Testing Metallic Iron Filtration Systems for Decentralized Water Treatment at Pilot Scale

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Abstract: There are many factors to consider for the design of appropriate water treatment systems including: cost, the concentration and type of biological and/or chemical contamination, concentration limits at which contaminant(s) are required to be removed, required flow rate, level of local expertise for on-going maintenance, and social acceptance. An ideal technology should be effective at producing clean, potable water; however it must also be low-cost, low-energy (ideally energy-free) and require low-maintenance. The use of packed beds containing metallic iron (Fe₀ filters) has the potential to become a cheap widespread technology for both safe drinking water provision and wastewater treatment. Fe₀ filters have been intensively investigated over the past two decades, however, sound design criteria are still lacking. This article presents an overview of the design of Fe₀ filters...
for decentralized water treatment particularly in the developing world. A design for safe
drinking water to a community of 100 people is also discussed as starting module. It is
suggested that Fe⁰ filters have the potential for significant worldwide applicability, but
particularly in the developing world. The appropriate design of Fe⁰ filters, however, is
site-specific and dependent upon the availability of local expertise/materials.

**Keywords:** decentralized water treatment; pilot study; water supply; zerovalent iron

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1. Introduction

1.1. Background

In recent years the use of decentralized water treatment systems has increased worldwide, but
particularly in the developing world [1–10]. Due to their relatively small spatial scale and low carbon
footprint, decentralized water filtration systems have a low environmental impact on water resources
because they do not discharge effluent into waterways [4]. Furthermore, each decentralized water
treatment system can be customized to suit local water quality objectives, climatic and topographic
conditions as well as aesthetic requirements. Conventional water treatment often calls for complex
multistage processes (namely coagulation, disinfection, flocculation, sand filtration, screen-filtration,
ozonization, sedimentation) and requires a wide array of chemicals (namely chlorine, flocculents,
hydrogen peroxide, lime, ozone). The processes therefore also typically require specialist expertise for
installation and maintenance [4,10–13]. As a result conventional wastewater treatment facilities are
often expensive to install and maintain and have a high carbon footprint. This has motivated the
development of alternative “one-step” technologies which include membrane filters (e.g., reverse
osmosis, ultra-filtration), functionalized adsorbents, and ion exchange resins.

Membrane filtration technologies typically exhibit a number of advantageous attributes including [3,7]:
(i) the ability to produce very high quality water (low aqueous contaminant concentrations); (ii) simple
modular design and the ability to be automated; (iii) no requirement for chemicals; and (iv) the ability
to effectively remove bacteria, viruses and other microorganisms. Conventional membrane filtration
can be divided into three stages: pre-filtration (media filtration), ultra-filtration and reverse osmosis.
However, three key disadvantages associated with membrane filtration systems are the high installation
cost, high energy requirement, and the necessity for maintenance (removal of membrane fouling material).
As a consequence, over the past few decades much research has been conducted into the development
of more cost effective and simple water treatment systems for the developing world [3,4,7,12–18].
These efforts include the development of renewable energy powered membrane (RE-membrane)
technologies [19,20]. Resulting water treatment systems are flexibly scalable from devices using individual
modules for household water supply (e.g., Lifestraws and Homespring) to large scale water supplies
for mega-cities [20]. Process selection can be adjusted to any water quality and desired contaminant
removal. The Portable Aqua Unit for Lifesaving (PAUL), also known as Water Backpack [13] is
presented as an example in the following section.
1.2. Membrane Technology Can Be a Bridging Solution

PAUL is a portable membrane water filter (pore size: 20–100 nm) developed at the University of Kassel (Germany) for humanitarian aid [13]. It is designed for the specific purpose of decentralized water treatment in emergency and disaster situations. PAUL functions without chemicals or energy input and during its “lifetime” there is no need for technical maintenance. A PAUL device can produce safe water for up to 700 persons for several months. Furthermore PAUL has been demonstrated as highly effective for the removal of 99.999% of bacteria (Escherichia coli and coliform) and 99.9% of viruses (coliphages). The performance of PAUL and other similar gravity driven membrane filtration systems for the treatment of chemical contaminant species (e.g., hydrocarbons, metals, metalloids, radionuclides), however, has been demonstrated as less effective. PAUL and similar membrane filtration systems are therefore inappropriate for the procurement of potable water in remote communities where such aqueous contaminant species (including arsenic, fluoride, and nitrate) are above minimal threshold levels. Accordingly, there remains a need for the development of alternative and/or complimentary technologies for water treatment. Gravity filtration using Fe\(^0\) has been discussed in the literature as a potential candidate technology due to the ability of Fe\(^0\) to remove microorganisms, degrade organic contaminants and also immobilize metal and metalloid species [14,15,18–27].

1.3. The Suitability of Fe\(^0\) Filters for Safe Water Provision in the Developing World

The idea of using metal corrosion for the in-situ generation of metal hydroxides for aqueous contaminant removal is the basis of electro-coagulation, essentially using Al\(^0\) and Fe\(^0\) as electrodes. Noubactep and Schöner [28] discussed the similarities between decontamination by electro-coagulation and by using granular metallic materials. According to Bojic et al. [29], the great efficacy of voluminous insoluble Al(OH)\(_3\) for aqueous removal of many chemical and microbiological pollutants implies efficient water decontamination by a microalloyed aluminium based composite. Bojic et al. [29] positively tested this idea to eliminate Escherichia coli from a model surface water and later for various chemical contaminants including Cr, Cu, halogenated trihalomethanes, and Zn [30–32].

In an independent approach, Fe\(^0\) was used as a reducing agent or generator of iron hydroxides for water treatment [33–35]. By 2002, Fe\(^0\) had already been tested for many relevant groups of chemical contamination. With the publication of the work of You et al. [36] entitled “Removal and inactivation of waterborne viruses using zerovalent iron”, Fe\(^0\) was demonstrated to be a universal material for water treatment. Since then, a great deal of work has established the potential of Fe\(^0\) for water disinfection [37–41].

The suitability of Fe\(^0\) filters for safe water provision in the developing world arises from their ability to treat chemical and biological contamination. Pathogens, arsenic and fluoride are arguably the three main pollutants of worldwide relevance [42–45]. The ability of Fe\(^0\) filters to remove salt ions (e.g., fluoride) and trace contaminants (e.g., arsenic) makes this technology more suitable than gravity-based membrane technology for deployment in remote areas.

1.4. Fe\(^0\) Filters for Self-Reliance in Water Supply

This article presents a comprehensive overview of the science and rationale for the use of Fe\(^0\) for decentralized water treatment. In particular a working methodology is presented which is intended to
function as a basis for future work; this comprises a hypothetic scenario wherein Fe\textsuperscript{0} is used for the provision of potable water to a community with 100 inhabitants (1 m\textsuperscript{3} water/day). This population size has been selected because it is considered to be one “production module” which can then be scaled upwards for communities of different population. For example, a community with 400 inhabitants would require four production modules.Downwards scaling for communities with less than 100 inhabitants is not discussed but the expertise from the present effort would also enable downscaling.

2. Water Supply Systems

2.1. Centralized Water Supply

Centralized water supply represents the current conventional water supply approach [4,6]. This approach is based on providing water through water supply schemes including components such as: water source development (namely boreholes, rainwater, or rivers), water distribution systems (namely piping systems), water storage systems (namely overhead tanks). A ready source of power supply is needed to run the schemes and a distribution network [4,6,18,46,47]. Individual homes are expected to be connected to the distribution network (Figure 1). Centralized water supply systems are typically available in cities both in the developed and developing world, and small communities in the developed world. In urban areas of the developing world, water is often quantitatively available. However, ineffective water supply chains frequently result in low quality [11,46].

![Figure 1. Concept of water treatment train based on filtration on granular materials and including at least one sand filter for media filtration (roughing filter). Further units may comprise: (i) biosand filter; (ii) activated carbon filter, Fe\textsuperscript{0}/sand filter. Treated water is stored for distribution.](image-url)
2.2. Disadvantages of the Centralized Water Supply

Centralized water supply systems may be prohibitively expensive to install, operate, and maintain for low-income and/or remote communities. This is due to a number of reasons including: (i) intermittent power supply (e.g., lack of fuel); (ii) lack of infrastructure; and (iii) lack of technical knowledge to maintain infrastructure [1,6,11,21]. Accordingly, there is an urgent need to develop affordable yet also low maintenance technologies for the water supply of low-income and remote communities.

2.3. Decentralized Water Supply

Some rural locations are equipped with obsolete water supply systems with an overhead storage tank. In the developing world, however, such small scale centralized water supply systems are fraught with financial and managerial problems [6,11,46]. Boreholes are sometimes available for domestic water supply systems; however, the quality of these and all other water sources (rain, river, source) is not typically monitored [11,48]. Alternative decentralized technologies for the developing world should be small-scale, energy efficient, environmentally sound, and use locally available resources [7]. They should also be capable of being controlled and maintained by the local community.

2.4. Appropriateness of Decentralized Water Treatment Systems

Decentralized water treatment systems have three main unique advantages: (i) the ability to tailor the technology for specific contaminant species; (ii) low cost; and (iii) ability for deployment in remote locations. Ideal systems should be able to produce clean drinking water without power input, or with energy input via renewable sources such as solar power [19,20]. Ideally, the system should also be constructed using local materials and use local technical labor. These requirements exclude the use of chemicals (including chlorine), particularly in low skill communities. The requirement of using endogenous or easily transferable technical skills exclude membrane technology (Section 1.2) as long as used membranes are not locally manufactured [49].

3. Decentralized Water Treatment with Metallic Iron

3.1. An Overview of the Fe\(^0\)/H\(_2\)O System for Contaminant Mitigation

The potential utility of Fe\(^0\) filters for decentralized safe drinking water provision has been intensively investigated over the past 15 years [14–16,18,24,26,27,50–54]. Fe\(^0\) is considered an appropriate material for water treatment because it is a relatively strong reducing agent (\(E^0 = -0.44\) V) and was applied initially to transform recalcitrant halogenated organic compounds into less toxic and/or biodegradable species [55–59]. More recently, Fe\(^0\) was demonstrated as highly effective for the treatment of several other classes of substances, including aromatic nitro compounds, bacteria, heavy metals, herbicides, nitrates, pesticides, radionuclides and viruses [18,60–79]. Fe\(^0\) materials have also been used in Fenton oxidation reactions [71,72,80–82]. It appears from the overview of treatable contaminants that Fe\(^0\) might be regarded as a technology with the potential to manage all classes of contaminants (inorganic ions, organic poisons, and harmful germs). This impression is supported by
articles which reported on quantitative removal of species (e.g., 1,2-dichloroethane, dichloromethane, methylene blue, triazoles) which were proven to be not reducible by Fe⁰ [83–85].

The performance of Fe⁰ materials for water treatment has been adapted in recent years via multiple different methodologies, for a wide range of applications, including: (i) decreasing the particle size to nano-scale in order to enhance the reactivity of the material as a function of mass [86]; (ii) embedding noble bimetallic particles into the Fe⁰ structure in order to improve the galvanic properties of the material [87]; (iii) embedding Fe⁰ into appropriate porous support materials [14,48]; and (iv) embedding Fe⁰ with complimentary adsorbent materials [88–90]. In addition, other metallic elements (namely Al⁰, Cu⁰, Sn⁰, Ti⁰ and Zn⁰) have also been tested as alternatives to Fe⁰ [91–95]. However, Fe⁰ has typically been demonstrated as superior due to its cost-effectiveness, bio-compatibility and long-term reactivity under natural conditions [96,97]. The presentation herein is limited to granular mm- and μm-scale particles (d > 215 μm) [25,98]. Furthermore, the suitability of micron scale Fe⁰ particles arises from the evidence that resulting system (Fe⁰ filters) must be efficient in the long term, with sub-micron scale particles becoming exhausted over relatively short timescales [25,71,72,99,100].

3.2. The Nature of the Fe⁰/H₂O System

It has been demonstrated/recalled that under environmental conditions, the Fe⁰/H₂O interface does not exist [101–106]. Rather, there is a minimum of two interfaces: Fe⁰/Fe-oxides and Fe-(hydr)oxide/H₂O, with the material comprising a “core-shell” structure [80,81,107–112]. Moreover, the (hydr)oxide layer comprises the location for H/H₂ and Fe²⁺ formation which is driven by Fe⁰ corrosion. Given that the outermost (hydr)oxide layer is not typically electronic conductive, extensive chemical reduction of aqueous contaminant species upon this interface does not typically occur [113]. In contrast, chemical reduction of aqueous contaminants due to chemical interactions with H/H₂ or Fe²⁺ has been demonstrated as more likely [114–118]. As a consequence Fe⁰ corrosion is an electrochemical reaction mediated by water (H₂O or H⁺) and resulting in H₂ evolution. However, contaminant reduction, when it occurs, is not the primary coupled cathodic reaction [82,112,118,119].

This clarification coupled to the consideration of the formation of voluminous Fe⁰ corrosion products is the theoretical starting point for the design of next generation Fe⁰ filters [16,17,25,98,100,120–123]. No consideration of these key issues has led to controversial reports which render the assessment of progress in designing Fe⁰ filters difficult. For example, in a recent article entitled “South African sands as a low cost alternative solution for arsenic removal from industrial effluents in permeable reactive barriers”, Trois and Cibati [54] demonstrated the suitability of admixing 25 or 50% (v/v) Fe⁰ with natural sand to treat as contamination. The title of their article is justified by the volumetric abundance of sand but the experimental design did not test any pure sand system (100% sand).

4. Rationale for Fe⁰ Filter Design

At present there are conflicting views with regard to the design of Fe⁰ filters [8,9,54,58,88–90,124–126]. In particular, the usefulness of mixing Fe⁰ and inert (anthracite, pumice, sand) or reactive but non-expansive (Fe₃O₄, MnO₂, TiO₂) materials is still controversially discussed [125,127,128]. Furthermore, an empirical approach has been used to screen selected operational factors like grain sizes and grain size distributions [126], grain packing [129] or the mixture of Fe₃O₄ and external Fe²⁺
solutions [88–90]. This empirical approach is certainly costly but not necessarily effective [121–123]. An alternative approach is to develop the science of the system, which will serve as compass to evaluate experimental results [25,130,131].

A key factor which prevents cross correlation between Fe⁰ water filtration studies is the absence of a standard reference material for Fe⁰ [132]. Accordingly, even results obtained under similar conditions are not really comparable. A methodology to compare the intrinsic reactivity of Fe⁰ materials was introduced [133], and was recently revisited [132,134], however it is yet to receive universal acceptance. On the other hand, a universal design rationale for the design of Fe⁰ filters was presented by Noubactep and Caré [130,131] and progressively revisited [25,98,121–123].

The following text provides the example of a pioneering work by Westerhoff and James [124] and enumerates the lesson that could have been learnt from it. The discussion is limited to relevant design aspects.

4.1. A Non-Exploited Pioneering Work

Westerhoff and James [124] investigated nitrate removal in Fe⁰ packed beds both at lab and field scales. Laboratory columns (\( V = 600 \text{ mL}, D = 5 \text{ cm}, L = 30 \text{ cm} \)) were used. The columns were packed with two different Fe⁰ samples (Fe⁰ is generally termed as zero-valent iron (ZVI)): 1636 g of ZVI1 (6.32 g/cm³) and 2271 g of ZVI2 (7.89 g/cm³). The field columns were packed with ZVI1. The dimensions of the field columns were: \( V = 4000 \text{ mL}, D = 7.5 \text{ cm}, L = 91 \text{ cm} \). While the lab columns contained 100% Fe⁰, a series of field experiments implied a column with a bottom half packed with 50% sand and 50% Fe⁰ to sustain the permeability. The other run had 100% Fe⁰ throughout the column (Figure 2).

![Figure 2. Schematic diagrams of the common design of individual Fe⁰-based filters: (a) pure Fe⁰ (100%); (b) a pre-treatment hybrid zone (e.g., Fe⁰/sand) followed by 100% Fe⁰; and (c) a fully hybrid system. It has now been established that only fully hybrid systems are sustainable because of the volumetric expansive nature of iron corrosion at pH > 4.5.](image-url)
Field experiments lasted for approximately 12 months. During this period, operational problems occurred frequently in the pure Fe\textsuperscript{0} system. The most significant operational problem was a decrease in hydraulic conductivity (permeability loss) over time. Permeability loss was also documented in the hybrid Fe\textsuperscript{0}/sand system, but to a lower extent. The experiments were stopped when the residual value of the hydraulic conductivity was less than 10\% of the initial value (for the hybrid Fe\textsuperscript{0}/sand system). Other operational problems included: (i) air entrainment; (ii) electrical power outage at the site; and (iii) cracks in reactor.

The most important results of Westerhoff and James [124] could be summarized as follows: (i) permeability loss was more pronounced under field conditions (due to a continuous supply of dissolved O\textsubscript{2}); (ii) there was a deficiency in the nitrogen mass balance (co-precipitation or enmeshment); (iii) a large difference in intrinsic reactivity was documented between the two tested Fe\textsuperscript{0} materials; and (iv) from the bottom (influent side) to the top the material in the column exhibited differential compaction behavior. In particular, for the hybrid system (bottom Fe\textsuperscript{0}/sand, top pure Fe\textsuperscript{0}) the three following layers were observed: (i) the bottom 10 cm consisted of highly cemented Fe\textsuperscript{0} and sand; (ii) the intermediate layer (still within the Fe\textsuperscript{0}/sand zone) was black in color and was visually similar to the original Fe\textsuperscript{0} material, but the Fe\textsuperscript{0} particles were irregular in shape; and (iii) the upper pure Fe\textsuperscript{0} layer maintained the irregular shape of the iron but changed to a black-gray color. Despite increased cementation in the influent zone, the same trend was observed in the pure Fe\textsuperscript{0} column.

4.2. Lessons from the Pioneering Work

The observations from the hybrid system of Westerhoff and James [124] correspond to the recent results of Miyajima [135] and Phukan [136]. These authors used a 1:1 Fe\textsuperscript{0}/sand (vol/vol) for methylene blue discoloration in column studies for four and three months respectively and observed that at the influent of the column, about 3 cm was brown colored (Figure 3) and hardly compacted, while the remaining Fe\textsuperscript{0}/sand layer was less or not compacted and black colored. Recent works have recalled that the availability of dissolved O\textsubscript{2} is the major factor causing particle cementation and permeability loss [25,100,122,123]. However, even without this “recent” knowledge, a pragmatic approach would have consisted of comparing the density of Fe\textsuperscript{0} (7.8 g/cm\textsuperscript{3}) and Fe oxides (e.g., β-FeOOH; 3.6 g/cm\textsuperscript{3}) [137]. Because the volume of a filter is constant, iron corrosion is necessarily coupled with a decrease of the pore volume because in-situ generated oxides are less dense or more voluminous than parent Fe\textsuperscript{0}. If Fe\textsuperscript{0} (ZVI) is to be transformed to Akageneite (β-FeO(OH,Cl)), a volume of about 2V\textsubscript{ZVI} is necessary for the reaction to be quantitative.

4.3. Disregarding Lessons from the Pioneering Work

Despite the clear message from Westerhoff and James [124] that pure Fe\textsuperscript{0} systems are not sustainable, research with pure systems has continued. For example, Ruhl et al. [125] recently tested four binary mixtures (Fe\textsuperscript{0}/anthracite, Fe\textsuperscript{0}/gravel, Fe\textsuperscript{0}/pumice and Fe\textsuperscript{0}/sand) for their efficiency in removing trichloroethylene (TCE) in column experiments for up to 200 days. No accompanying Fe\textsuperscript{0} system (reference system) was tested, the work of Westerhoff and James [124] was not considered for the discussion. The authors concluded that “the mixed reactive filters” are not applicable for treatment of the “tested groundwater with its indigenous microorganisms”. This conclusion is erroneous for at least
three reasons: (i) it is unlikely that used Fe$^0$ is depleted within 200 d; (ii) a pure Fe$^0$ system is not tested (no operative reference); and (iii) TCE removal by binary systems containing only 22% Fe$^0$ (w/w) was the system used to demonstrate the feasibility of Fe$^0$ for groundwater remediation [58].

Figure 3. Photograph of a column design depicting the typical sequence of coloration described by Westerhoff and James [124]. It can be seen that the entrance zone of the Fe$^0$/sand zone is colored brown while the upper section is black. The sand layer after the Fe$^0$/sand zone more or less maintains its “white” color or is brownish by Fe$^{III}$ iron oxide from the Fe$^0$/sand zone. Sand in the reference system (first column) and the sand layers preceding the Fe$^0$/sand zone is blue colored by methylene blue (see text).

A second example showing how the results of Westerhoff and James [124] were not properly considered is presented by Huang et al. [88–90]. The authors reported on a new efficient hybridized Fe$^0$/Fe$_3$O$_4$/Fe$^{II}$ system for environmental remediation and water treatment. However, it is not clear from the related works, what makes the system sustainable and why magnetite is the suitable additive. In a system in which Fe$^{II}$ is generated (iron corrosion) in-situ and is suitable at the long-term, the
introduction of external FeII could be regarded as counterintuitive (LeChatelier Principle). In other words, Huang and colleagues have not yet rationalized the functionality of their system.

4.4. Evaluation

The two examples in Section 4.3 and the parameters in Tables 1 and 2 testify that various μm- and mm-scale Fe0 materials have been tested as filter materials for water treatment. Most of the experiments were performed at laboratory scale. Field experiments were reported as well. Some 200 Fe0 permeable reactive barriers (Fe0 PRBs) have been installed worldwide [138–140]. Results from laboratory experiments (controlled conditions) have typically shown promising treatment efficiencies. Results from the field trials, including commercial Fe0 PRBs, have been more various. In particular, failures of Fe0 PRBs [83,141] have not been satisfactorily rationalized [122,123]. It is evident that the composition of contaminated waters and effluents vary to a large extent. This is also one of the reasons why comparison of published results is challenging.

Tables 1 and 2 clearly relate the diversity among Fe0 material tested or used for environmental remediation and water treatment. Although Fe0 has been used in some 200 PBRs, little progress has been made toward characterizing the variability in reactivity among Fe0 samples from different sources [132–134,142,143].

Table 1. Selected operational conditions of column studies presented as examples in Section 4 with regard to their diversities. X is the contaminant of concern and “n.s.” stands for non specified. Generally, diversities of operational conditions including the amount of Fe0 materials and their proportion in hybrid systems render fixation of reported discrepancies between studies difficult.

<table>
<thead>
<tr>
<th>Fe0/Solid Ratio</th>
<th>Column Dimensions</th>
<th>Flow Rate (mL/min)</th>
<th>Duration (Days)</th>
<th>X</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>D</td>
<td>L</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe0 (100%)</td>
<td>5.0</td>
<td>30</td>
<td>variable</td>
<td>weeks</td>
<td>NO3−</td>
</tr>
<tr>
<td>Fe0/sand (3:1)</td>
<td>7.5</td>
<td>91</td>
<td>n.s.</td>
<td>365</td>
<td>NO3−</td>
</tr>
<tr>
<td>Fe0 (100%)</td>
<td>7.5</td>
<td>91</td>
<td>n.s.</td>
<td>365</td>
<td>NO3−</td>
</tr>
<tr>
<td>Fe0/sand (3:1)</td>
<td>16</td>
<td>107</td>
<td>4.2 to 201</td>
<td>0.21</td>
<td>As</td>
</tr>
<tr>
<td>Fe0/sand (1:1)</td>
<td>16</td>
<td>107</td>
<td>4.2 to 202</td>
<td>0.21</td>
<td>As</td>
</tr>
<tr>
<td>Fe0 (100%)</td>
<td>16</td>
<td>107</td>
<td>4.2 to 203</td>
<td>0.21</td>
<td>As</td>
</tr>
<tr>
<td>Fe0/anthracite (n.s.)</td>
<td>2.5</td>
<td>21.5</td>
<td>0.075</td>
<td>200</td>
<td>TCE</td>
</tr>
<tr>
<td>Fe0/gravel (n.s.)</td>
<td>2.5</td>
<td>21.5</td>
<td>0.075</td>
<td>200</td>
<td>TCE</td>
</tr>
<tr>
<td>Fe0/pumice (n.s.)</td>
<td>2.5</td>
<td>21.5</td>
<td>0.075</td>
<td>200</td>
<td>TCE</td>
</tr>
<tr>
<td>Fe0/sand (n.s.)</td>
<td>2.5</td>
<td>21.5</td>
<td>0.075</td>
<td>200</td>
<td>TCE</td>
</tr>
<tr>
<td>Fe0 (100%)</td>
<td>5.1</td>
<td>15.0</td>
<td>n.s.</td>
<td>14</td>
<td>PO4 3−</td>
</tr>
<tr>
<td>Fe0 (100%)</td>
<td>5.1</td>
<td>15.0</td>
<td>0.8 to 1.0</td>
<td>14</td>
<td>As, Cr, Se</td>
</tr>
<tr>
<td>Fe0 (100%)</td>
<td>5.1</td>
<td>15.0</td>
<td>0.8 to 1.0</td>
<td>14</td>
<td>Cd, Cu, Pb</td>
</tr>
</tbody>
</table>
Table 2. Selected characteristic of Fe\textsuperscript{0}-based filter materials used in studies presented as examples in § 4.

<table>
<thead>
<tr>
<th>Material</th>
<th>Availability</th>
<th>Origin</th>
<th>Mass (g)</th>
<th>Mean grain size (mm)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Iron filings</td>
<td>scrap iron</td>
<td>Masterbuilders Inc.</td>
<td>1636</td>
<td>0.05–0.6</td>
<td>[124]</td>
</tr>
<tr>
<td>Iron chips</td>
<td>commercial</td>
<td>Baker Iron</td>
<td>2271</td>
<td>0.5–5.0</td>
<td>[124]</td>
</tr>
<tr>
<td>Iron fillings</td>
<td>commercial</td>
<td>Connelly-GPM Inc.</td>
<td>n.s.</td>
<td>0.08–2.4</td>
<td>[54]</td>
</tr>
<tr>
<td>Iron fillings</td>
<td>commercial</td>
<td>Gotthart Maier</td>
<td>n.s.</td>
<td>1.0–2.0</td>
<td>[54]</td>
</tr>
<tr>
<td>Granulated cast iron</td>
<td>commercial</td>
<td>Gotthart Maier</td>
<td>100</td>
<td>0.3–2.0</td>
<td>[125]</td>
</tr>
<tr>
<td>Zero-valent iron (ZVI)</td>
<td>commercial</td>
<td>Connelly-GPM</td>
<td>n.s.</td>
<td>0.1–2.0</td>
<td>[8, 9]</td>
</tr>
<tr>
<td>Porous iron composite (PIC)</td>
<td>commercial</td>
<td>NA Höganås Inc.</td>
<td>n.s.</td>
<td>0.1–2.0</td>
<td>[8, 9]</td>
</tr>
<tr>
<td>Sulfur modified iron (SMI)</td>
<td>commercial</td>
<td>SMI_PS, Inc.</td>
<td>n.s.</td>
<td>0.1–2.0</td>
<td>[8, 9]</td>
</tr>
</tbody>
</table>

The Fe\textsuperscript{0} literature review has also revealed that it is very hard to normalize the data from different independent studies [144, 145]. In fact, despite an observed linear relationship between the first-order rate constant \(k_{\text{obs}}\) and the specific surface area of Fe\textsuperscript{0}, the introduction of the \(k_{\text{SA}}\)-concept (surface area normalized rate constant) did not enable comparison of experiments obtained by different Fe\textsuperscript{0} types. There are several reasons for this including: (i) the intrinsic reactivity of tested Fe\textsuperscript{0}; (ii) the various physical properties (shape, size, specific surface area) of tested Fe\textsuperscript{0}; (iii) the proportion of Fe\textsuperscript{0} in the system; and (iv) the solution pH and the redox potential. Furthermore, a standard protocol is missing which might enable the comparison of Fe\textsuperscript{0} materials based on their treatment efficiency for a given volume of water, with a given level of contamination, and flow through the Fe\textsuperscript{0}-based filter at a certain flow velocity. Only under such defined conditions can the complexity of processes occurring in Fe\textsuperscript{0}/H\textsubscript{2}O systems be properly addressed.

Summarizing the Fe\textsuperscript{0} literature, it can be observed that despite 20 years of intensive research and numerous field scale treatment facilities, the explanations for contaminant removal from water by Fe\textsuperscript{0} are still in their infancy [25]. Drawbacks have not been reported to a large extent, attention seems to have been focused on success stories [118, 122, 123]. The use of Fe\textsuperscript{0} as filter materials for water treatment is necessarily connected to two major “drawbacks” [41]: (i) “reactivity loss” or non-linearity of Fe\textsuperscript{0} corrosion; and (ii) permeability loss resulting from the loss of interconnectivity of the initial pore space \(\text{in-situ}\) generation of “cementing” agents). In essence, both “reactivity loss” and permeability loss are inherent to Fe\textsuperscript{0} filtration for water treatment. This implies that both apparent “disadvantages” have occurred (to different extents) at success sites. Therefore, the way forward is further systematic research. Such a collaborative research path has recently been initiated in our laboratories. Achieved results are summarized in the following section.

4.5. Rationally Designing Fe\textsuperscript{0} Filters

A critical literature review (Sections 4.1–4.4) has revealed unsatisfactory aspects related to the design of Fe\textsuperscript{0} filters. It has also shown that, based on Westerhoff and James [124], the Fe\textsuperscript{0} system design could have been given more attention during the past 10 years. This approach would have been beneficial for the procurement of new systems as well as the modification and operation of existing systems (e.g., three-pitcher household filter). It is of vital importance that these systems are designed
according to the intrinsic properties of Fe\(^0\) [101,104,108,109], and known principles for designing conventional granular filters [146–148]. Moreover, the intrinsic properties of contaminants should be considered because classifications like “organic contaminants (e.g., dyes, pesticides, and pharmaceuticals/drugs)” or “industrial organic wastes (e.g., phenols and aromatic amines)” say nothing about the chemical reactivity or the affinity of the species of concern for the Fe\(^0\)/H\(_2\)O system.

The realization that aqueous contaminant removal in the presence of Fe\(^0\) (Fe\(^0\)/H\(_2\)O system) is not a property of the reduction by Fe\(^0\) but a characteristic of corroding Fe\(^0\) [115,149–151] was decisive for the rational design of Fe\(^0\) filters. In other words, contaminant removal is not mediated by reductive degradation or reductive precipitation but by the interactions between dissolved contaminants and primary (Fe\(^{II}\), H/H\(_2\)), secondary (Fe\(^{II}/Fe^{III}\)-hydroxides/oxides) and tertiary (Fe\(^{III}\)-hydroxides/oxides) products of Fe\(^0\) oxidative dissolution. Here, Fe\(^0\) is oxidized by water (H\(^+\) or H\(_2\)O). This observation corresponds to reports of several researchers [33,35,152–155] with the subtle but decisive difference that reduction (if applicable) is subordinated to adsorption, co-precipitation, and size-exclusion.

Clearly, under experimental conditions, there is reduction in the presence of Fe\(^0\) (in Fe\(^0\)/H\(_2\)O systems) but not by Fe\(^0\) (no direct reduction).

The finding that direct reduction (reduction by Fe\(^0\)) is of secondary importance for the process of contaminant removal in Fe\(^0\)/H\(_2\)O systems implies that observed reduction was mediated by primary corrosion products (Fe\(^{II}\) and H/H\(_2\)) and that contaminants are removed by adsorption, co-precipitation, and size-exclusion. This finding was followed by a theoretical work discussing the suitability of using hybrid systems instead of pure Fe\(^0\) layers [120,130,131]. Results challenged the still popular view, that mixing Fe\(^0\) with non-reactive materials (e.g., pumice, sand) is a chance to save Fe\(^0\) costs while satisfying width requirements [125,127,128]. Rather, it can be demonstrated that mixing Fe\(^0\) and a non-expansive aggregate is a prerequisite for sustainable Fe\(^0\) filters, the most sustainable system containing 25% Fe\(^0\) (v/v) [98,100,121]. These theoretical results have been experimentally validated [135,156–159].

The ion-selective nature (charge exclusion) of Fe\(^0\)/H\(_2\)O systems was also demonstrated using methylene blue (MB) as operational indicator [135,156,158,159]. The suitability of MB for this purpose arises from the fact that MB (cationic) has a very low affinity for iron hydroxides/oxides (pH\(_{pzc}\) > 6.0) covering the surface of Fe\(^0\) in Fe\(^0\)/H\(_2\)O systems. Under these conditions, the most reactive system is the one quantitatively producing iron oxides, or the one in which “early” MB breakthrough is observed [132]. From Figure 3, a summary of the behavior of the Fe\(^0\)/sand/MB system can be read as: (i) negatively charged sand is an excellent adsorbent for MB; (ii) in the Fe\(^0\)/sand zone, MB discoloration is first “disturbed” by Fe\(^{II}\) and Fe\(^{III}\) ions (concurrence for negatively charged sand surface), and later by ion oxides, coated in-situ on sand [135,158]; and (iii) the preferential flow created in the Fe\(^0\)/sand zone makes the sand layer upwards not fully available for MB discoloration (no blue coloration).

This hypothesis from MB discoloration (charge exclusion) was further confirmed/validated using two anionic dyes: Orange II and Reactive Red 120 [136,160]. Using MB as operative indicator has solved the long lasting problem of reducing the experimental duration while achieving reliable results [113,161–166]. When using up to 100 g Fe\(^0\) packed in fixed beds, it has been clearly demonstrated that reliable results could be obtained only after two months. This should be regarded as the absolute minimal duration of a column study. Table 1 recalls that larger Fe\(^0\) amounts have been typically used for sometimes shorter experimental duration.
The demonstration that (i) the Fe$^0$/H$_2$O system is ion-selective and (ii) the evidence that only hybrid systems are sustainable are the cornerstones for the design of better laboratory and field Fe$^0$ filters. This issue is addressed in the following section. The ion-selective nature of the Fe$^0$/H$_2$O system suggests that available literature on the interaction of iron oxides/hydroxides with contaminants should be considered in designing Fe$^0$ filters. In other words, classifications like “heavy metals”, “industrial organic wastes”, “organic contaminants” or “personal care products” are not really useful. As an example, a recent review on dyes adsorptive removal [167,168] reveals that the science of dye interaction with metal oxides (including iron oxides) dates back to the years 1951–1970. Since then, it has been established that there are three classes of dye: anionic, cationic and non-ionic, behaving differently with various metal oxides and hydroxides [169–176]. The main open question for the Fe$^0$ research is how to correlate available knowledge on contaminant adsorption on iron oxides and the fact that these adsorbents are progressively generated in-situ in Fe$^0$/H$_2$O systems. A further question would be when the generation of iron oxides stops or when available Fe$^0$ will be completely exhausted in the filters. Answering this question would enable the prediction of service life of Fe$^0$ filters.

5. Designing Fe$^0$ Filters for On-Site Water Treatment

The Fe$^0$ amount necessary to treat a given volume of water depends on five main parameters: (i) the nature of the contaminant (solubility, affinity to iron corrosion products); (ii) the contamination level; (iii) the Fe$^0$ intrinsic reactivity; (iv) the water flow velocity and the resulting interaction time as well as (v) the water chemistry (including pH value and hardness). The first intrinsic issue associated with Fe$^0$ is that its physical structure and chemistry constantly changes during aqueous corrosion. This inherent difficulty makes stoichiometric determinations between Fe$^0$ and aqueous contaminant species extremely challenging, i.e., it is difficult to define operational parameters such as adsorption capacity which is a trivial process for non-reactive adsorbents [13,177–179].

5.1. Modular Fe$^0$ Filter Design

The present paper intends to provide rationale for the design of a pilot plan in which a Fe$^0$ filter operates as an independent treatment unit, similar to the pioneering work by Westerhoff and James [124]. The scientific basis for this design is summarized in Noubactep et al. [121] and Rahman et al. [98]. The basic treatment system should consist of a modular series of treatment processes and includes in the following sequence (i) roughing filters; (ii) slow sand filters (SSF) and (iii) Fe$^0$/sand filters. Fe$^0$/sand filters could also be followed by SSF or iron removing filters for maximum effluent water quality. As stated above, the goal is to build a module for the treatment of drinking water for 100 people. The World Health Organization estimates that the average daily water requirement for drinking and cooking per person is 7.5 L, which equates to 750 L for 100 persons. The system presented herein is therefore designed to treat a minimum of 1000 L (1 m$^3$) water per day.

It should be explicitly stated that slow sand filter (SSF) are currently more or less successfully used for drinking water provision at household and small community levels [54,120]. However, sand itself does very little in cleaning water contaminated with organic and inorganic species (chemical contamination) and the removal of biological contamination is not resolved to this day. In essence, sand filters in study [180] work only when the groundwater to be cleaned is Fe$^{II}$-rich. This Fe$^{II}$ is
oxidized to Fe$^{III}$ and derived Fe$^{III}$-(hydr)oxides *in-situ* coat sand grains making it suitable for the removal of contaminants such as As [50]. The effort presented herein could be regarded as an attempt to improve SSF.

5.2. Appropriateness of Fe$^0$ Filters

An implicit shortcoming for Fe$^0$/sand filters is that Fe$^0$ corrosion by natural water cannot be accelerated “on request”. Positively tested approaches to accelerate Fe$^0$ corrosion include (see Section 4.1)

(i) the reduction of the particle size down to nano-scale [86]; (ii) using porous materials [8,9,14];
(iii) using electro-dissolution and internal electrolysis [27,181]; (iv) using oxidizing agents like H$_2$O$_2$ or O$_3$ [24,52]; (v) using multi-metallic systems, including sulfur modified iron [8,9,27,69]; (vi) using minerals with the potential to act as Fe$^{II}$ scavengers (MnO$_2$) [159,182]; and (vii) controlling the O$_2$ level [124]. Of these approaches, only those implying no chemicals and no technical skills are suitable for small communities (in the developing world). These are: using porous materials, using multi-metallic systems, using minerals like MnO$_2$, controlling the O$_2$ level, and combinations thereof.

In preliminary studies, the initial water flow rate should correspond to that of a slow sand filter. This initial flow rate will be stepwise increased to take the maximum advantage of microbial processes in the SSF and chemical and physical processes in the Fe$^0$/sand filters.

5.3. Improving Available Designs

A compilation of literature data [24,121,124,183], suggests that an effective pilot system is likely to comprise: (i) a series of polyethylene tanks ($\geq 1200$ L) installed for instance beside a municipal water treatment plant (raw water storage); (ii) a series of roughing filters; (iii) a series of slow sand filter (SSF); (iv) a series of Fe$^0$/sand filters, eventually (v) a series of filters for the removal of Fe escaping from the Fe$^0$/sand filters; and (vi) a second series of polyethylene tanks to collect and store drinking water. The number of filters in each series depends on the quality of the raw water and the quality of potable water to be delivered to the community. In a pilot study, after each treatment step, the quality of water should be monitored.

As a starting point, a modified field column of Westerhoff and James [124] can be applied: $V = 4400$ mL, $d = 7.5$ cm, $L = 100$ cm. Each Fe$^0$/sand column comprises a 70 cm reactive layer sandwiched between 2 layers of gravel or fine sand (each 15 cm thick). Roughing filters and slow sand filters are filled in the same manner but the reactive layer (70 cm) is replaced by gravel and fine sand respectively (Figure 4).
Figure 4. Concept for the configuration of multi-barrier water treatment systems: (a) the sustainable biochar-based system; and (b) a comparable Fe\textsuperscript{0}-based system. Adapted from Kearns, 2012 [184]. A comparison of the cleaning efficiency of any material to Fe\textsuperscript{0} should be performed by replacing the Fe\textsuperscript{0}/sand unit by the corresponding unit(s) containing the material of concern. This approach will enable the extension of the multi-barrier concept for efficient water treatment.

5.4. Ways for Efficient Fe\textsuperscript{0} On-Site Water Treatment Plants

Section 4.4 demonstrated that a minimum of six variables (Fe\textsuperscript{0} intrinsic reactivity, Fe\textsuperscript{0} shape and size, Fe\textsuperscript{0} content (amount and proportion), solution pH, and the redox potential) have been shown to have an important impact on the decontamination efficiency of Fe\textsuperscript{0}/sand filters. This makes comparisons and correlation of available data difficult. However, a profound analysis of the fundamental reactions
involved, together with some recently obtained design criteria, bring out a number of important considerations which may simplify design efforts. They can be summarized as (i) use only volumetric Fe⁰ ratios ≤ 50%; (ii) characterize the intrinsic reactivity of used Fe⁰; (iii) use as little Fe⁰ as necessary for reliable observations; and (iv) avoid too short experimental durations.

Testing Fe⁰ materials at pilot scale (a given model water) can be summarized in the following: (i) test several well-characterized Fe⁰ materials; (ii) test several Fe⁰/sand ratios (Fe⁰ < 50%), for a given Fe⁰ material and a Fe⁰/sand ratio; (iii) test the number of each unit for satisfactory water treatment; (iv) insert wood charcoal units before Fe⁰/sand units; (v) partly or totally replace sand by porous materials including: anthracite, gravel, MnO₂ and pumice.

At each site and for each design water samples should be analyzed on a weekly basis. Besides the concentrations of relevant contaminants (including pathogen indicators), the Fe level, the turbidity and the pH values should also be monitored. The hydraulic conductivity should also be recorded. This ambitious program implies the equipment of water laboratories wherever pilot scales are planned: ideally in any country water contamination is potentially a problem. Clearly the whole developing world should be equipped with good water laboratories. At the end of the field operation, the Fe⁰/sand column should be dismantled and thoroughly characterized. In this effort the seminal work of Westerhoff and James [124] can be used as guide.

5.5. Comparing Fe⁰ Filters to Other Technologies

The constancy that the nature of Fe⁰ filters has not been properly considered in filter design efforts implies the necessity to revisit the approach to (i) compare Fe⁰ and other affordable filter materials for water treatment [185,186]; and (ii) implement Fe⁰ in a combination of treatment methods for more efficient systems [187,188]. The presentation above has already shown that O₂ scavengers including SSF should precede Fe⁰/sand filters. The situation of multiple contaminant mixtures which may not be efficiently treated by a single technology [189] is an opportunity to compare Fe⁰ filtration to other available affordable filtration systems [190] and shape multi-barrier systems involving Fe⁰ filters. Contaminants possess different properties with respect to adsorptive behavior, degradation potential, molecular size, solubility, and surface charge [191]. Fe⁰ filters are ionic selective in nature because at neutral pH values, the surface of iron oxides is positively charged [160,191]. This means that negatively charged contaminants are readily removed in Fe⁰ filters. Because multiple contaminant mixtures may contain positively charged species as well, it is important to comparatively test Fe⁰ and other materials under the same operational conditions. Relevant filter materials include activated carbons, bark, bio-carbons (biochar), wood charcoal, calcareous shale, chitin, chitosan, commercial ion exchangers, dairy manure compost, dolomite, fly ash, lignite, limestone, olivine, peat, rice husks, steel slag materials, vegetal compost, yeast, and zeolites [190,192].

Comparing Fe⁰ filters to other filtration systems is continued by replacing Fe⁰/sand units in a treatment train as described in Section 5.3 by a unit filled with the material to be tested (70 cm per unit). Here the materials are not compared based on their mass or their chemical reactivity, but on the efficiency of a 70 cm packed filter. Results of such an experiment give a strong basis for the design of multi-barrier systems involving Fe⁰/sand units.
5.6. Economic Considerations

The economy of Fe$^0$/sand filters has already been discussed. According to Gottinger et al. [24,52], Fe$^0$/sand filtration for removal of As and U is economically feasible for small-communities. The total treatment cost is less than $0.01/L water in their modular treatment train, including filter installation, media, operation and maintenance, and disposal. The system presented herein should be less expensive because no ozonization and no aeration are needed.

The mass of iron needed for each Fe$^0$/sand unit can be estimated using the data of Westerhoff and James [124]. Here, 2271 g of the densest Fe$^0$ material fill 600 mL with a porosity of 40%. Assuming the same porosity, the volume corresponding to $L = 70$ cm reactive zone can be calculated using Equation (1):

$$V = L \cdot S = \pi \cdot L \cdot D^2 / 4$$

where $S = \pi \cdot D^2 / 4$ is the cross section of the column with a diameter $D = 7.5$ cm. The calculations yield a volume of 3094 cm$^3$ or 3094 mL. The corresponding Fe$^0$ mass is given by the rule of proportion like Equation (2):

$$m = 3094 \times 2271 / 600$$

The calculations yield a mass of 11,710 g or 11.71 kg of Fe$^0$ for a 100% Fe$^0$ unit. For a 1:1 Fe$^0$ to sand mixture, just 5.86 or about 6 kg of Fe$^0$ is need. If it is further supposed that 4 Fe$^0$/sand units will be needed and the system will operate for one year, then a 100-inhabitant-community may need just 24 kg Fe$^0$ for one year.

In addition, having good water and the know-how to produce more at low cost or low money expense, small communities have the possibility to commercialize excess water. That is some 250 L per day for the model community of this study can be bottled and offered to consumers. In other words, Fe$^0$/sand filters are not only affordable for communities in need; they are also a potential source of income. Business models for commercialization of treated water in the developing world have already been presented [7].

5.7. Implementation of Fe$^0$-Based Water Treatment Plants

There are important aspects of the implementation of Fe$^0$ filters that should be pointed out before sending a construction crew to the field. The maintenance of the Fe$^0$ filters is not addressed herein as it will be considered during pilot testing under real life conditions. In recent years, there have been numerous pilot plant testing for water filters, partly involving Fe$^0$ [24,26,27,52,124,184,193–195]. Construction materials (ceramic, concrete, metallic, plastic) are numerous and include: (i) plastic tanks (e.g., polyethylene); (ii) modified shipping container; (iii) ferro-cement tanks and (iv) polyvinylchlorid (PVC) tubing. Fe$^0$ materials can be commercially obtained. In general, manufacturing of Fe$^0$ filters is comparable to other existing filters, making it a priori a feasible technology. This section enumerates the materials used by Kearns [195] and adaptable for Fe$^0$ filters.
5.7.1. Siting

Water is ideally moved by gravity. The water system is sited at lower elevation than the source water and at higher elevation than the location(s) where treated water will be used. This circumstance enables completely passive operation of the treatment system and very simple control using only a float valve. When water is withdrawn from the storage tank (Figure 4a) the water level in the system drops, opening the float valve. When the system is full, the float valve closes.

5.7.2. Containment

The Fe⁰-based water treatment system presented herein is an open architecture which could be constructed, modified, adapted, and improved on a site-specific basis. Filters containers can be locally built, for example from stackable prefabricated concrete rings commonly used for tank construction. Commercially available plastic tanks can be also modified and used. Appropriately skilled masons can construct custom ferro-cement tanks. In this case the dimensions should enable facile filling of filter media and routine maintenance including the removal/replacement of all materials.

Some cover material (lid) should be used to exclude sunlight and inhibit the growth of photosynthetic microorganisms (algae, cyanobacteria) in the system. Tank tops should be wrapped in fine mesh screening to prevent entrance of insects, bird droppings, leaves, and bits of debris, etc. into the system.

5.7.3. Plumbing

PVC pipe is ubiquitous and cheap in most locations. The most suitable diameter for most connections to and from the water system and between the tanks should be used. Plumbing in the bottom of filter tanks should be protected from physical damage and blockage by under drains made from rock and coarse gravel at least 20 cm in depth.

5.7.4. Filter Materials

Standard gravel (1.0–4.0 cm) should be used for the roughing filter. Standard fine sand (0.15–0.35 cm) should be used for the sand filter [194]. The sand to be used in the reactive zone (Fe⁰/sand) should be coarser (e.g., 0.5–2.0 cm) and of comparable particle size. Fe⁰ should be abundantly available and selected for its appropriate reactivity.

5.7.5. Implementation Plan

Whichever materials are tested at pilot scale, there is always the possibility to find cheaper options. A pilot plan should be able to be implemented in a single phase. Firstly the tanks, piping materials and other equipment listed need to be sourced, bought and transported to the pilot site. The transport can be made by any common trailer truck. Once at the site, it will be relatively simple to build, operate, and monitor the Fe⁰-based plant for at least six months.
6. Concluding Remarks

The need for a scientific-based approach to design and evaluate the efficiency of Fe\textsuperscript{0} filters is corroborated in this study. The urgency of such a consensual approach is evident as it has been established herein that the required knowledge (science of aqueous iron corrosion) is available, but has not been considered in the right way. All is needed is a systematic holistic approach enabling the characterization of aqueous iron corrosion as influenced by the water chemistry, including the pH value, the presence of contaminants and co-solutes. The present study intends to create/initiate a synergy among researchers who are working for more efficient Fe\textsuperscript{0} filtration systems for decentralized water treatment. The adoption or at least the general consideration of fundamental aspects presented herein would accelerate the understanding of the operating mode of Fe\textsuperscript{0} filters and thus, outline the strengths and limitations of this still innovative, but potentially highly efficient technology.

This study specifically restricted its attention to aspects relevant for filter design and the evaluation of their performance in long-term experiments, in particular at pilot scale. The specific objectives of the paper were to make recommendations based on the current state of the science concerning (i) aqueous iron corrosion and (ii) contaminant removal by iron oxides/hydroxides in the environment. The achievement can be summarized by a number of questions, including the following: (i) Which Fe\textsuperscript{0} materials are suitable for water treatment (intrinsic reactivity, porosity, size)? (ii) What level of dissolved O\textsubscript{2} is needed for sustainable systems? (iii) How can one warrant the required level of dissolved O\textsubscript{2}? (iv) What is the general filter design (depth of filter, layering arrangement with particles of different sizes, compaction of the media during construction)? (v) Is it feasible to classify chemicals according to their affinities to the Fe\textsuperscript{0}/H\textsubscript{2}O system? (vi) What types of data are needed for the evaluation of chemicals (molecular size, molecular weight, solubility)? (vii) How does one proceed when data for critical inputs are missing (molecular size)? (viii) Are transformation half-lives of any realistic significance in assessing the relative removal efficiency of species in Fe\textsuperscript{0}/H\textsubscript{2}O system? (ix) Which other treatment units, beside sand filters (e.g., activated carbons) are necessary to optimize the efficiency of the system (on a site specific basis)? (x) What is the optimal frequency of filtration events (intermittent filters).

If future experiments are performed with these ten questions in mind, it will soon be possible to establish the science of “Fe\textsuperscript{0} for environmental remediation” based on the science of aqueous iron corrosion and knowledge on the interactions of iron oxides and hydroxides with aqueous species.

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Author Contributions

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Conflicts of Interest

The authors declare no conflict of interest.

References


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