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*Communication*

# $Fe^0/H_2O$ Filtration Systems for Decentralized Safe Drinking Water: Where to from Here?

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**Abstract:** Inadequate access to safe drinking water is one of the most pervasive problems currently afflicting the developing world. Scientists and engineers are called to present affordable but efficient solutions, particularly applicable to small communities. Filtration systems based on metallic iron ($Fe^0$) are discussed in the literature as one such viable solution, whether as a stand-alone system or as a complement to slow sand filters (SSFs). $Fe^0$ filters can also be improved by incorporating biochar to form $Fe^0$-biochar filtration systems with potentially higher contaminant removal efficiencies than those based on $Fe^0$ or biochar alone. These three low-cost and chemical-free systems ($Fe^0$, biochar, SSFs) have the potential to provide universal access to safe drinking water. However, a well-structured systematic research is needed to design robust and efficient water treatment systems based on these affordable filter materials. This communication highlights the technology being developed to use $Fe^0$-based systems for decentralized safe drinking water provision. Future research directions for the design of the next generation $Fe^0$-based systems are highlighted. It is shown that $Fe^0$ enhances the efficiency of SSFs, while biochar has the potential to alleviate the loss of porosity and uncertainties arising from the non-linear kinetics of iron corrosion. $Fe^0$-based systems are an affordable and applicable technology for small communities in low-income countries, which could contribute to attaining self-reliance in clean water supply and universal public health.

**Keywords:** appropriate technology; bio-sand filter; developing world; household water filters; zero-valent iron

## 1. Introduction

The world's ambition to achieve universal access to safe drinking water by 2030 in the framework of the United Nations Sustainable Development Goals (UN SDGs) [1] relies on the ability of engineers and scientists to effectively translate existing knowledge into practical solutions [2–4]. Several technical solutions are available, but many of them require the following: (i) A good understanding of the decontamination processes, (ii) high level of maintenance, (iii) synthetic chemicals, and (iv) well-trained operators [5]. These requirements are not feasible in low-income communities, where the lack of access to safe drinking water is the most severe. A survey of the literature on using metallic iron ($Fe^0$) for water treatment suggests that this material is a viable alternative for small communities [5–8].

$Fe^0$- and $Fe^0$/biosand-filters effectively remove organic, inorganic, and indicator and pathogenic organisms, including bacteria and viruses in aqueous systems [9–15]. According to literature, microbiological organisms are removed via irreversible adsorption and inactivation by iron corrosion products [9]. $Fe^0/H_2O$ systems also remove toxic metals and their ions (e.g., Zn, Pb, chromate), radionuclides (e.g., U) and metalloids (e.g., As and Mo) in aqueous systems [16–26]. Several detailed reviews exist on the application of $Fe^0$ and its composites for environmental remediation [27–38], including those by the current authors [39–47].

$Fe^0$ has been used in household water filters since the 1850s [48]. The ancient development of household $Fe^0$ filters culminated in the 'spongy iron' filters (Figure 1) [48–54] that has been up-scaled and successfully tested at large scale in 1883 [51,53,54]. The large-scale 'spongy iron' filters (Bischof Process) could secure water supply of the city of Antwerp (200,000 inhabitants) for 18 months before clogging occurred [50]. The clogging problem was solved by using $Fe^0$ as a coagulant generator in a revolving purifier, which can be considered as a continuous flow reactor [52–54]. However, the fact that $Fe^0$ grains are continuously mixed with the water due to the rotation of the revolving purifier suggests some semblance of a batch system. Water from the revolving purifier was subsequently filtered through sand filters (Anderson Process). Recently, several research groups have independently rediscovered both the Bischof and the Anderson processes [8,55–59]. In particular, the SONO filters using a porous composite iron matrix (CIM) corresponds to the household 'spongy iron' filters (Figure 1) [55], while the community $Fe^0$ filters successfully used in West Bengal [5,60–62] corresponds to the Anderson Process. Flaws in designing $Fe^0$-based systems have been reported and updated during the past decades [63–66]. These fundamental flaws include: (i) The misconception that contaminant removal is a reductive process occurring via the cathodic reaction of electrochemical oxidative dissolution of $Fe^0$ [64], and (ii) failure to consider the volumetric expansive nature of iron corrosion during the design of $Fe^0/H_2O$ filtration systems [39,45]. There is actually no comprehensive review article tailored on using $Fe^0$-based systems for small communities. The present communication is not aimed at filling this gap, but rather, to demonstrate how better systems can be designed. This is achieved by discussing the design of two success stories: (i) The SONO filter at household-scale [19,67,68], and (ii) the IITB filter at the community-scale [5,60–62].

The presentation until now attests a 170-years-old technical expertise on using $Fe^0$ for safe drinking water provision both at household and community levels. This suggests that $Fe^0$ can help achieving the UN SDGs by translating existing knowledge into practical solutions [3,4]. However, the science of the $Fe^0/H_2O$ system, although established by Khudenko [69] and independently demonstrated by Noubactep and colleagues [70–80], has not been properly considered in designing $Fe^0$ filters [80–82]. In particular, the volumetric expansive nature of iron corrosion, which is the most useful property in contaminant remediation (i.e., generation of contaminant scavengers such as iron corrosion products), as well as a major cause of clogging, has not been properly considered [83,84]. Similarly, the non-linear kinetics of iron corrosion has not been adequately addressed in $Fe^0$ remediation technology [78,84].

While a number of reviews exist on $Fe^0/H_2O$ filtration systems [8,17,27,30–32,35,36,41–44,46], currently missing is a framework outlining how research on such systems should progress in the next decade. Therefore, rather than providing a comprehensive review, the purpose of the current communication is two-fold: (i) To highlight the need to address existing flaws in the design of $Fe^0/H_2O$ systems, particularly the misconceptions on contaminant removal mechanisms, and failure to consider the volumetric expansive nature of Fe corrosion, and (ii) to propose a framework and timeline on how future research can advance the $Fe^0$ technology and its potential contribution to achieving the SDG on universal clean drinking water provision within the next decade (i.e., by 2030). To achieve this, the current communication proposes a shift from the pragmatic approach currently used to design $Fe^0/H_2O$ filtration systems, to a framework based on a fundamental scientific understanding of $Fe^0$

reactivity and contaminant removal mechanisms. These two fundamental aspects have been often overlooked in existing literature on the design of $Fe^0/H_2O$ filtration systems.

Therefore, the objective of this article is to critically evaluate the recent literature on using $Fe^0$ for safe drinking water provision in order to identify flaws in system design, and recommend ways for better investigations. Suggestions are made to couple $Fe^0$ and other affordable materials such as biochars and conventional bio-sand filters in order to develop more robust and efficient water treatment systems. It is expected that, by using low-cost filter materials ($Fe^0$, biochar, sand) and appropriate designs, this would enable the achievement of universal safe drinking water during the remaining 12 years (to 2030). The term 'universal' entails the provision of clean drinking water meeting the acceptable standards such as the WHO guidelines to all people, including those in low-income countries.

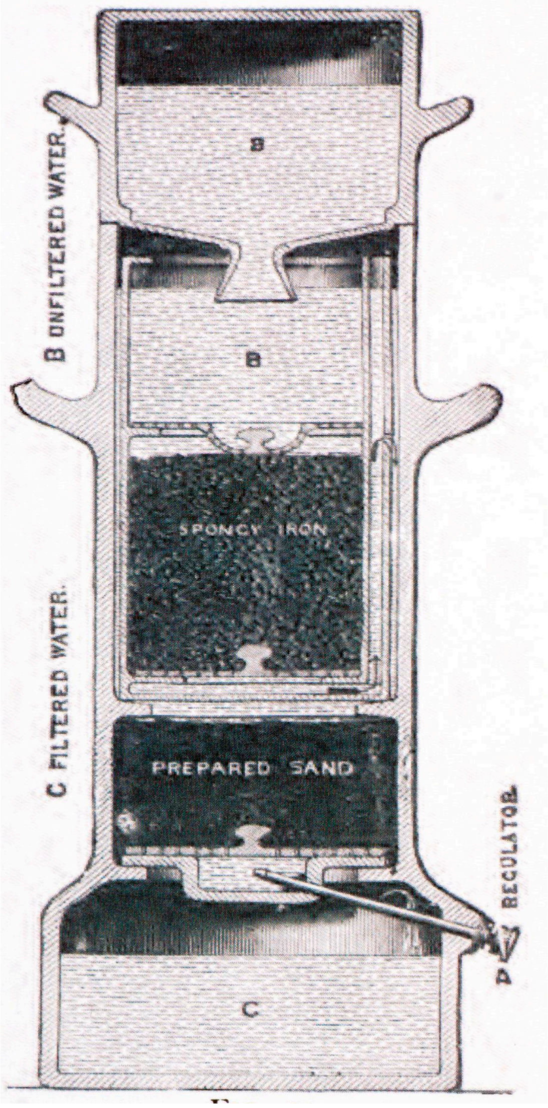

**Figure 1.** Photograph of a "spongy iron" filter as published by Nichols [48]. The raw water (**B**) is supplied from a bottle, which is inverted into the upper part of the inner vessel. Spongy iron ($Fe^0$) is contained in an inner vessel. Below the $Fe^0$ material are: (i) A layer of sand ('prepared sand'), (ii) a regulator (**A**), and (iii) a receptacle (**C**) with a tap for filtered water. Prepared sand consists of three layers of aggregates: Pyrolusite ($MnO_2$), sand, and gravel, which separate traces of iron from the filtrate.

## 2. The Operating Mode of $Fe^0/H_2O$ Systems

### 2.1. General Aspects

The large majority of investigations pertaining to use of $Fe^0$ for water treatment are conducted in the batch mode. In such studies, various types of $Fe^0$ materials were used, including: Iron foam, iron nails, scrap iron, steel wool, and various composites [85–90]. While batch laboratory studies provide useful information in the treatability studies for column experiments, continuous column studies provide more practical information for the design of $Fe^0$ filters. In batch experiments (static mode), the same solution remains in contact with a given amount of $Fe^0$ for a given experimental duration [91,92]. The removal process ideally continues, because $Fe^0$ as a filter material keeps evolving until a dynamic or transient pseudo-equilibrium is reached between the dissolved and removed contaminants [91–93]. This transient pseudo-equilibrium is related to the apparent corrosion kinetics (intrinsic reactivity) of the filter material (e.g., $Fe^0$). Unlike pure adsorbents [94], in $Fe^0/H_2O$ systems, the established equilibrium is not static in nature, as it changes further with time ('rust never rests') [94,95]. Contrary to the transient pseudo-equilibrium in batch systems, column experiments are dynamic, and solution continuously enters and leaves the column. Thus, a similar pseudo-equilibrium is not expected and is never established at any stage between dissolved contaminants and the amount removed. Similar to the pure adsorbent systems, equilibrium has to be continuously established, as each time a fresh influent solution (constant concentration) is added [92]. Hence, equilibrium in column mode is a real dynamic equilibrium. However, it should be considered that contaminant scavengers are generated in-situ by a dynamic process, whose non-linear kinetics has not been properly considered in the previous literature [78,84]. In-situ generated contaminant scavengers are also a major cause of porosity decrease and permeability loss [96]. The next section critically assesses how available information on the efficiency of $Fe^0/H_2O$ systems for decentralized safe water has been gathered in order to develop better designs of the next generation $Fe^0$-based systems.

### 2.2. Fundamental Principles

Treatability studies for Anderson-like processes are relatively simple to design. Whether the intrinsic reactivity is characterized or not, it suffices to identify the $Fe^0$ mass (e.g., kg) necessary to treat a given fixed volume (e.g., $m^3$) of contaminated water with certain characteristics (e.g., nature and extent of contamination, and solution chemistry) for a given operational duration (e.g., hours). The mixing type and intensity can be deliberately selected as it can be reproduced in field applications. Apart from the ways to separate and recycle used filter materials, the general experimental procedure for testing and utilizing powder activated carbons can be used without further modifications [97,98].

Treatability studies for Bischof-like processes are essentially more challenging to design. First, pure $Fe^0$ systems are not suitable due to the loss of porosity in the long-term [83,98,99]. Second, the size of $Fe^0$ particles and that of admixing aggregates (e.g., gravel, $MnO_2$, and sand) should be carefully considered as they determine the initial diameter of the pores and thus the long-term porosity and permeability of the system [83]. Third, no attempt should be made to accelerate iron corrosion using tools that cannot be reproduced under field conditions. In particular, the use of the rapid small-scale column test (RSSCT) should be avoided. RSSCT tests are scaled-down versions of pilot or full-scale systems for pure adsorbents [94,95,97]. Given that $Fe^0$ is a reactive material, the conventional RSSCT test cannot address the non-linear kinetics of its corrosion. Fourth, the experiments should last for long times to enable the formation of iron corrosion products. Considering the 18 months of Antwerp [50–52] as a reference, the goal can be to design systems running for at least six to twelve months, such that recommendations can be made to change the filters once or twice per year. Moreover, given that each individual $Fe^0$ material is unique in its intrinsic reactivity, a database of available materials subjected to detailed characterization is needed to develop site-specific designs for different types of water (e.g., carbonate rich and humic substance rich).

### 2.3. Design Aspects

$Fe^0/H_2O$ systems have been reported to effectively remove several classes of toxic substances from water [27–39,98,99]. Relevant contaminants include widespread ones such as toxic metals and metalloids (e.g., arsenic, zinc, cadmium, lead), organics (e.g., halogenated aromatics, nitrosoamines), nutrients (e.g., nitrates, phosphates), and radionuclides (e.g., uranium). However, removing these toxics in the presence of ubiquitous natural constituents (e.g., chlorides and humic substances) is challenging due to their interfering effects [100]. In many cases, the concentrations of ubiquitous natural constituents are up to nine orders of magnitude higher than that of the target contaminants [2]. On the other hand, efforts to buffer the experimental systems or to fix their ionic strength correspond to modifying the process of iron corrosion in a manner that will not be reproduced in nature [101–103]. In particular, ferrous and ferric chlorides are more soluble than their hydroxide counterparts [103]. Therefore, whenever chloride salts (e.g., NaCl and $CaCl_2$) are used to fix the ionic strength of experimental solutions, the whole corrosion process is modified. Using nitrate or sulphate salts induces similar modifications to different extents. However, unlike chlorides, which are conservative, sulphates and nitrates may induce microbial activities in the $Fe^0$/sand filter systems, including biofilm formation and hydrogen sulphide generation, can thus be consumed in the process.

The foregoing modifications have not been properly considered by past investigators. Table S1 presents the large variability of operational conditions used in a few selected independent investigations [11,15,104–108]. For example, it is seen that the $Fe^0$ aggregate ratio ranges from 1:1 to 1:3, while the type and form of materials used also vary. The variation of the experimental durations and contact times is also considerable (i.e., from a few seconds to months). Consequently, research findings on $Fe^0/H_2O$ systems have been mixed, resulting in some studies drawing the generalized and often misleading conclusion that $Fe^0/H_2O$ systems are not reliable for water treatment [109–114]. Fortunately, this negative impression co-exists with success stories, which are equally not based on the fundamental understanding of the chemistry of the system [5,61,62,115,116]. For example, the Indian Institute of Technology Bombay (IITB) has recently presented an efficient $Fe^0$-based filtration system for arsenic removal (Section 3.2). Several demand-designed IITB filters using iron nails as $Fe^0$ material are currently supplying small communities of West Bengal with arsenic-free drinking water. However, no single $Fe^0$ system is specific to arsenic [101,102], and the intrinsic reactivity of used iron nails has not been characterized [110,111]. The example of IITB filters demonstrates the urgent need of rational design concepts for more robust and efficient $Fe^0$-based systems [112].

## 3. Performance of Available $Fe^0/H_2O$ Systems

Several $Fe^0$-based clean water technologies are available for rural households in the developing world, and particularly Asia. Most of them have been designed in the framework of arsenic removal [116–128]. Water can be made available to a whole village, or alternatively, individual families may use household filters. Community filters are preferred as they are easier to monitor than household ones [5,120,121]. The literature on $Fe^0$-based systems for decentralized safe drinking water provision has not yet been critically reviewed. Available information is scattered in overview articles comparing $Fe^0$-based and other technologies [112,113]. It seems certain that $Fe^0$-based systems are efficient for decentralized safe drinking water [5,55,60–62,68,106,114–116]. This section presents two success stories and suggests ways for improvement.

### 3.1. SONO Arsenic Filter

#### 3.1.1. Overview

Figure 2 shows the SONO filter developed in a collaborative work between the George Mason University (Oregon, USA) and a non-governmental organization (NGO) from Kustia (Bangladesh) [55,68,118]. According to Etmannski [61], over 90,000 of these filters have been distributed in Bangladesh. They are often given to families for free through NGOs. Water treatment is achieved by running raw water through a porous composite iron matrix (CIM), which is made of cast iron with a small amount of manganese oxide ($MnO_x$). $MnO_x$ in the CIM is reported to catalyze oxidation of As(III) to As(V), which is then removed by complexation on the surface of iron corrosion products (e.g., FeOOH).

The filter consists of two plastic buckets placed one on top of the other in a rack. The buckets contain brick chips, layers of fine and coarse sand, wood charcoal, and CIM (Figure 2). The buckets are connected with plastic tubing and have valve controls near the bottom [55,61,62]. The process does not require any maintenance such as backwashing or regeneration of media [61,62]. Etmannski [61] reported that the life span of the filter is less than two years. The exhausted filter material is replaced and the used material is disposed of in an engineered landfill. The sole problem of SONO filter is that expiry dates are often ignored, partly because the filters are not really affordable for the population. Moreover, it is often difficult for communities with limited expertise to determine the end of the lifespan of the filter. To address this limitation, developing a simple $Fe^0$-based clock system to avoid the use of exhausted filters is part of the concept presented in the current communication [117].

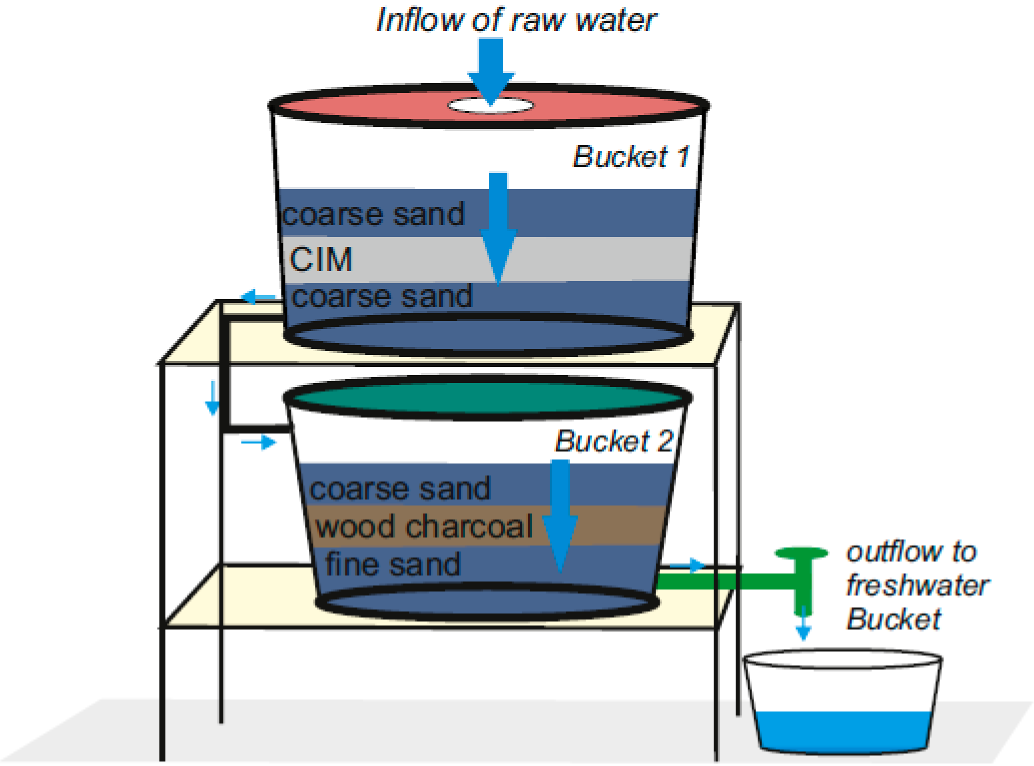

**Figure 2.** A conceptual sketch of the SONO filter initially developed for arsenic removal (adapted from Hussam and Munir [55]).

### 3.1.2. Operating Principles and Limitations

The SONO filter corresponds to the ‘spongy iron’ filter of Prof. Bischof [48–50]. It has been fully and independently developed, as is evident in the peer-reviewed literature from 2000 [118] and 2007 [55]. The SONO filter has two major characteristics: (i) Using a porous composite provides room (or space) for expansive corrosion, and thus extended service life (i.e., long-term permeability) [83,84,97], and (ii) using $MnO_x$ sustains long-term corrosion (i.e., less reactivity loss) [99,102]. Details of operating conditions and performance of the SONO filter are given in literature [61,67,68]. According to Munir et al. [99], at an optimum flow rate, 8.4 L/h, a SONO filter a can reduce arsenic concentration in groundwater from an initial value of 1600 ppb to about 20 ppb. This corresponds to an arsenic removal efficiency of 98.8%. It is still considered that the efficiency of SONO filters for As removal relies on some oxidation of $As^{III}$ to $As^{V}$. However, this assumption has not been scientifically demonstrated. Moreover, SONO filters have been found to remove a myriad of other contaminants, of which some are not redox active [112–114]. For example, Tuladhar and Smith [115] reported the removal of up to 24 different species including metals, nitrates, and pesticides. It is rather surprising that, despite its potential to contribute to the provision of clean water, the SONO design has not achieved widespread use, even in developing countries.

## 3.2. IITB Arsenic Filter

### 3.2.1. Overview

The Indian Institute of Technology Bombay (IITB) filter consists of two tanks that are similar in design (Figure 3). Well-water is pumped into the top of the first tank, and sprinkled into the top trays using a perforated pipe [5,61,62]. The aerated water comes into contact with iron nails ($Fe^0$), which are held in trays (Figure 3). $Fe^0$ is used to generate $Fe^{II}$ species, which are oxidized to $Fe^{III}$ by dissolved oxygen ($O_2$), thus generating iron corrosion products. Iron corrosion products (FeCPs) act as adsorbents for arsenic removal [118–122]. Sand is not used in the IITB filters as it would trap all the FeCPs in the top layers, and cause clogging, which would then require frequent cleaning. The filter bed is made up of stone chips and gravel, which reduce the head-loss and allow the FeCPs to penetrate deeper into the filter, thus achieving a higher contact time [5,61,62]. A sequence of two or three identical tanks forms a complete arsenic removal system, with a tap at the bottom of the last tank, which is used directly by the consumer [5,61]. The number of tanks depends on the initial concentration of arsenic in the local groundwater. The generated waste, supposedly FeCPs and $As^V$, is certainly stable under oxic conditions [123], meaning that substantial As leaching from the waste is not likely to occur. The operators of these systems are trained volunteers who are responsible for maintenance. Water is provided for free to the whole community.

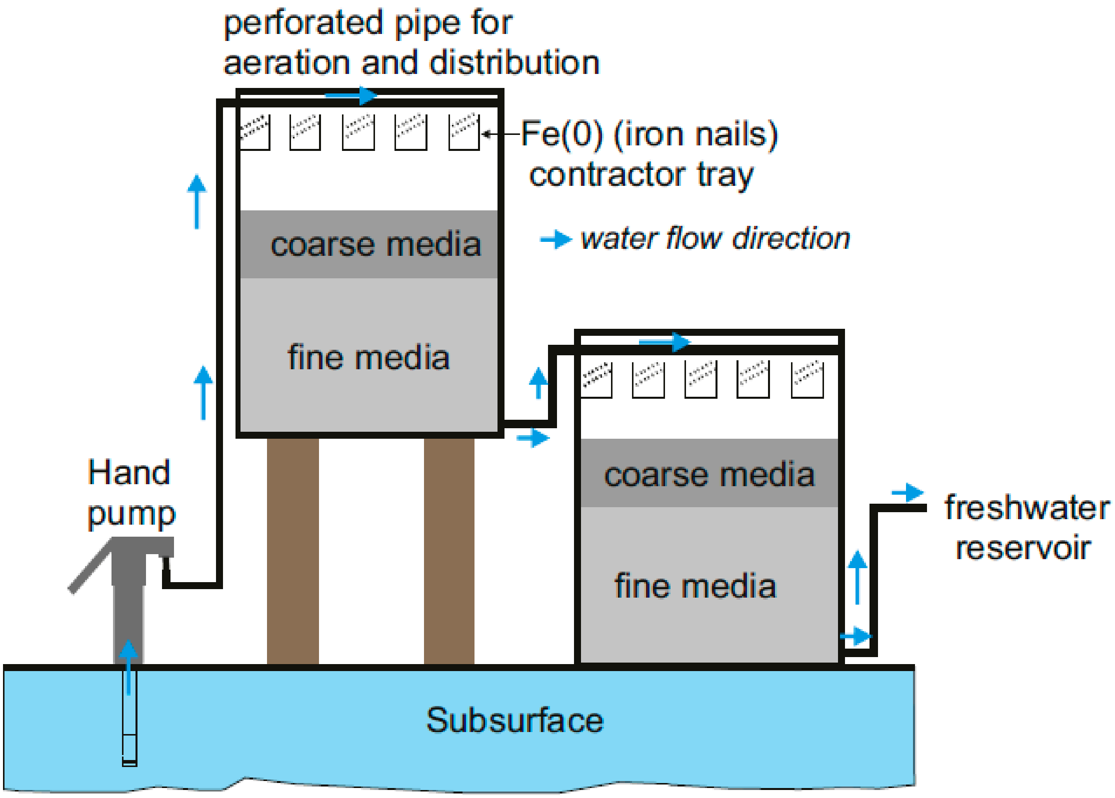

**Figure 3.** Conceptual sketch of the Indian Institute of Technology Bombay (IITB) filter (adapted from Etmannski [61]).

### 3.2.2. Operating Principles and Limitations

The IITB filter was conceptualized using the knowledge that multiple additions of Fe(II) to slow sand filters improve their removal capacity for arsenic [16,118–121,124–128]. As is adsorbed onto FeCPs, generally termed as hydrous ferric oxide (HFO) [125,129]. The system was designed and built by Tuhin Banerji, a PhD student under the supervision of Professor Sanjeev Chaudhari from IIT Bombay [5]. Detailed operating conditions of the IITB filter and its capacity to remove arsenic in groundwater are given in the literature [5,60–62].

The IITB filter corresponds to the Anderson Process wherein contaminants are adsorbed by FeCPs in a contactor, and then removed from the aqueous phase in the subsequent filter by size-exclusion. The contaminant-laden flocs are filtered out of the water. The major weakness of the IITB filter is that the used iron nails have not been characterized for their intrinsic reactivity. Hu et al. [111] have attested that commercial iron nails are a good long-term source of $Fe^{II}/Fe^{III}$, but considerable differences were reported in the kinetics of their dissolution in 0.02 M EDTA (ethylenediaminetetraacetic acid) for three months of column leaching experiments [110,111]. Accordingly, by considering the intrinsic $Fe^0$ reactivity, the design of IITB filters can be significantly improved. For example, a lower mass of a more reactive $Fe^0$ material can be used to design a more efficient system, or the more reactive material may just need two beds where three beds of the less reactive material are needed. Moreover, if a reactivity database of iron nails is made available, $Fe^0$ material selection criteria for specific water types (e.g., carbonate-rich) can be developed. Finally, the non-linear kinetics of long-term iron corrosion should be taken into consideration in the design process [78–80,83,84]. However, Banerji and Chaudhari [5] have also propagated the mistake in thinking that $As^{III}$ is oxidized to $As^V$ prior to removal. This simplification is not scientifically justified as the removal efficiency of $Fe^0$-based systems for both $As^{III}$ and $As^V$ species have been demonstrated [125–128], and is rooted on the availability of nascent iron hydroxides within the $Fe^0/H_2O$ system [129]. Therefore, rooting the design on the scientific understanding of the $Fe^0/H_2O$ system could lead to the development of the next generation IITB filters.

The history of $Fe^0/H_20$ systems, highlighting the key chronological events is presented in our earlier paper [80], and thus is not repeated here. In particular, current IITB filters were developed between 2008 and 2014 (6 years) [5,60–62]. This means that the remaining 12 years to 2030 are enough to harness existing scientific knowledge to optimize such systems for a universal safe drinking water supply. For contaminants that may not be efficiently removed in $Fe^0/H_2O$ systems [6,130,131], multi-barrier filter systems based on other low-cost materials, including biochars, can be developed [132–142].

## 4. $Fe^0$/biochar Dual Systems: Towards the Next Generation Water Filters?

### *4.1. $Fe^0$/Biochar Dual Systems: The Scientific Arguments*

Biochars are renewable carbon-rich low-cost biomaterials formed via the pyrolysis of diverse biomass materials, which are often cheap and readily available. Biochars possess several unique physico-chemical characteristics (e.g., high surface area, surface charges, high fixed carbon, and high stability) that make them excellent adsorbents for water filters [133]. An increasing body of literature, including experimental studies and reviews exists, demonstrating the capacity of biochars to effectively remove potentially toxic physico-chemical and microbial contaminants from aqueous systems [132,133]. The capacity of biochars and their activated derivatives to remove organic, inorganic, and microbiological contaminants in aqueous systems and removal mechanisms have been discussed in several reviews [137–142]. For example, a detailed discussion of the potential applications of biochars as adsorbents in drinking water filters for use in low-income countries is presented in Gwenzi et al. [132]. Similarly, as highlighted in Section 2, numerous studies, including recent reviews, have shown that various forms of $Fe^0$ are also low-cost and readily available materials for potential use in water filters [4,58,60–62,134–136].

$Fe^0$ technology was initially developed to remove color, pathogens, odors, and turbidity in aqueous systems [8,50–52]. Its application has extended to modern drinking water treatment, as evidenced by increasing suggestions for such applications, especially in low-income countries [5,61,62,134]. The capacity of biochars and $Fe^0/H_2O$ systems to remove potential toxic contaminants highlights their potential application for drinking water treatment, particularly in low-income countries. Rather than considering biochar and $Fe^0$ as competing technologies, the concept presented here seeks to harness the unique properties of the two technologies. Surprisingly, widespread use of $Fe^0/H_2O$ and biochar-based water filters is still lacking even in low-income countries, which are expected to benefit immensely from such low-cost technologies. This trend could be attributed to two reasons: (i) Lack of detailed design data and procedures for both $Fe^0/H_2O$ and biochar water filters, and (ii) in the case of $Fe^0/H_2O$ systems, inconsistencies in scientific data reported in literature [4,8]. Therefore, addressing these issues is critical in advancing the scientific knowledge, and enhancing the uptake and adoption of both technologies. The outlook is to develop the next generation of water filters, where $Fe^0$ and biochar complement each other to achieve synergies and better performance.

Current research on biochar and granular $Fe^0$ has largely developed independently of each other. Understandably, there have been limited attempts to simultaneously harness the unique contaminant removal attributes of the two materials to develop novel water filter systems. The exceptions are a few studies using biochar as a dispersant material for nanoscale $Fe^0$ ($nFe^0$/biochar) [137,138]. While subject to confirmation through experimental studies, the current article posits that dual material water filters consisting of both granular metallic iron ($gFe^0$) and biochar ($gFe^0$/biochar) will attain a higher contaminant removal than those with either $gFe^0$ or biochar alone. This proposition is premised on the fact that $gFe^0$/biochar systems will act as hybrid systems, where contaminant removal occurs on both the $gFe^0$ and biochar. In fact, the concept of a water filter combining $gFe^0$ and biochar is almost analogous to the SONO filter with CIM and wood charcoal (Figure 2).

Conventional granular $Fe^0$ filters are rendered sustainable by admixing $Fe^0$ with inert (e.g., gravel and sand) or reactive material (e.g., $MnO_2$) [4,86,87,102,143,144]. Considering the high stability and low bulk density of biochars [139–141,145–148], it is postulated that biochars can be used to increase porosity instead of gravel or sand. These anticipated biochar effects are similar to the increased soil porosity and hydraulic conductivity often reported on biochar-amended soils [139–142,145]. In summary, the novelty of the $gFe^0$/biochar water filters is premised on the understanding that such dual systems simultaneously harness the contaminant removal capacities of biochar and $gFe^0$ systems.

While the capacity of $gFe^0$ and biochar to remove contaminants in aqueous systems is beyond doubt, some controversies and/or knowledge gaps still exist for both technologies. For example, scientific literature on $Fe^0$ technology is fraught with inconsistencies on the mechanisms responsible for contaminant removal [70–77]. Some studies predominantly attribute contaminant removal by $Fe^0/H_2O$ systems to direct reduction by $Fe^0$, while overlooking the role of indirect reduction [23,70–74]. In fact, the confusion introduced by the electrochemical nature of aqueous iron corrosion, and the chemical nature of contaminant reduction (no electrons from $Fe^0$) appears to be a common source of confusion in environmental research. In a recent publication, Blackwood [149] highlighted that the electrochemical nature of microbial influenced corrosion (MIC) and was widely reported in scientific publications over the years has never been established. Several recent papers have discussed the controversy pertaining to the $Fe^0/H_2O$ systems, the broader effects associated with its perpetuation, and the need for consistency [150,151]. For example, inconsistencies pertaining to $Fe^0$ technology and their origins and future research directions to overcome them were recently highlighted [4,79,80]. Similarly, several knowledge gaps still exist on the application of biochars as water filter materials, and these are discussed in earlier review articles [133,148]. These inconsistencies and imperfect knowledge on both $Fe^0$ and biochar culminate in uncertainty. Therefore, combining $Fe^0$ and biochar may contribute to minimizing the limitations associated with each of these two technologies, and partly address this uncertainty. In summary, the novelty of developing filters based on both $Fe^0$ and biochars includes: (i) To develop 'hybrid' systems harnessing the contaminant removal capacities of the two materials, and (ii) biochar that is highly porous and stable [139–141] and can thus prevent loss of porosity and hydraulic conductivity, and enhance the sustainability of $Fe^0/H_2O$ systems.

### *4.2. Conceptual Designs of gFe$^0$/Biochar Dual Water Filters*

Literature on the design principles, and performance evaluation of water filters with dual active materials for small communities, is still limited [5,60,125]. Lacking that, a number of conceptual designs for such systems, and their potential applications based on the objectives of the water treatment system are highlighted here.

Three typical conceptual designs of $gFe^0$/biochar systems are as follows:

(1) $Fe^0$/biochar layer can be included as an additional mixed layer in dual or multi-layered fixed-bed systems in traditional bio-sand filters to increase contaminant removal efficiency. Since most communities in low-income countries are already familiar with conventional bio-sand filters, such an improved design may experience more rapid uptake and adoption than completely new stand-alone $gFe^0$/biochar water filters discussed below.

(2) Development of new stand-alone $gFe^0$/biochar water filters without the bio-sand component. Such $gFe^0$/biochar water filters will rely on dual effects of both $Fe^0$ and biochar to effectively remove contaminants. The design configuration of such $gFe^0$/biochar filters may consist of either a single fixed-bed column reactor or a series of such reactors, wherein $gFe^0$ and biochar will be either mixed or layered.

(3) A water filter system consisting of a sequential configuration of two or more $gFe^0$/biochar fixed bed columns, where the contaminated water first passes through biochar or biochar/sand fixed-bed adsorption column(s), and is then followed by $Fe^0$-fixed-fixed bed columns. In such as design, the $Fe^0$ filter serves to polish the water initially treated in a biochar-based water filter, thereby improving the contaminant removal efficiency of the system. In fact, the conceptual design of a water filter based on this principle is consistent with the pioneering vision envisaged during the early stages of development of $Fe^0$ remediation systems [4,134–136].

## 5. Future Research Directions

### *5.1. Key Knowledge Gaps*

To this end, the presentation has highlighted several concepts and propositions, but there is a paucity of data pertaining to several thematic areas on $gFe^0$/biochar water filtration systems, including: (i) Detailed material characterization and optimization of $Fe^0/H_2O$ systems, (ii) design and performance evaluation of the $gFe^0$/biochar filters, (iii) life cycle analysis and disposal of spent/loaded filter materials, and (iv) development and evaluation of a $Fe^0$ filter clock prototype. Therefore, the specific knowledge gaps that need to be addressed in future research on each of these thematic areas are highlighted here.

#### 5.1.1. Material Characterization and Optimization of $Fe^0/H_20$ Systems

The intrinsic reactivity of most $Fe^0$ materials reported in literature is often not characterized [110,111]. Therefore, future studies should characterize the intrinsic reactivity and non-linear corrosion kinetics of $Fe^0$ materials, with the goal of establishing a database of readily available materials (based on an operational reference). Moreover, the volumetric expansion associated with iron rusting, ion-selectivity of $Fe^0$ systems, and the role of co-solutes/agents, warrant consideration in the design and optimization of $Fe^0$ water filters.

#### 5.1.2. Design and Evaluation of $gFe^0$/Biochar Water Filters

The potential to develop low-cost water filters incorporating two (dual) reactive materials, specifically $gFe^0$ and biochars has not been fully exploited in previous research. Therefore, there is a need to design, fabricate, and evaluate such systems under laboratory and field setups. Such performance evaluation should use both batch and column experiments, and process modelling to investigate contaminant removal, hydraulic behavior (porosity, hydraulic conductivity, clogging), and potential to regenerate filter materials. Such studies must include: (i) Detailed characterization of materials used, and (ii) ensuring that experiments are conducted in a quiescent mode rather than agitation in order to acquire data that is practically relevant and useful for the determination of the system design parameters.

#### 5.1.3. Life Cycle Analysis and Disposal of Spent/Loaded Filter Materials

The widespread uptake and adoption of $gFe^0$/biochar water filters in households is likely to generate substantial quantities of spent filter materials loaded with potentially toxic contaminants. From a sustainability viewpoint, it is important to understand the behavior and fate of the contaminants and spent filter materials from a life cycle perspective along the water treatment chain. In this regard, it is important to ensure that contaminants are not released back into drinking water sources. Soil application of spent filter materials is one possible disposal method, but the leaching behavior and fate of contaminants, and their potential eco-toxicological effects on crop species, and soil organisms and biogeochemical processes will need to be investigated. Such studies may also investigate the risk of contaminant transfer into the human food chain via food crops grown on amended soils.

#### 5.1.4. Design and Evaluation of a $Fe^0$-based Filter Clock System

The idea to develop a $Fe^0$-based clock system for water filters was recently presented [117]. Future research should focus on translating this concept into a prototype. Such research may entail determining optimum $gFe^0$/biochar/inert material mixtures that provide a filter with various clogging times corresponding to the target design life of a filter. In this regard, Faraday's law, current intensity values, and any existing long-term $Fe^0$ corrosion data (e.g., from utility pipes and archaeological iron features and artefacts) may provide a starting point in developing such a filter clock. Such a prototype will need to be subjected to rigorous laboratory and field-testing under relevant field conditions.

### *5.2. $Fe^0/H_2O$ Filtration Systems: A Framework for the Next Decade up to 2030*

The fundamental flaws associated with the pragmatic approach currently used for design of $Fe^0/H_2O$ filtration systems, coupled with the knowledge gaps highlighted, and the urgent need to achieve universal clean drinking water provision in the remaining decade (i.e., by 2030), calls for the rapid translation of existing scientific knowledge into frugal and efficient $Fe^0/H_2O$ filters. Figure 4 presents a proposed framework and timeline for advancing research on $Fe^0/H_2O$ water filtration systems in the next decade up to 2013.

The proposed framework consists of six key steps summarized as follows (Figure 4):

(1) Detailed characterization of $Fe^0$ filter materials, and subsequent establishment of a database on $Fe^0$ reactivity;
(2) Development of detailed design protocols for $Fe^0$ filters based on fundamental scientific understanding of contaminant removal mechanisms and $Fe^0$ reactivity, particularly the volumetric expansive nature of iron corrosion;
(3) Development of a $Fe^0$ filter clock to determine the end of design or service life of $Fe^0$ water filters;
(4) Development and evaluation of robust and efficient prototypes of $Fe^0$ filters, including multi-barrier systems;
(5) Adaptive field-testing and optimization of $Fe^0$ filters, including participatory evaluation by target end-users and beneficiaries particularly in developing countries, where such low-cost technologies are most needed; and
(6) Out-research and dissemination of efficient $Fe^0$ filters, including capacity-building in their design, operation and maintenance, and follow-up studies on adoption.

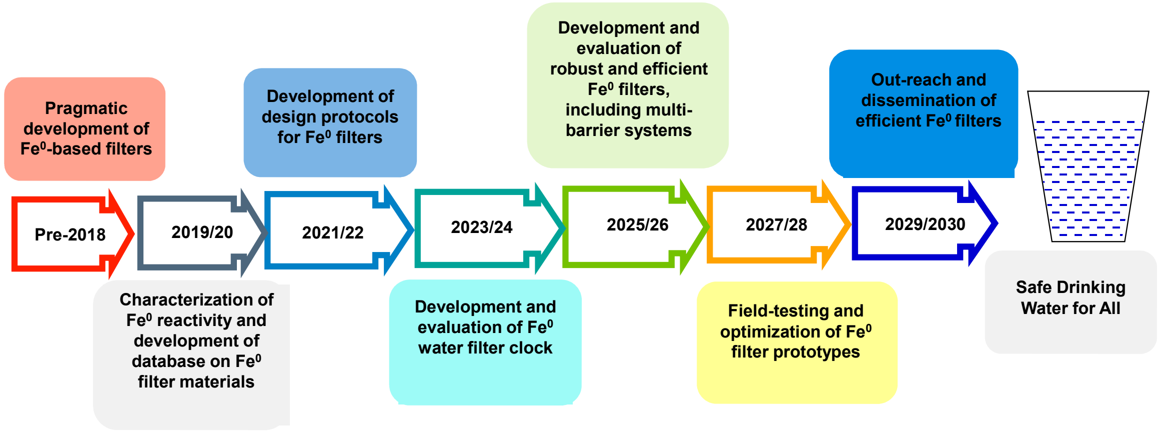

**Figure 4.** A proposed framework for advancing research on $Fe^0/H_2O$ filters in the next decade up to 2030.

## 6. Conclusions and Outlook

There is a broad consensus on a lack of capacity in the developing world to achieve the UN SDGs, among them, SDG 6 on ensuring the availability and sustainable management of water and sanitation for all. Based on this consensus, it then follows that appropriate water treatment technologies need to be introduced in these regions, a position which most scientists and engineers from the developing world seem comfortable with. Subsequent to Dr. Robert Koch's seminal work establishing the relationship between drinking water quality and human health in Hambourg and Altona (Germany) as early as 1892/1893, slow sand filters (SSFs) were developed to avoid cholera and other water borne diseases. During the past century, several attempts have been made to modify SSFs to optimize the removal of pathogens and chemical contaminants. Several readily available adsorbents, including biochars, were successfully tested, but are rarely used in practice. Metallic iron ($Fe^0$) has been demonstrated to be very efficient in removing pathogens and several classes of chemical contaminants. Moreover, efficient and affordable $Fe^0$-based systems such as the IITB filters were developed locally and partly implemented and maintained by their low-skilled owners.

The case of IITB filters should motivate scientists and engineers in low-income countries to challenge the notion that water treatment solutions should be introduced from somewhere. Moreover, achieving the SDGs is not a matter of big money (development aid), but rather a matter of political willingness to support and motivate local scientists, engineers, and communities [152,153]. The work highlighted herein is sowing the seeds for the establishment of $Fe^0$-based treatment systems as a universal solution for decentralized safe drinking water supply. The presented science-based approach will assist the design of more robust and efficient systems that are appropriate for several regions of the world. The resulting systems should be implemented with proper accompanying educational and capacity building strategies. It is the firm conviction of the authors that the necessary skills needed to achieve Goal 6 of the UN SDGs are already available everywhere, including in Africa. All that is needed is the courage and political will to start this collective work now (2019) in order to achieve the UN SDGs by 2030.

**Supplementary Materials:** The following are available online at http://www.mdpi.com/2073-4441/11/3/429/s1, Table S1: Summary of some experimental conditions used for experiments in selected peer-reviewed articles using $Fe^0$ materials for decentralized water treatment.

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