh statement and hopes that the research community will now follow the 10-year-old path concealing Fe<sup>0</sup> remediation and mainstream science. A promising application is safe drinking water provision on a decentralized manner.

**Keywords**

Environmental remediation; Frugal technology; Safe drinking water; Zero-valent iron.

**Introduction**

The use of metallic iron (Fe<sup>0</sup>) in water treatment was established in the second half of the nineteenth century [1-4]. Related investigations cleared up the nature of Fe<sup>0</sup> as coagulant-inducing agent both in the Bischof Process (packed-bed filtration) and in the Anderson Process (fluidized bed-like) [1,4-9]. During the past three decades, sophisticated scientific methods have been used to produce a confused mass of discordant and mostly unfounded individual opinions [10-12] demonstrating that Fe<sup>0</sup> might be an environmental reducing agent [8,9,13-33]. This fact alone explains the absolute empiricism which thus far has dominated and still dominates research and field application of Fe<sup>0</sup> for environmental remediation and water treatment. The theory of reducing Fe<sup>0</sup> was proven false one decade ago [34-37]. In the meantime, new scientific data from systematic and rigorous studies on the Fe<sup>0</sup>/H<sub>2</sub>O system have been presented [12,31,32,38-46].

Contaminant removal in Fe<sup>0</sup>/H<sub>2</sub>O systems was wrongly assumed to be an electrochemical process wherein Fe<sup>0</sup> is oxidatively dissolved and the contaminant is simultaneously reduced (cathodic reaction) [25,46]. This mechanism has never been scientifically demonstrated nor univocally accepted [34]. The first mistake was the absence of a mass balance of iron coupled to the incomplete mass balance of the contaminants [11,12,47,48]. In fact, non-recovered original species (contaminants) were considered reductively transformed [12,35,43,44,49]. It was properly considered that aqueous iron corrosion produces Fe hydroxides (and oxides) 'passivating' the reactive surface of Fe<sup>0</sup> (reactivity loss). However, two important facts were overseen: (i) water is the solvent meaning that hydroxide generation is quantitative, and (ii) quantitative hydroxide availability implies coagulation (Anderson Process). Accordingly, a real mass balance of any contaminant should go through reductive dissolution of hydroxides/oxides (e.g. sequential extraction) to liberate the 'coagulated' fraction [50,51]. The other aspect of in-situ generation of iron hydroxides (and oxides) is that they are larger in volume than their parent (Fe<sup>0</sup>), meaning that the initial porosity of a packed bed containing Fe<sup>0</sup> will be progressively filled. This is the first cause of permeability loss in porous and permeable media [52-54]. It is essential to recall that all these fundamental aspects were documented in the scientific literature before the advent for Fe<sup>0</sup> reactive walls in the 1990s (Tab. 1) [11,12, 33]

Even though recent results [52] have confirmed the reasonableness of the hypothesis of foreign mineral precipitates as major cause of permeability loss [55], yet the opinion continued to exist that mixing granular Fe<sup>0</sup> and other aggregates including sand is a tool to save Fe<sup>0</sup> costs. Such an opinion, which has in some respects been confirmed scientifically, gave rise to an immense number of attempts to improve the efficiency of nano-scale Fe<sup>0</sup>. However, these attempts had as their exclusive object the identification of suitable materials to disperse nano-Fe<sup>0</sup> for the reductive transformation of contaminants. The research on using nano-Fe<sup>0</sup> for environmental remediation is still conducted along exclusively empirical lines [56-58].

The whole research on Fe<sup>0</sup> for water remediation is merely a question of obtaining maximum contaminant removal in minimum time [11,12,36,37,59]. This approach has mediated the development of multi-metallic systems and nano-Fe<sup>0</sup> but is questionable because material selection should be based on site specific conditions. In particular, the solution of each individual problem requires precise knowledge including the flux of

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Time	Discovery / event	Author(s)	Application
1675	Erosion corrosion	Boyle	Anderson process
1763	Bimetallic corrosion	British Navy	Multi-metallics
1871	Sponge iron for drinking water	[5]	Household filters
1883	Sponge iron for drinking water	[5]	Large scale filters
1885	Iron filings in revolving purifiers	[6]	Fe <sup>0</sup> as coagulant
1923	Iron corrosion is volumetric expansive	[163]	Filter design
1938	Electrochemical aqueous Fe <sup>0</sup> corrosion	Evans	Fe <sup>0</sup> is not an own reducing agent
1951	Steel wool for safe drinking water	[144]	Drinking water
1986	Iron filings for agricultural wastewaters	[145]	Wastewater
1992	Iron metal for groundwater remediation	[13]	Groundwater
2001	Fe-micro-alloyed aluminium composites	[140]	Pathogen removal
2004	Fe-micro-alloyed aluminium composites	[139]	Chemical removal
2007	Porous composite iron material for DW	[99]	Household filters
2007	Iron nails for drinking water	[98]	Household filters
2009	Iron metal as universal material for DW	[71]	Drinking water
2017	Discovery of ancient iron technology	[8]	Water treatment

**Table 1:** Time-line of the history of metal corrosion outlining the main information for the design of sustainable Fe<sup>0</sup> filters. The key information is more than 70 years-old. DW stands for drinking water.

contaminants and the solution chemistry [33,60-65]. Based on such knowledge and the intrinsic reactivity of Fe<sup>0</sup> materials, the most suitable material should be selected for designing the water treatment plant (after pilot testing). Despite three decades of intensive research, there is no unified tool to assess the suitability of available Fe<sup>0</sup> materials [66-70].

The complexity of the Fe<sup>0</sup>/H<sub>2</sub>O system has been used to justify or explain why innumerable attempts made during the past 30 years to improve this remediation technology have not been really successful. Coming to the last decade [11,12,34,35,38-40,43,71], however, it is seen that investigations following a truly scientific direction is possible and even successful (Table 1). The alternative approach furnishes a sure means for determining rapidly a large number of operating parameters, such as the Fe<sup>0</sup>/sand ratio [72,73] the depth of the reactive zone, the selectivity of the system [74-76], the suitability of other aggregates [67,77,78] which are essential for pilot testing the Fe<sup>0</sup> remediation technology for (i) safe drinking water provision, (ii) wastewater treatment and (iii) environmental remediation.

The next section recalls the nature of the Fe<sup>0</sup>/H<sub>2</sub>O system to demonstrate that successful system design is possible despite confusing literature report.

### The essence of iron corrosion

Iron corrosion produces iron oxides and hydroxides that are larger in size than iron atoms (Fe<sup>0</sup>) in the lattice system ( $V_{\text{oxide}} > V_{\text{iron}}$ ) [79-81]. This evidence means that the volumetric expansive nature of iron corrosion should be considered wherever Fe<sup>0</sup> oxidative dissolution is likely to produce iron oxides/hydroxides. This is when the final pH value is larger than 4.5 [44]. This volumetric expansion should be considered before any other source of clogging is discussed. For ex-situ applications of Fe<sup>0</sup> filters, appropriate pre-treatment can totally eliminate all other sources of pore filling.

In cementation using Fe<sup>0</sup>, the operations take place at lower pH values (pH < 4.0) and there is less or no oxide accumulation [82]. The excess iron consumption is commonly given to characterize the efficiency of the system (1). Clearly, in the cementation reaction the mass balance of the metal ions to be removed (e.g. Cr<sup>VI</sup>, Cu<sup>II</sup>, Pb<sup>II</sup>) and that of iron are given to assess the system efficiency. For Pb removal for example, it is preferable that the Pb concentration of wastewater is low, to avoid clogging (permeability loss) due to the replacement of

Fe by larger Pb in the filters. As discussed in section 1, not considering the iron mass balance has been a brake for the development of Fe<sup>0</sup> filters.

Once the mass balance for both iron (Fe<sup>0</sup>, soluble Fe<sup>II</sup>/Fe<sup>III</sup> and precipitates) and contaminants is properly considered, the mechanism of decontamination can be addressed. At pH > 4.5, hydroxide precipitation is quantitative and contaminant co-precipitation unavoidable [34,35,83,84]. The Fe<sup>0</sup> surface is shielded by an oxide scale which is positively charged [85]. Negatively charged species are preferentially adsorbed but all contaminants are accumulated in the porous system by size-exclusion. Adsorption, co-precipitation and size-exclusion are thus the fundamental mechanisms of contaminant removal in Fe<sup>0</sup>/H<sub>2</sub>O systems [34,35,37,59,86]. The remaining question to answer before designing sustainable is the following: What is the rate of generation of iron hydroxides/oxides? In other words, what is the intrinsic reactivity of the Fe<sup>0</sup> material to be used? The time-dependent changes of its dissolution kinetics is implicitly considered [32].

### The paramount significance of Fe<sup>0</sup> intrinsic reactivity

Fe<sup>0</sup> materials relevant for water treatment are a large array of metallic materials having in common a high amount of iron ( $E^0(\text{Fe}^{\text{II}}/\text{Fe}^0) = -0.44 \text{ V}$ ) in their elemental composition: mostly cast iron and low alloyed steel. Fe<sup>0</sup> materials differ in their:

- Crystallinity, bulk density and porosity (granular, foam, filamentous, sponge)
- Conventional application (iron foam, iron nails, scrap iron, steel wire, steel wool)
- Particle size (mm,  $\mu\text{m}$  and nm)
- Particle shape (filings, plates, shavings, spheres) [11].

It has been demonstrated that none of the chemical or physical characteristics alone can enable the assessment of the intrinsic reactivity [67,68,70,87-91]. Moreover, experiments for characterizing the Fe<sup>0</sup> intrinsic reactivity should last for several weeks [92].

A central question that should be scientifically answered before engineers can accurately select appropriate Fe<sup>0</sup> materials for a specific application is the characterization of the intrinsic reactivity and its time-dependent changes. Again, information on elemental

composition, particle morphology and size, and surface area are not really helpful in characterizing the reactivity [67-70,88,90]. In essence, the intrinsic reactivity is a characteristic of each material and cannot be measured, but just assessed by appropriate procedures. In fact, the efficiency of  $\text{Fe}^0$  for any specific application is a complex cause-and-effect relationship between a large number of operational parameters, including

- i.  $\text{Fe}^0$  reactivity
- ii. Mode of generation and long-term permeability of oxide scales on  $\text{Fe}^0$
- iii. Solution chemistry
- iv. Proportion of  $\text{Fe}^0$  in the reactive zone
- v. Thickness of the reactive zone

There is currently no standard method to characterize the intrinsic reactivity of  $\text{Fe}^0$  and thus facilitate material selection [67,68,70,90]. Btatekue-K and colleagues have summarized the most applicable methods that are additionally frugal in nature [78]. They are contaminant independent as they directly characterize  $\text{Fe}^0$  dissolution (EDTA test) or indirectly the extent of  $\text{Fe}$  hydroxide generation (MB test). These two tests are regarded as the cornerstone on which the design of  $\text{Fe}^0$  filters for safe drinking water provision will be built. The EDTA test evaluates the chemical reactivity of  $\text{Fe}^0$  materials as the extent of iron dissolution in a diluted solution of ethylenediaminetetraacetate (e.g. 2 mM). The suitability of methylene blue (MB) to characterize the intrinsic reactivity of  $\text{Fe}^0$  materials arises mainly from the historical observation by [93] that iron oxide coated sand exhibits a very low adsorptive affinity for MB. Accordingly, using MB is a powerful tool to comparatively characterize the extent of iron oxide generation by various  $\text{Fe}^0$  materials or the same material under various operational conditions.

## $\text{Fe}^0$ filters for universal safe drinking water provision

Providing the whole world with safe drinking water is challenging because efficient, affordable and transferable water treatment systems should be developed and made available at the smallest scale and in decentralized manner [94-115]. With the information given herein, designing a sustainable  $\text{Fe}^0$ /sand filter for a household or a small community becomes a simple routine exercise similar to the one used in designing cementation beds (e.g. for lead removal) [116,162]. Here, there is an excess iron consumption but there is no need to quantify it. Rather the dissolved iron in the effluent should be measured to assess/decide whether further units for iron removal are needed [49,117,118]. Experiments should be performed for (i) beds packed with different available materials (including the same in different particle sizes), (ii) beds of different lengths (e.g. identical columns in series), and (iii) at different flow conditions. The experimental data should be compared by means of the efficiency of decontamination, the pH value, electric conductivity, the iron breakthrough and changes in the hydraulic conductivity. Such tests should use well-characterized real world samples or natural-near waters (e.g. tap water) artificially contaminated with relevant species (e.g. As, F, U).

If such systematic experiments are performed in several laboratories across the world,  $\text{Fe}^0$  will soon be the savior of several communities still lacking safe drinking water while waiting for their governments to bring along appropriate solutions. The next section present ways to test  $\text{Fe}^0$  filters for water defluoridation ( $\text{F}^-$  removal).

## Testing $\text{Fe}^0$ filters for fluoride mitigation

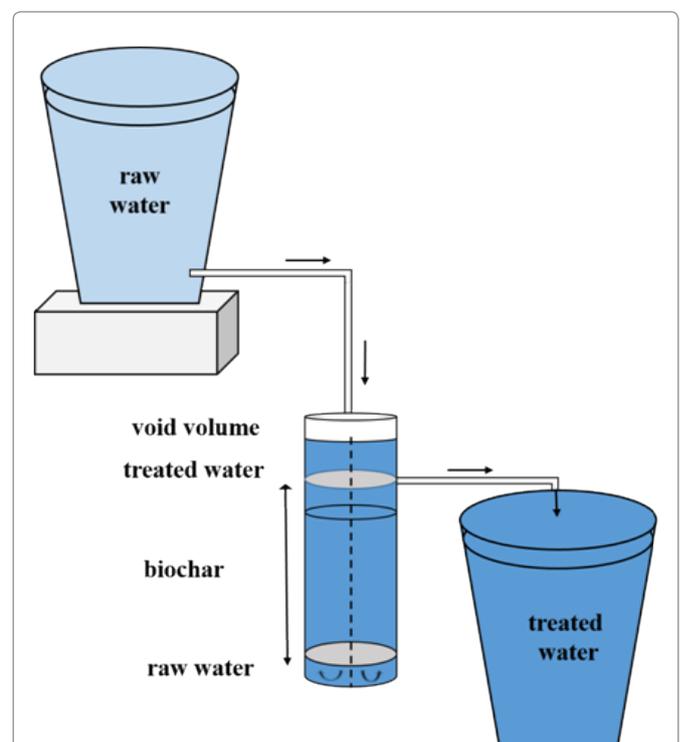
The African Great Rift Valley accommodates the world's most severe fluoride belt [29,119-122]. Here, fluoride mitigation by means of alum (Nalgonda technique) has been used, with limited success since the 1980s [121]. In recent years, packed-bed adsorption of columns filled with bone char have been more successfully implemented for water defluoridation [121,123-125]. This adsorption technique is currently the process of choice for fluoride mitigation in Ethiopia, Kenya and Tanzania [121]. Despite some real

progress, none of the countries has approached universal access to safe drinking water. However the good news is that proven designs are made available that could be modified to supply both households and small communities with safe drinking water. Larger communities can be supplied with several small modules operating in series [118].

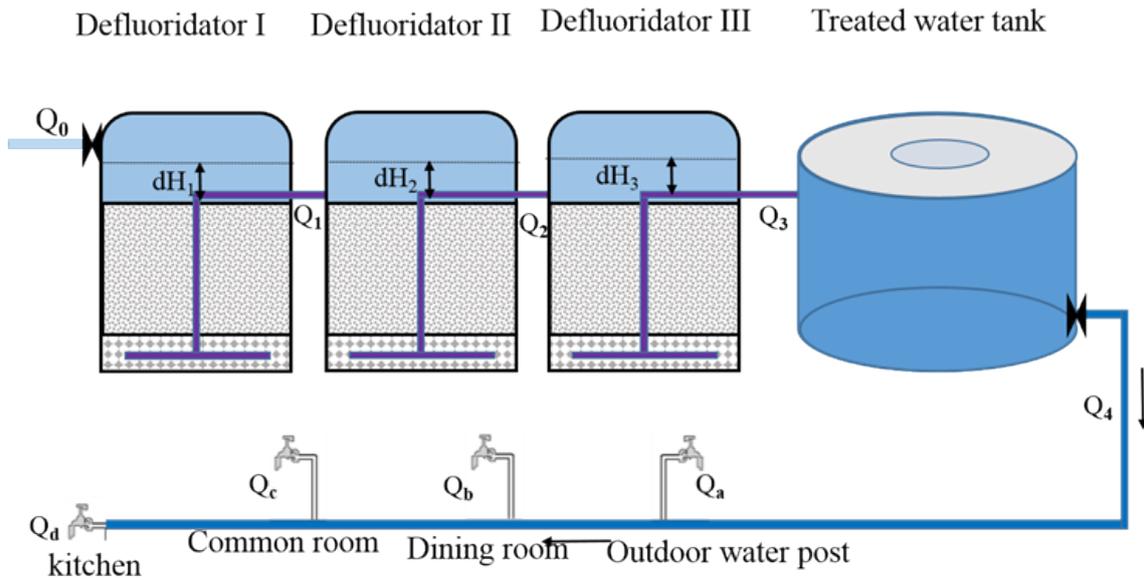
Figure 1 schematically presents a household bone char filter for defluoridation, as commercially available in the Arusha region in Tanzania. Such filters were disseminated to about 2000 families [121]. Dahi [121] also reported on the development and implementation of about 20 different types of larger bone char filters, mainly for use in schools and other institutions (Fig. 2). The drum type filters [126] are easy to recharge and are recommended for pilot testing  $\text{Fe}^0$  filters. Upon successful testing, field water treatment plants can use more robust cement tanks [121,125].

Testing  $\text{Fe}^0$  filters for defluoridation at household (Fig. 1) or at small community (Fig. 2) levels is regarded as a continuation of previous works on the wide field of filtration technology for drinking water and sanitation [49,71,86,89,98-100,114,117,121,125,127,128]. The exceptional stability of fluoride in aqueous solution [129,130] implies that larger depths of  $\text{Fe}^0$  layers (e.g. several columns in series) will likely be necessary to achieve the treatment goal (e.g.  $\leq 1.5$  mg/L for drinking water). The particular properties of  $\text{Fe}^0$  aqueous corrosion imply that sustainable filters operate under anoxic conditions [53,54]. Thus, either a Slow Sand Filter (SSF) is used and the operational flow rate is that of a conventional SSF, or the  $\text{Fe}^0$  filters are preceded with one or several  $\text{O}_2$  scavenging units.  $\text{Fe}^0$  itself is a powerful  $\text{O}_2$  scavenger [131]. However, while using  $\text{Fe}^0$  as  $\text{O}_2$  scavenger, care must be taken to avoid that preferential flow created within the scavenger unit don't disturb the flow regime in the effective  $\text{Fe}^0$  filters.

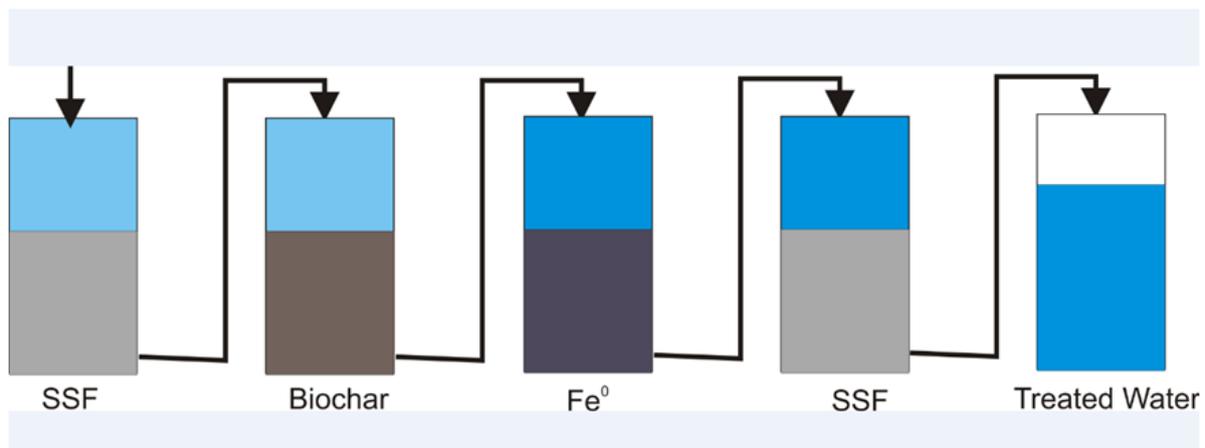
In testing  $\text{Fe}^0$  filters for water defluoridation, a special attention is due to the presence of anions, such as  $\text{Cl}^-$ ,  $\text{CO}_3^{2-}/\text{HCO}_3^-$ ,  $\text{NO}_3^-$ ,  $\text{PO}_4^{3-}$  and  $\text{SO}_4^{2-}$ , as well as natural organic matter [36,59,125]. However, only the concentration range relevant to field situations should be investigated. Ideally, relevant natural waters should be used and



**Figure 1:** Schematic diagram of the domestic bone char filter distributed in the region of Arusha (Tanzania). Here bone char is replaced by biochar. It is expected that  $\text{Fe}^0$ /sand layers are tested in the near future. Considering the intrinsic nature of  $\text{Fe}^0$ , more than one unit can be necessary for efficient water treatment.



**Figure 2:** Schematic diagram of the triple bone char filtration plant installed at an international school in Arusha (Tanzania).  $Q_{i,\alpha}$  is the water flow rate through point  $i$  or  $\alpha$ .  $dH_{i,\alpha}$  is the hydraulic head at point  $i$  or  $\alpha$ . In pilot testing  $Fe^0$ -based filtration plants, both  $dH$  and  $Q$  as well as the corresponding water quality should be routinely monitored.



**Figure 3:** Concept of water treatment train based on filtration on granular materials and including at least one slow sand filter (SSF) for lowering the  $O_2$  level. Further units may comprise biochar filter and  $Fe^0$ /sand filter. Treated water is stored for distribution.

artificially contaminated with  $F^-$  (and other relevant contaminants). As stated in section 3, the  $Fe^0/H_2O$  is very complex and no single solute has a unique mode of impact on the system. For example,  $Cl^-$  is known to enhance corrosion because soluble  $FeCl_2/FeCl_3$  is generated. Accordingly, if the presence of  $Cl^-$  is coupled with a relative high flow velocity, generated  $FeCl_2/FeCl_3$  can be transported out of the system without having time to precipitate. This does not mean that  $Fe^0$  filters is not fundamentally efficient or that iron corrosion is not enhanced. It suffices to reduce the flow velocity (increase residence time) to achieve better results.

Another fundamental aspect is the used  $Fe^0$  material. Porous  $Fe^0$  materials have been shown more efficient than compact ones in  $Fe^0$  filters [5,89,91,99,132]. In essence, a porous  $Fe^0$  is a continuous network of metal co-existing with a network of interconnected voids. The metal is less dense and could be depleted under field conditions. The more the metal is compact the more uncertain is its long-term corrosion behavior. Another important tool that has been used to modify the efficiency (not the reactivity) of  $Fe^0$ /

$H_2O$  systems is alloying [27,41,43,55]. In the context of fluoride removal using electro coagulation, Al-alloyed have shown better efficiency than  $Fe$  electrodes [133]. The similitude of both processes [36,37,134-136] suggests that Al-alloyed materials will be better for fluoride mitigation [137,138]. Coincidentally, such materials have already been developed and used for other contaminants including pathogens [134,135,139,140]. It can be postulated, that one efficient group of materials for  $Fe^0$  filters is an Al-alloyed sponge  $Fe^0$ . Ideally, such a material is comparable to sand in its bulk density (2.5 to 3.0  $cm^3/g$ ) to facilitate homogenous mixture to build the reactive zone. As regarding the pH value, natural waters ( $6.0 \geq pH \leq 8.5$ ) correspond to the range of minimal  $Fe$  solubility [44]. This is the major property making  $Fe^0$  a remediation agent [34,35,43,72]. Wherever necessary, starting water treatment at slightly acidic pH values (e.g. pH 4.0 to 4.5) should be investigated because  $Fe^0$  has the inherent property to enhance the pH value through aqueous corrosion. For small community treatment plants, further filtration through pH-enhancing units (e.g.  $CaCO_3$  or  $CaMg(CO_3)_2$ ) could be envisaged.

Figure 3 gives the general treatment train involving Fe<sup>0</sup> filters. A slow sand filter (SSF) would eliminate pathogens and lower the dissolved O<sub>2</sub> level. A second filter (e.g. biochar) would serve as pre-treatment for the Fe<sup>0</sup> filter. Excess Fe from the Fe<sup>0</sup> filter can be scavenged by a second SSF or a wood charcoal filter. Where necessary, each type of filter can be made up of several filters in series.

### Economic aspects

The bone char technology has been adopted in Tanzania, Kenya, and Ethiopia due to its superiority in comparison with the Nalgonda technique [121]. However, the process costs of the bone char technology are still too high to be affordable by the low income population. Moreover, self-reliance in operation and maintenance, essential for the technology sustainability, are yet to be secured. On the other hand, the bone char has an inbuilt limitation in treating waters with high bicarbonate levels [121,125]. These limitations are the key points on which testing Fe<sup>0</sup> filters will insist.

As clearly discussed elsewhere [49] it is difficult to reliably assess the costs of Fe<sup>0</sup> filters. There are at least three reasons for this:

- i. There is no reference Fe<sup>0</sup> material to which available or new materials could be compared
- ii. The efficiency of Fe<sup>0</sup> filters is contaminant- and site-specific
- iii. The technology is not yet developed to the point where comparison of independent data is possible. Nevertheless, because Fe<sup>0</sup> filters are using the same construction materials like other filtration techniques, while using sand and readily available (and mostly affordable) Fe<sup>0</sup> materials like iron nails, steel wire or steel wool, it is expected that efficient Fe<sup>0</sup> filters will be more affordable than existing filters [79,105,108,141,142]. Two other key points support the likely affordability of Fe<sup>0</sup> filters:
  1. The local availability of the technology of iron making [143]
  2. The recyclable nature of filter materials (iron oxides). Iron oxides are the main raw materials of iron making [49].

Despite the lack of a sound comparison basis, the Fe<sup>0</sup> filtration technology is generally perceived and presented as an affordable technology for low-income communities [21,22,98,99,144-149].

### Concluding remarks

Available information about Fe<sup>0</sup> filter design is insufficient and not organized [9,25,31,32,42,44,46,57,150,151]. A number of previous studies have identified major design flaws [11,25,46] and presented some paths to the next generation efficient and sustainable Fe<sup>0</sup> filters [31,37,49,52,53,71,73,76,86,89,117,118,152,154,155]. There is no doubt on the fundamental appropriateness of Fe<sup>0</sup> filters for safe drinking water provision (and sanitation) [49,153]. Here are just four examples to corroborate the evidence that the open issue is a proper design [31,49,71,152]:

1. Fe<sup>0</sup> filters (containing spongy iron) were efficient at household level before 1881 and were successfully tested at the water works of Antwerpen (Belgium). They were efficient for 18 months without any maintenance [3,8,9].
2. Fe<sup>0</sup> filters (containing steel wool) were efficient at removing radioactive contaminants from waters. Simple filters were designed for two weeks [144]. This can be used as starting point for further investigations [10,117].
3. Fe<sup>0</sup> filters (containing iron filings) were very efficient at removing selenium and several other toxic species from agricultural wastewaters [145]. These filters were not sustainable as they contained 100 % Fe<sup>0</sup> in their reactive zones [11]. In other words, using hybrid systems (e.g. Fe<sup>0</sup>/sand) would have yielded more sustainable systems [10,52-54,72,73,156].
4. Fe<sup>0</sup> filters (containing iron nails) were very efficient at removing arsenic and other toxic species from drinking water mostly in Nepal [98].

Researchers are encouraged to long-term pilot test Fe<sup>0</sup> filters. Ideally, the first design and implementation should be supervised by

scientists. Fe<sup>0</sup> materials should be routinely characterized unless a patented own material is used. In all the cases the efficiency of the systems should be analytically documented [117,157]. It would be beneficial, if the results are made available to a broader audience [158]. It is certain, that Fe<sup>0</sup> alone can solve the long lasting universal crisis of shortage in safe drinking water provision in low-income communities [159,160]. However, combination with other affordable technologies have been recommended [31,144,161,162].

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