Filtration Process and Alternative Filter Media Material in Water Treatment

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Supplementary Materials

1.   Legislation and Standards for Safe Drinking Water

The first standards for drinking water were established by the World Health Organisation (WHO) in 1958. The "Guidelines for Drinking Water Quality" provided by the WHO did not have any legal status and aimed mainly at guiding countries towards the development of their own regulations [1].

The values reported are generally considered to be measurements at the consumers' taps. This later work presents a shift from maximum levels to guideline values; the shift was maintained in the following editions (1993, 2004) including the most recent one, published in 2011. The number of parameters considered increased in the latest editions (Table S1). It has to be highlighted that guideline values are established by the WHO only when the parameter has a direct effect on human health [2]. Guidance is, however, also provided for the quality from the aesthetic point of view [3].

With regard to the UK and the EU, the defining document in terms of drinking water standards was the Drinking Water Directive (80/778/EEC) in 1980 [4]. The directive set desirable and maximum acceptable values, sampling frequencies and analytical practices for 62 parameters. Limited power was left to governments, with only the possibility of applying more stringent values or set values for additional parameters [2,3]. The revised and current version of the European Directive was passed in 1998 after the 1993 WHO guidelines, and it brought some changes. The number of parameters was reduced. Parameters were divided into mandatory and indicator parameters, the latter not requiring action if the values are exceeded unless human health is at risk.

England, Scotland, Wales and Northern Ireland have separate regulations for water quality, though all of them include the requirements of the 1998 European Directive [5,2]. The standards, called Prescribed Concentrations or Values, are enforced by the Drinking Water Inspectorate (DWI) [1]. It is relevant to mention that not only the final quality of the water is regulated but also the processes and materials used for treatment [2]. This is once again a duty of the DWI, acting not only for England and Wales but also for Scotland on behalf of the Scottish ministers [6]. For Scotland, the standards were initially set into the Water Supply Regulations 2001 [7,8]. This was later substituted by the Public Water Supplies Regulation 2014, setting standards for around 50 parameters with no substantial changes from the previous legislation [8,9,10].

Table S2 summarises the values required by English, EU and Scottish regulations for some of the parameters, which are also discussed in the following paragraphs.

The parameters discussed in the following paragraphs were selected based on their occurrence in the literature; indeed, these parameters are widely used to evaluate the performance of drinking water treatment processes. They give an overview of the removal from the organic, chemical and aesthetic point of view. Thus, based on the literature and on the removal aspects, these parameters were also used as indicators of the media performance in this project.

Table S1. List of substances in the WHO Guidelines, 4th edition, incorporating the 1st addendum [11].

Substances to be Regulated			
Acrylamide	Chloroform	Dichlorprop	Molinate
Alachlor	Chlorotoluron	Di(2-ethylhexyl)phthalate	Monochloramine
Aldicarb	Chloropyrifos	Dimethoate	Monochloroacetate
Aldrin and dieldrin	Chromium	1,4-Dioxane	Nickel
Antimony	Copper	Edetic acid	Nitrate (as NO_3^-)
Arsenic	Cyanazine	Endrin	Nitriloacetic acid
Atrazine and its chroro-s-triazine metabolites	2,4-D	Epichlorohydrin	Nitrite (as NO_2^-)
Barium	2,4-DB	Ethylbenzene	N-Nitrosodimethylamine
Benzene	DDT and metabolites	Fenoprop	Pendimethalin
Benzo[a]pyrene	Dibromoacetonitrile	Fluoride	Pentachlorophenol
Boron	Dibromochloromethane	Hexachlorobutadiene	Selenium
Bromate	1,2-Dibromoethane	Hydroxyatrazine	Simazine
Bromodichloromethane	Dichloroacetate	Isoproturon	Sodium dichloroiscocyanurate
Bromoform	Dichloroacetonitrile	Lead	Styrene
Cadmium	1,2-Dichlorobenzene	Liindane	2,4,5-T
Carbofuran	1,4-Dichlorobenzene	MCPA	Terbuthylazine
Carbon tetrachloride	1,2-Dichloroethane	Mecoprop	Tetrachloroethene
Chlorate	1,4-Dichloroethane	Mercury	Toluene
Chlordane	Dichloromethane	Methoxychlor	Trichloroacetate
Chlorine	1,2-Dichloropropane	Metolachor	Trichloroethene
Chlorite	1,3-Dichloropropene	Microcystin-LR	2,4,6-Trichlorophenol
Trifluralin	Trihalomethanes	Uranium	Vinyl chloride
Xylenes	Burkholderia pseudomallei	Campylobacter jejuni, C.coli	Escherichia coli
Francisella tularensis	Legionella	Leptospira	Mycobacteria
Salmonella Typhi	Other Salmonellae	Shigella	Vibrio cholera
Adenoviruses	Astroviruses	Enteroviruses	Hepatitis A virus
Hepatitis E virus	Noroviruses	Rotaviruses	Sapoviruses
Acanthamoeba	Cryptosporidium hominis/parvum	Cyclospora cayetanensis	Entamoeba histolytica
Giardia intestinalis	Naegleria fowleri	Dracunculus medinensis	Schistosoma

Turbidity: the effluent from a water treatment plant should present turbidity levels lower than 1 Nephelometric Turbidity Unit (NTU). The WHO suggests a median value below 1 NTU for effective disinfection [1]. One of the objectives of filtration within the treatment chain is to reduce the effluent turbidity to less than 0.3 NTU, with a peak limit of 1 NTU. When there is concern over oocysts, such as *Cryptosporidium Parvum* or *Giardia Lamblia*, the target is reduced to 0.1 NTU with acceptable peaks within 0.3 NTU [3,5]. Cleasby & Logsdon encourage considering 0.2 NTU as turbidity breakthrough in order to minimise the risk of oocysts in the effluent. Production of low turbidity water does not, however, ensure their complete removal [12].

Colour: It is generally due to the presence of humic and fulvic acids in the water and can be enhanced by iron or manganese [3]. Though no specific limits are set in the European Directive, in the UK a limit has been set which refers to the measurement of colour with the Platinum/Cobalt method [8].

Table S2. Comparison of several parameters for water quality in the UK and Europe [6,7,8,9].

Parameter	England and Wales, 2010	Scotland, 2014	European Drinking Water Directive, 1998
Colour (mg/l Pt/Co)	20	20	Acceptable to consumers with no abnormal change
Turbidity (NTU)	4 (1 as an indicator parameters at the treatment works)	4 (1 as an indicator parameter at the treatment works)	Acceptable to consumers with no abnormal change, <1 NTU at the treatment works
pH	6.5–9.5	6.5–9.5	6.5–9.5
Aluminium (µg/l)	200	200	200
Iron (µg/l)	200	200	200
TOC (mg/l)	-	No abnormal change	No abnormal change, only for flows >10000 $m^3 day^{-1}$
Total THMs (µg/l)	100	100	100
Coliform bacteria (N/100 ml)	0	0	0
Escherichia Coli (N/100 ml)	0	0	0
Cryptosporidium Parvum	-	-	-
Pesticides total (µg/l)	0.5	0.5	0.5

Among the various metals considered by the legislation, it is particularly important to discuss aluminium and iron. Their impact on treatment is substantial, as their metal ions are the main species used during coagulation. In addition, research has proved that exposure to these metals can have a negative effect on human health; aluminium has shown toxic effects on the nervous system [3].

The presence of aluminium in water supplies is mostly due to its incomplete removal during treatment. Besides issues related to the aesthetic quality of the water, health issues have been connected with exposure to the metal [3,13], though research on the matter is still on-going. Limitations on the concentration of iron are mostly related to the risk of deposits within the distribution systems and the aesthetic quality of the water [3]. Residual iron can create some problems in domestic settings if precipitation is not complete [13]. The WHO guidelines place them both among chemicals for which guideline values have not been established; for aluminium practicable levels are suggested (0.1 mg/l for large treatment facilities, 0.2 mg/l in small facilities) [7]. The European Directive, and consequently the UK legislation, has set a 200 µg/l limit for both the metals, as a preventive measure.

In terms of organic material, the attention is currently focusing on pesticides, though their impact on human health is not wholly understood. Limits have been set for single pesticides and values below 0.5 µg/l are required as total concentration.

There is interest about the removal of NOM, though it is due more to concern over disinfection by-products [2,3,14]. Indeed, the recommended action to limit the formation of DBP is an efficient removal of the organic precursors; especially when it comes to trihalomethanes (THM) [3]. The drive is towards maintaining the final NOM at a concentration as low as possible. NOM is generally quantified via the amount of organic carbon (Total Organic Carbon, TOC) [15]. Dissolved Organic Carbon (DOC) represents the dissolved part and it is operationally defined as the organic matter passing through a 0.45 µm filter [14]. Besides DOC and TOC, other parameters characterising NOM are UV absorbance at 254 nm, pH, turbidity and colour [16]. The concentration of organic matter in

raw water is generally between 0.1 mg/l and 10-20 mg/l [17], the majority of which is removed via coagulation. Specific standards for TOC and generally NOM are not mentioned in the legislation; the only requirement being the absence of abnormal changes.

With regard to microbiological parameters, analyses are focused on understanding which bacteria might come from faecal contamination – animal or human – as its total absence is required. Coliforms are generally measured first. If they are detected the presence of *E. Coli* is investigated, as it confirms the faecal origin of the bacteria [15].

Cryptosporidium is a persistent protozoan, which is difficult to detect. Several outbreaks of the oocyst in the UK at the end of the 1980s prompted analyses on the matter, leading to the publication of reports (the Badenoch reports) in the 1990 and 1995 [18,19]. These included recommendations in terms of filter operations and monitoring, but no standard value. After additional research, a standard of 1oocyst per 10 l based on continuous sampling of 1000 l of effluent was set in the 1999 Water Supply (Water Quality) (Amendment) Regulations [20]. Contrary to the limits in the WHO standards, this is based on operational experience rather than on the infective dose [1]. It was removed as a standard in the revision of the Regulations in 2007, but it has been maintained as a value to strive for [2]. In Scotland the issue is currently addressed by the *Cryptosporidium* (Scottish Water) Directions 2003 [21]. The objective is the introduction of more widespread, frequent and effective testing depending on the risk associated with the treatment plant and with the amount of water treated [10]. The definition of the best practices and trainings required from Scottish Water to avoid possible contamination is also included [21].

In addition to the parameters discussed in the previous paragraphs, new contaminants are being discovered and evaluated as a result of scientific research. Among those, there are pharmaceutical residuals and endocrine disrupting chemicals, which have been linked to an increase in certain types of cancer and increased antibiotic resistance. The variety of compounds and the structures make the removal difficult, though a good performance has been obtained with reverse osmosis, ozone and activated carbon [3]. Several other contaminants classified under chemical (Metaldehyde, Methyl tertiary butyl ether, Perchlorate, Perfluorooctanesulfonate) and microbiological issues (Aeromonas, Arcobacter, Enterovirulent Escherichia Coli, Microsporidia, Cyanobacteria) are presented by Ratnayaka et al. [3]. It is also worth to mention the challenge presented by climate change, through the modification of the weather patterns and the modification of concentration and composition of contaminants.

2.   The Double Layer Model and Hydration Forces Acting onto Particle Surfaces

The van der Waals force is due to the permanent or instantaneous dipole moment generated by electron distributions and acts as an attractive force. The attraction is stronger the closer the particles are to each other [15]. It can occur between two particles or between particle and grain; in the latter case the attraction is twice as strong [22].

The double layer characterisation and interaction is conveyed in the Derjaguin, Landau, Verwey and Overbeek (DLVO) theory. The theory describes the forces involved in the stabilisation of colloids; these have an effect on both the attraction/repulsion between two particles and between particle and grain [15]. The interaction energy for the latter has, however, been calculated as twice that obtained for two particles with similar distance and potentials [22]. The electric double layer is due to the surface charge presented by the particle, as shown in Figure S1. The charge has to be balanced in the solution by an outer layer (Stern layer) of counter charged ions, to maintain a net zero charge. The layer becomes less and less dense the farther away from the surface, creating a second diffuse layer. The Stern and diffuse layers have a difference in potential, called the zeta potential, which represents the potential between the surface and the bulk solution [15,22]. When two particles, or a particle and a grain, approach each other the interaction of the two double layers occurs via the overlapping of their diffuse layers. Both grain and particles generally have negative zeta potential, which works against a possible attachment and becomes more repulsive the shorter the distance between them [23].

Hydration forces tend to repel particles from the surface because of the bond created by water molecules with the surface itself or with the particles [24,25]. They might be opposed by the action of polymers, which, being adsorbed on both particle and surface, create a bridge that draws the particle closer (mutual adsorption) [23]. Polymers can also increase the repulsion via an increase in potential and thus of the energy barrier that has to be overcome, or via steric repulsion [22].

The overall effect of these and other minor forces is dependent upon the distance of the particle from the surface. It also depends upon the particle size; repulsive forces are more effective on small particles transported by diffusion [24,26].
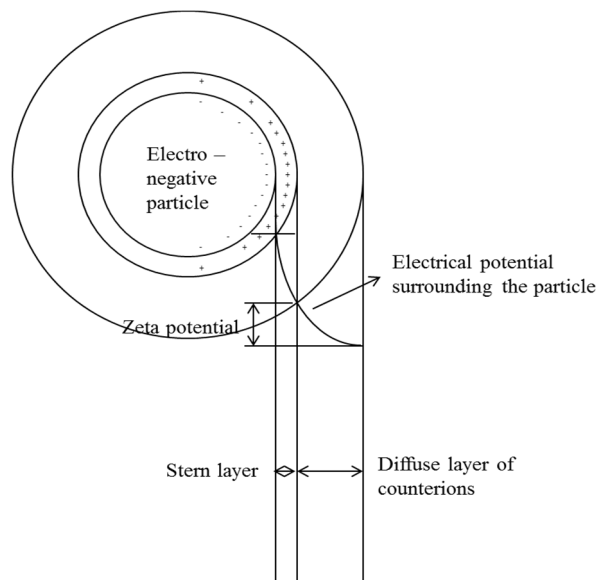
Figure S1. Double layer model [15].

3. Development of Mathematics Models for the Bed Media Filters

Many mathematical models have been developed to predict and evaluate the granulated media filters performance. The model from Mintz [27] and Ives [28] describes the entire filtration cycle using empirical functions derived from bench scale experiments and partial differential equations. Additionally, the model from O'Melia and Ali [29] describes the clean bed filter performance and the ripening stage, which is based on calculations of particle volume balances, applied to a given filter bed depth without considering the entire filtration cycle. Both models did not consider the deposit porosity. On the other hand, the model by Adin and Rebhun [30] describes the bed retention ability and the clogging process during the entire filtration cycle using the bed capacity expressed in weight of accumulated material per unit of bed volume (the specific deposit), and bed hydraulic conductivity expressed in velocity terms.

Using their microscopic model [31] and modifying the model by Adin and Rebhun [30], Vigneswaran and Chang [32] developed a model to describe the entire filtration cycle with experimental results. The model is mainly based on the detachment of deposited particles and on the concept that there exists a maximum limit on particle deposition on the filter grain. Further development of it was the models by Mackie [33], Hunt [34] and Boller and Kavanaugh [35] which express the size-density relationships for aggregated particulate and describes the accumulation of particulate deposits in the pore space of granular media filters. Goodarz [36] used the floc size-density relationship for homogeneous particles and a head loss expression to present the effects of particle size, particle density, filtration rate and media grain size on head loss development during in-line filtration. For the models to evaluate the deep bed filtration of waste water effluent, few cases

emphasized the relation between filtration parameters and sizes of particles in the deposit, though the work by Cikurel et al [37] used existing filtration models to calculate and compare size density and attachment strength of filter deposits, which were produced by the interaction of effluents with primary flocculants and flocculation aids and the filter media during the contact filtration. They found that in order to obtain an effective removal of particles without causing excessive pressure gradients, a correlation must be established for the floc size, density, and strength, and filtration parameters such as bed characteristics and approach velocity.

Although the development of mathematical models is one of important aspects in the fields of filtration for water and waste water treatment, the current manuscript focuses on the alternative filter media development and application. Subject of filtration models should constitute to an independent review and one of such comprehensive reviews can be read in [24]. Efforts are still under way to develop a model that can describe the entire filter cycle and to accurately predict filtration performance; since "the long history in the deep bed filtration research would definitely make this an achievable task in the future" [24].

References

1. Hess, A.; Logsdon, G.S.; Chipps, M.J.; Rachwal, A.J. Filter maintenance and operations guidance manual; AWWA Research Foundation and American Water Works Association: Denver, United States, **2002**.

2. Binnie, C.; Kimber, M. Basic Water Treatment 5th ed.; ICE Publishing: London, **2013**.

3. Ratnayaka, D.D.; Brandt, M.J.; Johnson, K.M. Twort's Water Supply 6th ed.; Butterworth-Heinemann: Oxford, MA, **2009**.

4. Drinking Water Directive (80/778/EEC), **1980**. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:31980L0778&from=EN

5. Twort, A.C.; Ratnayaka, D.D.; Brandt, M.J. Water supply 3rd ed.; Butterworth-Heinemann: Oxford, MA, **2000**.

6. The Scottish Government. Approved Products and Processes for use in the Public Water Supply, **2015**. Available online: http://www.gov.scot/Topics/Environment/Water/17670/approvals

7. Gray, N.F. Water technology 3rd ed.; IWA Pub, **2010**.

8. Water Supply (Water Quality) Regulations, **2001**. Available online: https://www.legislation.gov.uk/wsi/2001/3911/contents/made

9. Public Water Supplies (Scotland) Regulations, **2014**. Available online: https://www.legislation.gov.uk/sdsi/2014/9780111024782/contents

10. The Scottish Government. Legislation, **2015**. Available online: http://www.gov.scot/Topics/Environment/Water/17670/9395

11. WHO, Guidelines for drinking-water quality, 4th edition, incorporating the 1st addendum, **2017**.

12. Cleasby, J.L.; Logsdon, G.S. Granular bed and precoat filtration. In R. D. Letterman, ed. Water Quality and Treatment: A Handbook of Community Water Supplies; McGraw-Hill, **1999**.

13. Tebbutt, T.H.Y. Principles of water quality control 5th ed.; Butterworth-Heinemann, **1998**.

14. Gough, R.; Holliman, P.J.; Willis, N.; Freeman, C. Dissolved organic carbon and trihalomethane precursor removal at a UK upland water treatment works. *Science of The Total Environment*, **2014**, *468-469*, 228–239.

15. Letterman, R.D.; Amirtharajah, A.; O'Melia, C.R. Coagulation and flocculation. In Water Quality and Treatment: A Handbook of Community Water Supplies; McGraw-Hill, **1999**.

16. Matilainen, A.; Vepsäläinen, M.; Sillanpää, M. Natural organic matter removal by coagulation during drinking water treatment: a review. *Advances in Colloid and Interface Science*, **2010**, *159*, 189–197.

17. Volk, C.; Wood, L.; Johnson, B.; Robinson, J.; Zhu, H.W.; Kaplan, L. Monitoring dissolved organic carbon in surface and drinking waters. *Journal of Environmental Monitoring*, **2002**, *4*, 43–47.

18. Badenoch, J. Cryptosporidium in water supplies. Report of the Group of Experts; Department of the Environment, Department of Health: London, UK, **1990**.

19. Badenoch, J. Cryptosporidium in water supplies. Second Report of the Group of Experts; Department of the Environment, Department of Health: London, UK, **1995**.

20. The Water Supply (Water Quality) (Amendment) Regulations, **1999**. Available online: https://www.legislation.gov.uk/uksi/1999/1524/contents/made

21. The Scottish Government. Cryptosporidium (Scottish Water) Directions, **2003.** Available online: https://www.webarchive.org.uk/wayback/archive/20180515204154/http://www.gov.scot/Publications/2004/01/18727/31490

22. Gregory, J. Interfacial phenomena. In The scientific basis of filtration; Noordhoff - Leyden, **1975**.

23. Ives, K.J. Rapid Filtration. *Water Research*, **1970**, *4*, 201–223.

24. Jegatheesan, V.; Vigneswaran, S. Deep bed filtration: mathematical models and observations. *Critical Reviews in Environ. Sci. Technol.* **2005**, *35*, 515–569.

25. Zamani, A.; Maini, B. Flow of dispersed particles through porous media—deep bed filtration. *Journal of Petroleum Science and Engineering*, **2009**, *69*, 71–88.

26. O'Melia, C.R. Particles, Pretreatment and performance in water filtration. *Journal of Environ. Eng.*, **1985**, *111*, 874–890.

27. Mintz, D.M. Modern Theory of Filtration. International Water Supply Ass, 7th Cong. Vol.1 . Special Subject No. 10 (Oct. **1966**).

28. Ives, K. J. Theory of Filtration. International Water Supply Ass, Vienna. (**1969**).

29. O'Melia, C.R.; Ali, W. The role of retained particles in deep bed filtration, *Prog. Water Technol.,* **1978**, *10*, 167-182.

30. Adin, A.; Rebhun, M. A model to predict concentration and head-loss profiles in filtration. *J. AWWA.* **1977**, *8*, 444-451.

31. Vigneswaran, S.; Chang, J.S. Mathemathical modelling of the entire cycle of deep bed filtration. *Wat. Air Soil Pollut.* **1986**, *29*, 155-164.

32. Vigneswaran, S.; Chang, J.S. Experimental Testing of Mathematical Models Describing the Entire Cycle of Filtration, *Water Res.* **1989**, *23*, 1413-1421.

33. Mackie, R.I. Numerical solution of the filtration equations for polydisperse suspensions. 6th World Filtration Congress, Nagoya, Japan, pp. 244-247 (**1993**).

34. Hunt J. The accumulation of solids within deep bed filters. 6th World Filtration Congress, Nagoya, Japan. pp. 236-239 (**1993**).

35. Boller, M.A.; Kavanaugh, M.C. Particle characteristics and headloss increase in granular media filtration. *Water Res*. **1995**, *29*, 1139-1149.

36. Goodarz-Nia, I. Floc density, porosity and void ratio in colloidal systems and aerosols. *J. Coll. Interface Sci.,* **1977**, *62*, 131-141.

37. Cikurel, H.; Rebhun, M.; Amirtharajah, A.; Adin, A. Wastewater effluent reuse by in-line flocculation filtration process. *Water Sci. Technol*. **1996**, *33*, 203–211.