Communication

Avoiding the Use of Exhausted Drinking Water Filters: A Filter-Clock Based on Rusting Iron

Arnaud Igor Ndé-Tchoupé [1], Mesia Lufingo [2], Rui Hu [3], Willis Gwenzi [4], Seteno Karabo Obed Ntwampe [5] , Chicgoua Noubactep [6,*] and Karoli N. Njau [2]

[1] Department of Chemistry, Faculty of Sciences, University of Douala, Douala BP 24157, Cameroon; ndetchoupe@gmail.com

[2] Department of Water and Environmental Science and Engineering, Nelson Mandela African Institution of Science and Technology, Arusha 447, Tanzania; lufingom@nm-aist.ac.tz (M.L.); karoli.njau@nm-aist.ac.tz (K.N.N.)

[3] School of Earth Science and Engineering, Hohai University, Fo Cheng Xi Road 8, Nanjing 211100, China; rhu@hhu.edu.cn

[4] Biosystems and Environmental Engineering Research Group, Department of Soil Science and Agricultural Engineering, Faculty of Agriculture, University of Zimbabwe, P.O. Box MP167, Mount Pleasant, Harare, Zimbabwe; wgwenzi@yahoo.co.uk

[5] Bioresource Engineering Research Group (BioERG), Department of Biotechnology, Cape Peninsula University of Technology, P.O. Box 652, Cape Town 8000, South Africa; NtwampeS@cput.ac.za

[6] Department of Applied Geology, University of Göttingen, Goldschmidtstraße 3, D-37077 Göttingen, Germany

* Correspondence: cnoubac@gwdg.de; Tel.: +49-551-393-3191

Abstract: Efficient but affordable water treatment technologies are currently sought to solve the prevalent shortage of safe drinking water. Adsorption-based technologies are in the front-line of these efforts. Upon proper design, universally applied materials (e.g., activated carbons, bone chars, metal oxides) are able to quantitatively remove inorganic and organic pollutants as well as pathogens from water. Each water filter has a defined removal capacity and must be replaced when this capacity is exhausted. Operational experience has shown that it may be difficult to convince some low-skilled users to buy new filters after a predicted service life. This communication describes the quest to develop a filter-clock to encourage all users to change their filters after the designed service life. A brief discussion on such a filter-clock based on rusting of metallic iron (Fe^0) is presented. Integrating such filter-clocks in the design of water filters is regarded as essential for safeguarding public health.

Keywords: adsorptive filtration; frugal innovation; permeability loss; water treatment; waterborne diseases; zero-valent iron

1. Introduction

Safeguarding public health within a given community has two key aspects: preventing the occurrence of diseases and controlling the spread of such diseases, with disease curing being the function of medicine. However, these three aspects are inter-connected. If the cause of a specific disease is understood or at least well-characterized, then the spread of that disease within a community can be reduced to a minimum or completely eradicated. Supplying human communities with safe drinking water has been established as a necessity, thus warrants monitoring and scrutiny [1–4], to mitigate against waterborne diseases.

The development of affordable, appropriate, efficient and sustainable water filtration systems is regarded as one of the fundamental research goals in efforts to achieve the United Nations' Sustainable

Development Goals (UN SDGs) [5]. The role of filtration systems based on metallic iron (Fe^0 filters) has been largely discussed in this context [2,5–11]. Fe^0 filters are regarded as a key technology because suitable Fe^0 materials are universally available and design criteria have been established [12–14]. One shortcoming of Fe^0 filters is the time-dependent permeability loss of the porous bed. This limitation has the potential to become advantageous when designing next generation filters for low-skilled and low-income communities. Such next generation Fe^0-based filters will rely on rusting to develop a clock system to signal filter exhaustion.

Household water treatment systems are extremely difficult to maintain. Their maintenance is mostly left to the owners because necessary specialised supervision by experts would be difficult to secure [10,15]. Impoverished communities are usually less educated, with few members of such a society being equipped with few saleable skills. Therefore, they may lack money for even vital water filters and would attempt to continue to use an exhausted filter. Moreover, such communities often lack access to an analytical laboratory to regularly analyse the quality of drinking water from such filters [16]. Therefore, to safeguard their health, low-cost and appropriate means should be found to encourage water filter changes after the estimated service life of the filter. One option includes exploiting the volumetric expansive nature of iron corrosion or rusting in aqueous systems [17,18]. In this regard, a small Fe^0 filter designed to clog promptly at the end of the service life can be added to any water filtration system (not only Fe^0-based ones). When the system is clogged, no water is released, thereby ensuring the user to change the filtration device.

This communication is structured as follows; first, an overview on existing household filtration systems is presented, and then the scientific knowledge behind the filter-clock and some practicable designs of such a monitoring system are discussed.

2. Commercial Household Water Filters: Now and Then

Commercial water filtration systems for households are antecedent to centralized water provision systems [19]. Historically, communities initially used water from wells as an alternative to surface water or collected and stored rainwater. They then realized that stored water might need some sort of treatment. With the development of science, it was later established that virtually each water source needs some degree of treatment before use as a potable water source [4]. As early as the 1860s, commercial household water filters were commonplace in Europe [19–21]. Nichols [19] documented the following aggregates for use as filter materials: animal charcoal, broken bricks, broken stone, carbide of iron (Fe^0), cotton, flannel, gravel, iron turnings (Fe^0), iron wire (Fe^0), pebble stone, porous stone, powdered glass, sand, sawdust, spongy iron (Fe^0), wood-charcoal, and wool among others.

Nichols [19] explicitly noticed that there was nothing better at removing dissolved organic materials than 'well-burned animal charcoal (bone-coal)'. In the 1930s and in the 1980s 'bone-coal' then termed as bone char was rediscovered and mostly used for fluoride removal ([22]). The application of metallic iron (Fe^0) for safe drinking water provision has a similar history: (i) established before 1900 [19]; (ii) independently rediscovered during the 1950s [23] and the 1990s [10,24–27].

It might be surprising to learn that technical expertise in filtration for decentralized safe drinking water provision is a century old. Recent review articles [1,2,6–9] revealed that modern filtration devices have the following two common characteristics: (i) are made of similar materials [2,6,7] and (ii) experience the same limitations: i.e., their filtration capacity is not indefinite; hence, they require renewal at proper intervals [10]. A special material, termed activated carbon, was introduced in the 1940s and was intensively investigated for some 30 years [9,28,29]. Granular activated carbon (GAC) is the form commonly used to design household water filters. Just like Fe^0-based filters, each designed GAC filter has a limited service life. Therefore, appropriate filter replacement or changes in filtration units is required after the exhaustion of the designed service life.

Despite being the best available adsorbent for water treatment due to its excellent adsorption capacity, GAC could not be used at a large scale due to its high cost of production (and regeneration). Moreover, such costly adsorbents are unaffordable to low-income households in developing countries.

This prohibitive cost has prompted scientists to develop more-affordable adsorbents from agricultural, industrial and geo-materials [2,6,7,9,30,31]. Factually, resulting filters vary in their service lifes, which are intuitively shorter than those of GAC filters. For the developers of filtration units, their production is based on the availability, affordability and efficiency of the resulting filters at household level, with their regenerability being of secondary importance, although a system to collect used filtration materials could be introduced. It is not rare that most water filters on the market contain an operating manual describing the filter, and specifying its expected service life. The given service life of a filter is considered a reliable way to determine the longevity of filtration systems. However, the question is how should even an illiterate user be knowledgeable about filter life span, and how to change a filter after the expected service life? This aspect is discussed herein focusing on Fe^0 filtration systems.

3. Fe^0-Based Filter Clocks

3.1. General Aspects

Water treatment technology using Fe^0 materials consists of 'putting corrosion to use' [32]. In this case, iron corrosion products (FeCPs) are mainly used as contaminant collectors and degraders [10,26,27,33–39] or as catalysts for chemical reactions ([40]). Herein, a different aspect of using FeCPs is presented: The volumetric expansive nature associated with FeCPs generation [17] can be used to control filter life span. In fact, each FeCP (oxide) is larger in volume than the parent Fe^0 (iron) in the metal lattice ($V_{oxide} > V_{iron}$). In practice, an expansion coefficient (η) has been introduced such that $V_{oxide} = \eta * V_{iron}$ ($\eta \geq 2.01$) [18]. The η value depends on the availability of dissolved O_2. This means that, each Fe^0-based filter is prone to clogging 'by virtue' of the volumetric expansive nature of aqueous iron corrosion [18]. In other words, by using different Fe^0 materials and/or different Fe^0 proportions in hybrid systems (e.g., Fe^0/sand, Fe^0/pumice, Fe^0/pozzolane) [39], it is possible to determine the service life (t_∞) of a filter. That is, for example, the time frame for which a household-based filter delivers enough safe drinking water for a typical number of people in a household.

Although Fe^0-based filters are mostly a stand-alone technology for safe drinking water provision [5,10,21], the idea herein is to develop miniaturized units with capacity to clog after a defined operational duration (e.g., t_∞ = 2, 4, 8, 12, 18 or 24 months) and under given operational conditions (e.g., presence of O_2, ambient temperature, water quality). Accordingly, the development of a Fe^0-based filter-clock is therefore regarded as a futuristic independent domain of research in public health engineering.

The principle behind a filter-clock is to let the polluted water flow through two filters: (i) a conventional filter (e.g., a granular activated carbon filter) that mitigates the contamination and (ii) a second Fe^0-based filter (filter-clock) designed to clog promptly after the capacity of the conventional filter is exhausted (at t_∞). Both filters can be combined in a single unit or the order of the relative position of the two adjoining units can be inverted. Figure 1 shows the basic concept of the conventional filter.
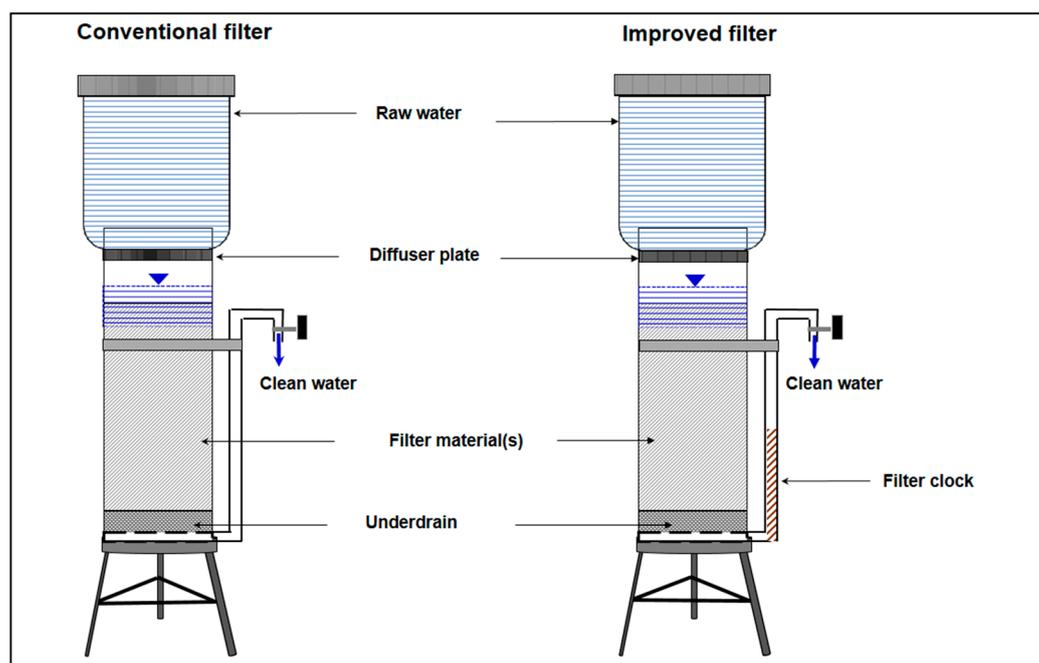
Figure 1. Basic concept of a filter-clock subsequent to a conventional filter. The Fe^0-based filter-clock clogs promptly after the exhaustion of the capacity of the conventional filter (at t_∞). This operating principle corresponds to the one of colored indicators widely used in Analytical Chemistry (titration reaction).

3.2. Design Aspects

The universal equation of a Fe^0 filter is given by Equation (1):

$$V_{oxide} = A^*V_{solid}^*[\tau_{iron}^*(\eta - 1)] \tag{1}$$

where $V_{oxide} = 0$ for $t = 0$ (t_0), t_0 corresponds to the freshly designed filter. Valid V_{oxide} values correspond to $V_{oxide} < V_{pore}$, V_{pore} corresponding to the initial porosity of the filter. t_∞ corresponds to $V_{oxide} = V_{pore}$ and is characterized by system' clogging. t_∞ thus corresponds to the service life of the conventional filter to be equipped with a clock. 'A' is a factor considering the geometry of the Fe^0 particle and its time-dependant variation [13,14]. V_{solid} is the initial volume of solid materials (Fe^0 and other aggregates) and τ_{iron} is the Fe^0 proportion in the solid mixture. η reflects the availability of dissolved O_2 [14].

The primary equation of a filter is given by Equation (2):

$$V_{filter} = V_{solid} + V_{pore} \tag{2}$$

Knowing τ_{iron} ($V_{iron} = \tau_{iron}^*V_{solid}$), V_{filter} (fixed by the designer), the η value (determined by the O_2 level) and assuming spherical particles, V_{pore} can be derived and Equation (1) solved. However, the time-dependent law accounting for FeCPs generation is typically missing [41–45]. Although assumptions can be made using the Faraday Law and current intensity values from the literature, the lack of long-term corrosion data on granular Fe^0 corrosion makes the solution of Equation (1) challenging. This communication is about contributing to solving this challenge by encouraging the scientific community to record data on long-term corrosion of relevant Fe^0 materials under various field conditions. This is establishing t_∞ values for various systems and practically used them on a situation-characteristic basis.

A survey of the literature on Fe^0-based filtration reveals many examples of systems that have clogged after some weeks or months [25,35,46–50]. The remaining task is to bring some systematic

approach in the investigations to come up with a set of applicable tools. This is a typical case where the Know-Why will be derived from the Know-How.

As an example, using a pure layer of iron filings (Fe^0: 6 g) in a small column (inner diameter = 3.0 cm and length = 11.5 cm), Ndé-Tchoupé [39,51] reported on almost complete clogging after 45 days. The Fe^0 layer occupied just 2.2 cm of the column. This result suggested that the material tested by this author can be used as a filter-clock in all cases the filter capacity is expected to exhaust after one month. This system can be used as a reference with Fe^0 admixed with sand in different proportions (e.g., Fe^0:sand 3:1, 1:1, 1:3) in order to obtain filter-clocks corresponding to a longer service life (t_∞ > 4 weeks). Factually, the design from the observations of Ndé-Tchoupé [39,51] should be miniaturized to produce the first generation filter-clock.

4. Conclusions

Metallic iron (Fe^0) has been used for environmental remediation and water treatment for decades. A great deal of research has been done to improve the understanding of the Fe^0/H_2O system in the context of environmental remediation. The lack of a standardised method to characterize the intrinsic reactivity of used Fe^0 materials and their impact of the system's efficiency often causes confusion in interpreting achieved results. This communication introduces the concept of a filter-clock to fix the service life of filtration systems. Fe^0 is chosen because permeability loss due to expansive iron corrosion is currently the major challenge of Fe^0/H_2O remediation systems. In an effort to establish miniaturized Fe^0 filters to assess a filter-clock, reasonable iron corrosion rates for modelling purposes must be known.

It is expected that measured Fe^0 corrosion rates under various operational conditions (including water quality characteristics) will accelerate the development of the Fe^0 filtration technology as a whole. With this simple operational procedure, the service life of all available filters can be known thus fixed. This would be a major contribution to public health, particularly in developing countries, where the provision of safe drinking water remains a challenge. Furthermore, Fe^0-based filter clocks will strengthen frugal innovations for safe drinking water provision worldwide, in particular, for impoverished communities.

Author Contributions: Willis Gwenzi, Rui Hu, Mesia Lufingo, Arnaud Igor Ndé-Tchoupé, Karoli N. Njau, Chicgoua Noubactep and Seteno Karabo Obed Ntwampe contributed equally to manuscript compilation and revisions.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Shannon, M.A.; Bohn, P.W.; Elimelech, M.; Georgiadis, J.G.; Marinas, B.J.; Mayes, A.M. Science and technology for water purification in the coming decades. *Nature* **2008**, *452*, 301–310. [CrossRef] [PubMed]
2. Ali, I. New generation adsorbents for water treatment. *Chem. Rev.* **2012**, *112*, 5073–5091. [CrossRef] [PubMed]
3. Chankova, S.; Hatt, L.; Musange, S. A community-based approach to promote household water treatment in Rwanda. *J. Water Health* **2012**, *10*, 116–129. [CrossRef] [PubMed]
4. Howe, K.J.; Hand, D.W.; Crittenden, J.C.; Trussell, R.R.; Tchobanoglous, G. *Principles of Water Treatment*; John Wiley & Sons, Inc.: Hoboken, NJ, USA, 2012; p. 674.
5. Ali, S.I. Alternatives for safe water provision in urban and peri-urban slums. *J. Water Health* **2010**, *8*, 720–734. [CrossRef] [PubMed]
6. Naseri, E.; Ndé-Tchoupé, A.I.; Mwakabona, H.T.; Nanseu-Njiki, C.P.; Noubactep, C.; Njau, K.N.; Wydra, K.D. Making Fe^0-based filters a universal solution for safe drinking water provision. *Sustainability* **2017**, *9*, 1224. [CrossRef]

7. Ali, I. The quest for active carbon adsorbent substitutes: Inexpensive adsorbents for toxic metal ions removal from wastewater. *Sep. Purif. Rev.* **2010**, *39*, 95–171. [CrossRef]

8. Ali, M.A.; Khan, T.A. Low cost adsorbents for removal of organic pollutants from wastewater. *J. Environ. Manag.* **2012**, *113*, 170–183. [CrossRef] [PubMed]

9. Ali, I. Water treatment by adsorption columns: Evaluation at ground level. *Sep. Purif. Rev.* **2014**, *43*, 175–205. [CrossRef]

10. Banerji, T.; Chaudhari, S. A cost-effective technology for arsenic removal: Case study of zerovalent iron-based IIT Bombay arsenic filter in West Bengal. In *Water and Sanitation in the New Millennium*; Nath, K., Sharma, V., Eds.; Springer: New Delhi, India, 2017.

11. Makota, S.; Nde-Tchoupe, A.I.; Mwakabona, H.T.; Tepong-Tsindé, R.; Noubactep, C.; Nassi, A.; Njau, K.N. Metallic iron for water treatment: Leaving the valley of confusion. *Appl. Water Sci.* **2017**. [CrossRef]

12. Noubactep, C.; Temgoua, E.; Rahman, M.A. Designing iron-amended biosand filters for decentralized safe drinking water provision. *Clean Soil Air Water* **2012**, *40*, 798–807. [CrossRef]

13. Caré, S.; Crane, R.; Calabrò, P.S.; Ghauch, A.; Temgoua, E.; Noubactep, C. Modelling the permeability loss of metallic iron water filtration systems. *Clean Soil Air Water* **2013**, *41*, 275–282. [CrossRef]

14. Domga, R.; Togue-Kamga, F.; Noubactep, C.; Tchatchueng, J.B. Discussing porosity loss of Fe^0 packed water filters at ground level. *Chem. Eng. J.* **2015**, *263*, 127–134. [CrossRef]

15. Ahmad, J.; Goldar, B.; Misra, S. Value of arsenic-free drinking water to rural households in Bangladesh. *J. Environ. Manag.* **2005**, *74*, 173–185. [CrossRef] [PubMed]

16. Ndé-Tchoupé, A.I.; Crane, R.A.; Mwakabona, H.T.; Noubactep, C.; Njau, K.N. Technologies for decentralized fluoride removal: Testing metallic iron-based filters. *Water* **2015**, *7*, 6750–6774. [CrossRef]

17. Pilling, N.B.; Bedworth, R.E. The oxidation of metals at high temperatures. *J. Inst. Met.* **1923**, *29*, 529–591.

18. Caré, S.; Nguyen, Q.T.; L'Hostis, V.; Berthaud, Y. Mechanical properties of the rust layer induced by impressed current method in reinforced mortar. *Cem. Concr. Res.* **2008**, *38*, 1079–1091. [CrossRef]

19. Nichols, W.R. *Water Supply, Considered Mainly from a Chemical and Sanitary Standpoint*; John Wiley & Sons: New York, NY, USA, 1883; p. 260.

20. Devonshire, E. The purification of water by means of metallic iron. *J. Frankl. Inst.* **1890**, *129*, 449–461. [CrossRef]

21. Mwakabona, H.T.; Ndé-Tchoupé, A.I.; Njau, K.N.; Noubactep, C.; Wydra, K.D. Metallic iron for safe drinking water provision: Considering a lost knowledge. *Water Res.* **2017**, *117*, 127–142. [CrossRef] [PubMed]

22. Dahi, E. Africa's U-Turn in defluoridation policy: From the Nalgonda technique to bone char. *Res. Rep. Fluoride* **2016**, *49 Pt 1*, 401–416.

23. Lauderdale, R.A.; Emmons, A.H. A method for decontaminating small volumes of radioactive water. *J. Am. Water Works Assoc.* **1951**, *43*, 327–331.

24. Gillham, R.W.; O'Hannesin, S.F. Enhanced degradation of halogenated aliphatics by zero-valent iron. *Ground Water* **1994**, *32*, 958–967. [CrossRef]

25. Henderson, A.D.; Demond, A.H. Long-term performance of zero-valent iron permeable reactive barriers: A critical review. *Environ. Eng. Sci.* **2007**, *24*, 401–423. [CrossRef]

26. Gheju, M. Hexavalent chromium reduction with zero-valent iron (ZVI) in aquatic systems. *Water Air Soil Pollut.* **2011**, *222*, 103–148. [CrossRef]

27. Ghauch, A. Iron-based metallic systems: An excellent choice for sustainable water treatment. *Freib. Online Geosci.* **2015**, *32*, 1–80.

28. Sontheimer, H.; Crittenden, J.C.; Summers, R.S. *Activated Carbon for Water Treatment*, 2nd ed.; DVGW-Forschungsstelle Karlsruhe: Karlsruhe, Germany, 1988; p. 722.

29. Vidic, R.D.; Suidan, M.T.; Traegner, U.K.; Nakhla, G.F. Adsorption isotherms: Illusive capacity and role of oxygen. *Water Res.* **1990**, *24*, 1187–1195. [CrossRef]

30. Gwenzi, W.; Chaukura, N.; Noubactep, C.; Mukome, F.N.D. Biochar-based water treatment systems as a potential low-cost and sustainable technology for clean water provision. *J. Environ. Manag.* **2017**, *197*, 732–749. [CrossRef] [PubMed]

31. Igalavithana, A.D.; Mandal, S.; Niazi, N.K.; Vithanage, M.; Parikh, S.J.; Mukome, F.N.D.; Rizwan, M.; Oleszczuk, P.; Al-Wabel, M.; Bolan, N.; et al. Advances and future directions of biochar characterization methods and applications. *Crit. Rev. Environ. Sci. Technol.* **2018**. [CrossRef]

32. Tratneyk, P.G. Putting corrosion to use: Remediating contaminated groundwater with zero-valent metals. *Chem. Ind.* **1996**, *13*, 499–503.

33. Fairweather, V. When toxics meet metal. *Civ. Eng.* **1996**, *66*, 44–48.

34. Gheju, M.; Balcu, I. Removal of chromium from Cr(VI) polluted wastewaters by reduction with scrap iron and subsequent precipitation of resulted cations. *J. Hazard. Mater.* **2011**, *196*, 131–138. [CrossRef] [PubMed]

35. Henderson, A.D.; Demond, A.H. Impact of solids formation and gas production on the permeability of ZVI PRBs. *J. Environ. Eng.* **2011**, *137*, 689–696. [CrossRef]

36. Antia, D.D.J. Desalination of water using ZVI, Fe^0. *Water* **2015**, *7*, 3671–3831. [CrossRef]

37. Noubactep, C. Metallic iron for environmental remediation: A review of reviews. *Water Res.* **2015**, *85*, 114–123. [CrossRef] [PubMed]

38. Gatcha-Bandjun, N.; Noubactep, C.; Loura Mbenguela, B. Mitigation of contamination in effluents by metallic iron: The role of iron corrosion products. *Environ. Technol. Innov.* **2017**, *8*, 71–83. [CrossRef]

39. Ndé-Tchoupé, A.I.; Makota, S.; Nassi, A.; Hu, R.; Noubactep, C. The Suitability of Pozzolan as Admixing Aggregate for Fe^0-Based Filters. *Water* **2018**, *10*, 417. [CrossRef]

40. Enthaler, S.; Junge, K.; Beller, M. Sustainable metal catalysis with iron: From rust to a rising star? *Angew. Chem. Int. Ed.* **2008**, *47*, 3317–3321. [CrossRef] [PubMed]

41. Moraci, N.; Lelo, D.; Bilardi, S.; Calabrò, P.S. Modelling long-term hydraulic conductivity behaviour of zero valent iron column tests for permeable reactive barrier design. *Can. Geotech. J.* **2016**, *53*, 946–961. [CrossRef]

42. Noubactep, C. Designing metallic iron packed-beds for water treatment: A critical review. *Clean Soil Air Water* **2016**, *44*, 411–421. [CrossRef]

43. Noubactep, C. No scientific debate in the zero-valent iron literature. *Clean Soil Air Water* **2016**, *44*, 330–332. [CrossRef]

44. Noubactep, C. Research on metallic iron for environmental remediation: Stopping growing sloppy science. *Chemosphere* **2016**, *153*, 528–530. [CrossRef] [PubMed]

45. Noubactep, C. Predicting the hydraulic conductivity of metallic iron filters: Modeling gone astray. *Water* **2016**, *8*, 162. [CrossRef]

46. Westerhoff, P.; James, J. Nitrate removal in zero-valent iron packed columns. *Water Res.* **2003**, *37*, 1818–1830. [CrossRef]

47. Comba, S.; Di Molfetta, A.; Sethi, R. A Comparison between field applications of nano-, micro-, and millimetric zero-valent iron for the remediation of contaminated aquifers. *Water Air Soil Pollut.* **2011**, *215*, 595–607. [CrossRef]

48. Wacław ek, S.; Nosek, J.; Cádrová, L.; Antoš, V.; Černík, M. Use of various zero valent irons for degradation of chlorinated ethenes and ethanes. *Ecol. Chem. Eng.* **2015**, *S22*, 577–587. [CrossRef]

49. Noubactep, C.; Makota, S.; Bandyopadhyay, A. Rescuing Fe^0 remediation research from its systemic flaws. *Res. Rev. Insights* **2017**. [CrossRef]

50. Noubactep, C. Metallic iron (Fe^0) provide possible solution to universal safe drinking water provision. *J. Water Technol. Treat. Methods* **2018**, *1*, 102. [CrossRef]

51. Ndé-Tchoupé, A.I. Conception et Dimensionnement des Filtres à fer méTallique Pour Emploi Domestique. Ph.D. Thesis, Université de Douala, Douala, Cameroon, 2018.