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BIOSCIENCE ENGINEERING

RAINWATER HARVESTING: THE ASSESSMENT OF LONG TERM-TREATMENT EFFICIENCIES AND ITS POSSIBILITIES AS PROCESS WATER

NUMBER OF WORDS: 46 844

Stijn Vanacker
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Promotors: Prof. dr. ir. Stijn Van Hulle, Prof. dr. Ann Dumoulin
Tutor: Ing. Pieter-Jan De Buyck

Master’s Dissertation submitted to Ghent University in partial fulfilment of the requirements for the degree of Master of Science in de industriële wetenschappen: chemie

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Woord vooraf

Er is al veel gezegd, geschreven en onderzoek gedaan omtrent regenwater en het was dan ook een hele uitdaging om alles samen te vatten en te bundelen in een vlot lezend eindwerk. Uiteraard kon ik dit niet alleen en via deze weg wil ik dan ook een aantal mensen bedanken.

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List of abbreviations

BCR	Benefit-cost ratio
BHA	Beta hydroxy acid
BPA	Bisphenol A
BV	Bed volume
CFU	Colony forming units
COD	Chemical oxygen demand
DBPs	Disinfection by-products
DEHP	Diethylhexyl phthalate
DIDP	Diisodecyl phthalate
DIHP	Diisoheptyl phthalate
DINP	Diisononyl phthalate
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
E. coli	*Escherichia coli*
EC	Electrical conductivity
ECF	Electrocoagulation-flotation
FAD	Filtration, adsorption and disinfection
FIB	Faecal indicator bacteria
GAC	Granular activated carbon
GC-MS	Gas chromatography – mass spectrometry
ICP-OES	Inductively coupled plasma – optical emission spectrometry
IEX	Ion exchangers
IOD	Immediate ozone demand
LOD	Limit of detection
LOQ	Limit of quantification
MCPP	Methylchlorophenoxypropionic acid/mecoprop
MF	Microfiltration
NF	Nanofiltration
NOM	Natural organic material
NPV	Net present value
PAC	Powdered activated carbon
PAHs	Polycyclic aromatic hydrocarbons
POM	Particulate organic matter
POPs	Persistent organic pollutants
POU	Point-of-use
RO	Reverse osmosis
RWH	Rainwater harvesting
SODIS	Solar disinfection
SOPAS	Solar pasteurisation
SPE	Solid phase extraction
TCEP	Tris(2-carboxyethyl)phosphine
TCO	Total cost of ownership
TCPP	Tris(1-chloro-2-propyl)phosphate
TN	Total nitrogen
TP	Total phosphorus
TSS	Total suspended solids
UF	Ultrafiltration
UFEB	Up-flow fluidized expanded bed
UV-VIS	Ultraviolet- visible
VAT	Value added tax
VMM	*Vlaamse Milieumaatschappij*
WLC	Whole life costing

Summary

Alternative sources of water such as rainwater are of interest to be used as process water in industrial sectors. Although roof runoff is already fairly clean, concentrations of pesticides, polycyclic aromatic hydrocarbons and phthalates along with soluble heavy metals may be higher than health safety levels due to various pollutant processes. Physicochemical treatment followed by disinfection may therefore be needed before application.

This research mainly focussed on the long term granular filtration of runoff. Rainwater was filtered over a period of two months, both continuously and discontinuously. Discontinuous filtration was implemented to investigate the implications of periods of drought on the effectiveness of the treatment set-up. Continuous filtration was chosen to determine the effects of biofilm growth in the set-up and breakthrough of activated carbon. The second part of the research was dedicated to carrying out filtration tests in order to obtain an effluent which is fit-for-use for the industrial laundry sector. This effluent has to meet a certain range of predetermined quality parameters, both regarding physicochemical as well as microbial quality.

For the long term filtration tests, runoff was spiked with 9 metals and 8 organic compounds (PAHs, diuron, DEHP and DIDP). Treatment consisted of sand filtration followed by granular activated carbon (GAC) filtration both continuously and discontinuously (48 h/week), performed in separate yet identical set-ups. This schedule is based on the real life water consumption pattern for a family of three people. The evolution of pH, turbidity, EC, COD, TN, UV_{254} along with the concentrations of the spiked metals and organic compounds was followed-up. Microbial quality was assessed during the filtration experiment under normal operating conditions and after the performance of a shock chlorination (5 mg/l). UV-irradiation was used as disinfection step to remove pathogens from the continuous GAC-effluent.

The conclusion for the long term filtration tests is that the removal of organic micropollutants decreased over time, as can be expected. More and more adsorption sites became occupied leading to a breakthrough of the GAC. Standard water quality parameters indicated that all effluents were conform the drinking water standards. The removal of metals was fluctuating but for the discontinuous set-up, it could be observed that longer periods without backwashing of the sand filter enhanced the removal by sand filtration. Removal of organic compounds was higher than that of the metals because of the specific type of GAC. A decrease in removal over time was observed for all organics with the exception of the phthalates. When comparing the effluent quality of the long term filtration tests to the drinking water standard, it could be concluded that regarding the standard parameters and metal concentrations, water quality after treatment equalled drinking water quality with an occasional outlier of nickel. However, when comparing the concentration of organic pollutants with the drinking water standards, it became clear that the quality was insufficient. Half of the samples exceeded the limit of total PAHs in drinking water, eight and four samples of the continuous and discontinuous set-up respectively exceeded the concentration limit for pesticides in drinking water. The guideline of the WHO for

DEHP was occasionally exceeded. An important remark here is that the collected runoff was spiked before treatment with concentrations of metals and organics which are relevant according to the literature. The influent concentrations were thus elevated and were not equal to concentrations of runoff found in the literature. The influence of biofilm growth on the removal efficiency should not be underestimated. Because the storage containers were subject to ambient temperatures, accelerated microbial growth was observed. To control this, a shock chlorination of the whole set-up was performed. As a decreasing trend for the removal of organics was observed, predicted breakthroughs for every spiked compound could be calculated, both for the continuous and discontinuous set-up. For the metals, only the breakthrough of copper could be predicted as the removal behaviour of the others was too variable. The breakthrough when using a commercial GAC cartridge on a household level (225 bed volumes per day) was also predicted based on the obtained results from the long term filtration tests. Replacing this type of cartridge in this specific situation should be done after 6 months when considering copper and after 1 year when considering the organics (with exception of the phthalates). Even after UV disinfection, the effluent did not meet the drinking water standard for both total viable count and total coliforms. No microbial activity was detected after performing the shock chlorination.

A laundry company, Excelsior, provided the opportunity to apply the theory regarding RWH treatment with a practical case study. The goal was to determine if simple and robust techniques could be used to achieve a sufficient water quality which met the quality standards as defined by the industry. This water would then be used as washing water in the company and partially substitute the used tap water. Runoff from three different roofs was mixed in a ratio corresponding with the roof surfaces. Treatment consisted of a lava rock filter followed by a sand filter and ultimately GAC filtration. Then, the effluent of the granular filtration was parallelly disinfected with UV-irradiation, hydrogen peroxide, chlorine and ozone. Parameters that were evaluated were pH, EC, turbidity, the concentration of iron, copper, manganese and calcium. Total viable count (22°C) and total coliforms (37°C) were determined to assess the microbial quality.

The effluent of the laundry company case almost met, after treatment with a sand and GAC filter, the predetermined physicochemical requirements. Only the pH and the manganese concentration were too high. This could be solved by using IEX techniques or by adjusting the mixing ratio of the different runoffs. The microbiological quality was only assured by performing a chlorination (2 mg/l). The creation of disinfection by-products and its effect on the washing process should be further investigated.

Samenvatting

Er wordt gekeken of alternatieve waterbronnen zoals hemelwater interessant kunnen zijn als proceswater in industriële sectoren. Hoewel afspoelend hemelwater reeds redelijk zuiver is, kunnen de concentraties van pesticiden, polycyclische aromatische koolwaterstoffen en ftalaten en daarnaast ook van opgeloste, zware metalen een gezondheidsrisico's vormen door het optreden van verschillende vervuilingsprocessen. Fysicochemische behandeling gevolgd door disinfectie kan nodig zijn alvorens het water aangewend kan worden.

Dit onderzoek focuste vooral op de langdurige filtratie van afspoelend hemelwater. Regenwater werd gedurende twee maanden gefilterd zowel in een continue als discontinue uitvoering. Discontinue filtratie werd toegepast om de gevolgen van droogte op de effectiviteit van het behandelingssysteem te onderzoeken. Continue filtratie werd gekozen om de effecten van biofilmgroei en de doorslag van de actieve koolfilter te bepalen. Het tweede deel van het onderzoek focuste op filtratietesten waarbij er getracht werd een effluent te verkrijgen dat geschikt was voor gebruik in de wasserij-industrie. Het effluent moest voldoen aan een waaier van vooropgestelde kwaliteitsparameters betreffende zowel de fysicochemische als microbiële kwaliteit.

Tijdens de lange termijn filtratietesten werd afspoelend hemelwater gespiket met 9 metalen en 8 organische componenten (PAK's, diuron, DEHP en DIDP). De behandeling bestond uit zandfiltratie gevolgd door een actieve koolfiltratie in zowel een continue als discontinue opstelling (48 h/week), uitgevoerd in aparte maar identieke opstellingen. Het discontinue regime was gebaseerd op een bestaand waterverbruiksprofiel van een familie bestaande uit drie personen. De veranderingen in pH, turbiditeit, conductiviteit, COD, TN, UV_{254} en daarnaast ook de concentraties van de gespikete metalen en organische componenten werden opgevolgd. De microbiologische kwaliteit werd onderzocht onder normale werkomstandigheden en na het uitvoeren van een shock chlorering (5 mg/l). Een UV-lamp werd gebruikt als disinfectiestap om pathogenen uit het continu GAC effluent te verwijderen.

De conclusie voor de lange termijn filtratietesten was dat de verwijdering van organische componenten daalde na verloop van tijd, zoals verwacht kon worden. Meer en meer adsorptieplaatsen werden bezet wat leidde tot een doorbraak van de actieve kool. Standaard water kwaliteitsparameters gaven aan dat de waterkwaliteit na behandeling conform was met de drinkwaternormen. De verwijdering van metalen was fluctuerend maar voor de discontinue opstelling kon vastgesteld worden dat langere periodes zonder terugspoeling van de zandfilter de verwijdering van metalen door zandfiltratie verbeterden. De verwijdering van organische componenten was beter door het specifieke type actieve kool dat gebruikt werd. Een dalende verwijdering doorheen de tijd kon waargenomen worden voor alle organische componenten met uitzondering van de ftalaten. Wanneer de effluentkwaliteit van de lange termijn filtratietesten vergeleken werd met de drinkwaternorm, kon er geconcludeerd worden dat wat betrof de standaard kwaliteitsparameters en de metaalconcentraties de waterkwaliteit na behandeling bijna de drinkwaterkwaliteit haalde met hier en daar een overschrijding voor nikkel. Maar wanneer de concentraties aan organische polluenten vergeleken werden met de

drinkwaternormen werd duidelijk dat de effluentkwaliteit wat dit betreft onvoldoende was. De helft van de stalen van beide opstellingen overschreden de maximaal toegelaten concentratie aan totale PAK's in drinkwater, acht en vier stalen van de continue en discontinue opstelling respectievelijk overschreden de concentratielimiet voor pesticiden in drinkwater. Ook de richtlijn van de WHO betreffende DEHP werd occasioneel overschreden. Een belangrijke opmerking hierbij was dat het verzamelde, afspoelend hemelwater gespiked werd voor de behandeling met concentraties aan metalen en organische componenten die relevant waren volgens de literatuur. Hierdoor waren de influentconcentraties hoger en niet gelijk aan de typische concentraties van afspoelend hemelwater die te vinden waren in de literatuur. De invloed van biofilmgroei op de verwijderingsefficiëntie mocht ook niet onderschat worden. Versnelde microbiële groei kon waargenomen worden doordat de opslagvaten onderhevig waren aan kamertemperaturen. Om dit te controleren werd een shock chlorering doorheen het volledige systeem uitgevoerd. Door de dalende verwijderingstrend kon de doorslag voor iedere gespikete component berekend worden, dit voor zowel de continue als discontinue opstelling. Voor de metalen kon enkel de doorslag van koper bepaald worden omdat de verwijdering van de overige metalen te variabel was. De doorslag wanneer een commerciële actief koolfilter gebruikt werd op huishoudniveau (225 bedvolumes per dag) werd ook berekend op basis van de verkregen resultaten van de lange termijn filtratietesten. De voorgestelde filter in deze situatie zou vervangen moeten worden na 6 maanden wanneer rekening gehouden werd met de metalen en na 1 jaar wanneer rekening gehouden werd met de organische componenten (met uitzondering van de ftalaten). Het effluent voldeed niet aan de drinkwaternorm betreffende het totaal kiemgetal en totale coliformen, zelf na disinfectie met UV. Na de shock chlorinatie werd geen microbiële activiteit meer vastgesteld.

Een wasserijbedrijf, Excelsior, bood de kans om de theorie omtrent regenwaterbehandeling om te zetten in een praktische case studie. Het doel was om te bepalen of simpele en robuuste technieken gebruikt konden worden om een voldoende hoge waterkwaliteit te behalen zodat de vooropgestelde kwaliteitseisen van de sector gehaald werden. Dit water zou gebruikt worden in het bedrijf als waswater en zou een deel van het gebruikte drinkwater vervangen als proceswater. Afspoelend hemelwater van drie verschillende daken werd gemixt overeenkomend met de verhoudingen van de dakoppervlaktes. De behandeling bestond uit een lavasteen-filter gevolgd door een zandfilter met tenslotte een actieve koolfiltratie. Het effluent werd parallel gedesinfecteerd met UV-straling, waterstofperoxide, chloor en ozon. De opgevolgde parameters zijn pH, conductiviteit, turbiditeit, de concentraties aan ijzer, koper, mangaan en calcium. Het totaal kiemgetal (22°C) en totale coliformen (37°C) werden bepaald om de microbiële kwaliteit te beoordelen.

Het effluent van de wasserijcase, na behandeling met een zand en actief koolfilter, voldeed bijna volledig aan de vooropgestelde fysicochemische eisen. Enkel de pH en de mangaanconcentratie waren te hoog. Dit zou opgelost kunnen worden door een ionwisselaar te gebruiken of door het mixratio van de verschillende stromen aan te passen. De microbiologische kwaliteit kon enkel gegarandeerd worden door het uitvoeren van een chlorering (2 mg/l). Het ontstaan van disinfectie bijproducten en de typische geur en smaak zouden verder onderzocht moeten worden.

1. Introduction

In 2019, around 785 million people had no access to safe drinking water and 2 billion people used a drinking water source that was contaminated with micro-organisms. Population growth, climate change and urbanization are some of the main reasons that an increased number of people are going to be affected by an elevated water stress by 2025 (World Health Organization, 2019). Besides the direct effects of the lack of potable water, water scarcity could also lead to economic, political and military conflicts in the future (Helmreich and Horn, 2009; Mogelhaard, 2012). To cope with these problems, new water management strategies should be implemented. One of them is the implementation of rainwater harvesting, a practice that goes back to the very beginning of mankind but that has recently gained a renewed interest (Yannopoulos, Giannopoulou and Kaiafa-Saropoulou, 2019). Rainwater is a naturally "clean" source of water in comparison with surface water (Helmreich and Horn, 2009). To date, rainwater is traditionally used for toilet flushing or irrigation of crops (Li, Boyle and Reynolds, 2010). Nevertheless, with an appropriate treatment it is also possible to use rainwater as an alternative source of potable water. This way, millions of people in certain parts of the world could have access to a safer water source. Water scarcity is not the only reason why rainwater has gained interest as an alternative water resource. A second benefit is that rainwater harvesting can attenuate peak flows and the water can be transformed into an asset rather than a problem (Kim, Lee and Kim, 2005). Climate change is starting to affect large economic sectors such as farming, tourism and transport (Hofman-Caris *et al.*, 2019). With rising sea levels and an increased rainfall (both in quantity and intensity) in certain parts of the world, water will become a destructive force for cities and urban regions (Kim, Lee and Kim, 2005). In most cities, the current runoff infrastructure is not designed to handle intense rainfall events. As such, sewer systems will be overloaded and the risk of urban flooding will increase in the future. A third benefit of harvesting rainwater is the possibility of cost saving. In Flanders for example, 817 mm of rain fell on average in 2019 (Brouwers, 2020) and with an estimated total roof surface in Belgium of 624 km² (Van Noordt and Bieseman, 2018), assuming that 80% can be harvested, about 408 million m³ of rainwater could potentially be used instead of tap water (at a tap water cost of €4.3/m³ on average (VMM, 2020a)). This would lead to a saving of roughly €1.75 billion minus the costs related to the treatment, storage or transportation of the rainwater. The most recent numbers on tap water consumption show that 511 million m³ of potable water was consumed in Belgium (353 million m³ in Flanders and 158 million m³ in Wallonia in 2018) (Etat de l'Environnement Wallon, 2019; Statistiek Vlaanderen, 2020). This indicates that 80% of tap water consumption could potentially be replaced by harvested rainwater. As a result, there is an ongoing trend in developed countries towards disconnecting from the central water system and to use the harvested rainwater for toilet flushing, laundry or watering plants (Hofman-Caris *et al.*, 2019). The reason for this is that potable water is relatively expensive in comparison with rainwater which is perfectly fit for irrigation and cleaning. This practice has been widely adopted not only in households but also in larger buildings (e.g. universities, companies, apartment buildings) and this both on a small (single house) and large (neighbourhoods) scale

(Lynch and Dietsch, 2010; Farreny, Gabarrell and Rieradevall, 2011; Rahman, Keane and Imteaz, 2012; Vargas-Parra *et al.*, 2014; Wang and Zimmerman, 2015).

When discussing the reuse and quality of rainwater in literature, the most commonly investigated parameters are standard water quality parameters (e.g. pH, turbidity, electrical conductivity (EC)) and microbiological quality parameters. Nevertheless, recent studies also showed that roof runoff and harvested rainwater can be contaminated by a wide range of organics as well as inorganic micropollutants such as heavy metals, pesticides, polycyclic aromatic hydrocarbons (PAHs) etc (Moreira Neto *et al.*, 2012; Khayan *et al.*, 2019; De Buyck *et al.*, 2021; Tran *et al.*, 2021). Although microbiological contamination of harvested rainwater poses the greatest acute risk to human health when consumed (without the appropriate disinfection), the presence of micropollutants can pose serious health risks in the long term. On the removal of organic and inorganic micropollutants from rainwater, only limited information is available to date. Indeed, the application of treatment technologies for the production of water suitable for various purposes mainly focuses on the adjustment of standard water quality parameters and microbiological control. Also, experiments are nearly always performed within a limited time frame. Either results are given after the immediate start-up of the treatment system or at just one point in time, missing the full scope of the treatment behaviour. In addition, much research has been conducted but no conclusive evidence has been presented to indicate whether rainwater harvesting systems are economically feasible or not. Last but not least, the importance of a legislative framework relating to the further development and implementation of rainwater harvesting systems (RWH-systems) cannot be underestimated. A non-incentivising and unclear set of rules and laws can deter households and the industrial sector from embracing rainwater harvesting as a potential alternative to tap water (Llopart-Mascaró *et al.*, 2010).

Therefore, this study investigates and explores the potential of rainwater harvesting as an alternative source of process water. Longer term filtration tests were performed to see if the effluents from the set-up conformed to predetermined quality requirements. An examination on the behaviour of the less frequently studied pollutants (micropollutants) in filtration systems over time was conducted as well. In addition, a practical case study for a laundry company was included to verify whether harvested rainwater could reach the required quality (put forward by the sector) by the use of different filtration media and disinfection techniques. Furthermore, the economic feasibility of the different treatments was evaluated. Before experimenting, a literature study was performed in which various sources for rainwater runoff contamination are discussed along with an overview of the contaminants and their concentration levels. Furthermore, the current legal framework for rainwater harvesting and reuse and the (quality) requirements for different (industrial) applications are discussed. Next, an enumeration of generally applied techniques and various state-of-the-art treatment technologies and their efficiencies are listed. Finally, the literature study also gives an overview of the typical costs associated with rainwater harvesting and treatments to determine whether an implementation of rainwater harvesting system (RWH-system) is cost effective for the user. Water treatment experiments were performed and the obtained results are discussed and compared with values found in the literature study. A conclusion was then reached whether or not an RWH-system is an economically feasible choice over a long period of time. Also, the analysis of the results obtained from the performed tests concerning the process water for the laundry case led to a proposal regarding the appropriate rainwater treatment technology.

2. Literature study

The first part of the literature study deals with the different types of pollution that can occur during rainwater harvesting. The influence of conventional roofs on the runoff quality is discussed as well as the effect of the first flush. In the second part, an overview of typical rainwater quality parameters is presented. Next, the legislative framework for the reuse of rainwater in Flanders and other countries is discussed along with quality requirements for the use of water in different sectors. Then, state of the art treatment technologies, both physicochemical treatment and advanced disinfection techniques, are reviewed along with their typical removal efficiencies. Furthermore some case studies on rainwater harvesting, where rainwater is used as an alternative source of fresh water, are discussed. Finally this literature study gives a summary of the typical costs of rainwater harvesting and treatment systems.

2.1 Sources of rainwater runoff contamination

Roof runoff can be contaminated by several pathways, such as wet deposition, dry deposition and material leaching. Figure 2-1 summarises these pathways and in the sections below, some more details on the most important contaminant sources are given (De Buyck et al. 2021).

2.1.1 Wet deposition

When raindrops travel through the air, the clean condensed water vapour can be contaminated with pollutants via the phenomenon called washout, especially in industrial and agricultural areas. Air pollution can play a major role in the incorporation of metals and organics such as polycyclic aromatic hydrocarbons (PAHs) in precipitation. The most significant sources for aerial deposition of PAHs are exhaust fumes and industrial aerosols (De Kwaadsteniet *et al.*, 2013; De Buyck *et al.*, 2021). Not only PAHs can be present but also inorganic acids such as nitric (HNO_3) and sulfuric (H_2SO_4) acid and organic acids can be formed due to the presence of precursors such as sulphur, nitrogen oxides (NO_x) and hydrocarbons during complex reactions in the atmosphere (Rocha *et al.*, 2003). As a result, the pH of the rainwater will decrease and acidic rain will form. Parameters affecting deposition include meteorological data (wind velocity, humidity…) and particle characteristics (size and shape). Precipitation scavenging is a process where particles are incorporated in water droplets. If hydrophobic organics adhere to these particles, this process is responsible for the majority of wet deposition and also acts as the most important process for the transportation of these particles to the roof surface. Volatile and semi-volatile organic compounds are thus primarily being washed out from the atmosphere by the process of incorporation in rain droplets. Wet deposition will provide nitrogen and a small part of suspended solids, phosphorus, dissolved organic carbon (DOC) and heavy metals as contaminants to the rainwater before it touches the roofing material (Sánchez, Cohim and Kalid, 2015).

2.1.2 Dry deposition

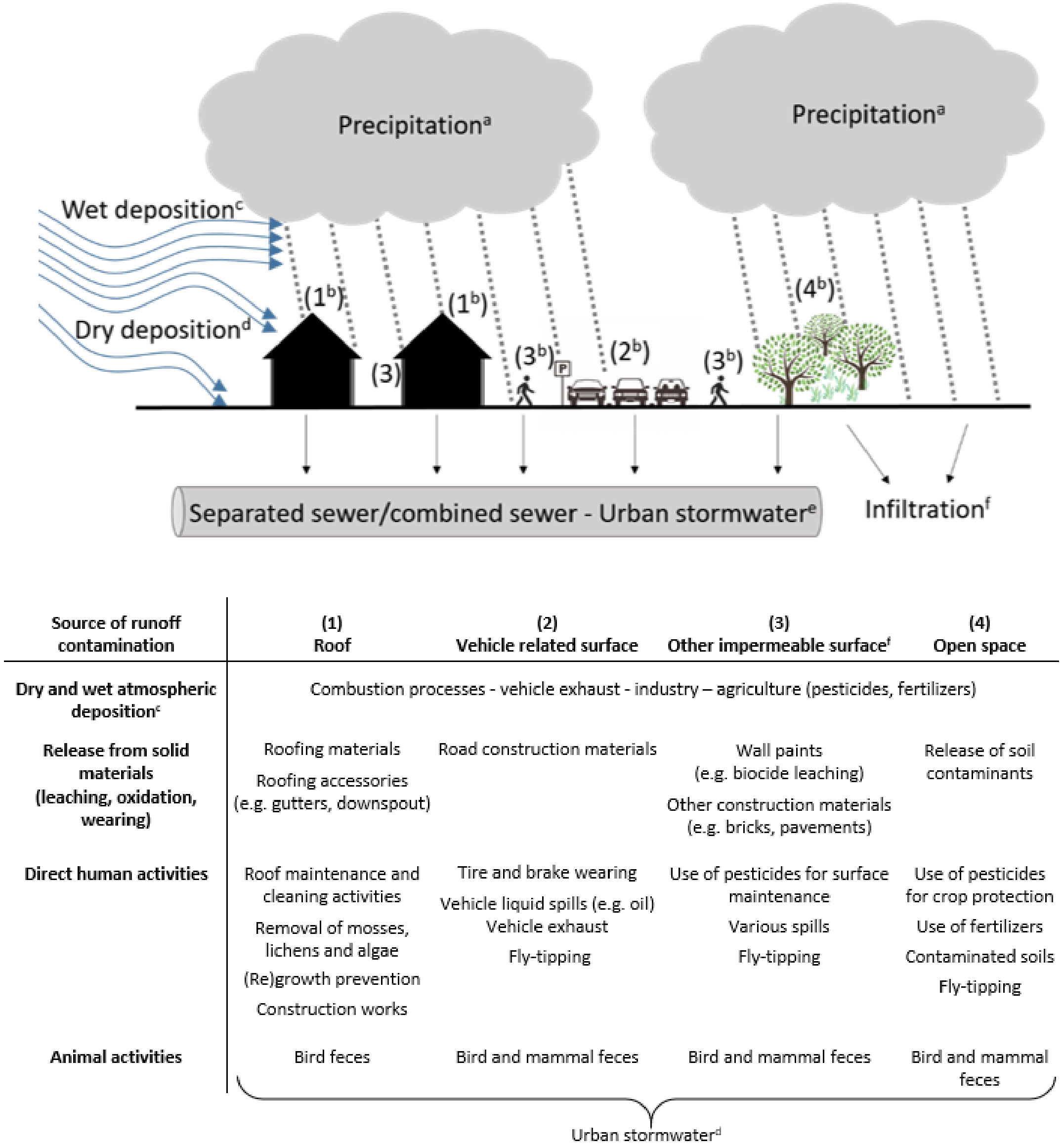
This pathway involves the settling of particles on the catchment surface area. Dust and other particulate matter containing heavy metals all end up on the roof surface. The wind can also play an important role in the transportation of particles over longer distances. Various mechanisms are involved in these movements: turbulent diffusion, Brownian diffusion, sedimentation, inertial forces etc. (Sánchez, Cohim and Kalid, 2015). When rain falls, particles are entrained and flushed into the gutters and storage tanks. The duration of the antecedent dry period is important for the amount of dry deposition: the longer the dry period is, the more particles will settle on the catchment area and the more polluted the runoff will be (Lee *et al.*, 2004). This can be separately collected by installing a first flush device (see 2.2.1) that considers the rainfall pattern on the location of the RWH system (Sánchez, Cohim and Kalid, 2015) and treatment of this volume should be done accordingly. The preference of compounds for the occurrence in the particulate (dry deposition) or gaseous phase (wet deposition) depends on the nature of the compound. High molecular weight compounds are more likely to be present in the particulate phase, low molecular ones in the gaseous phase. Inorganic compounds, e.g. heavy metals, are characterized by their solubility in water, affinity for adsorption onto particles and are affected by the pH. These parameters determine the distribution ratio between dry and wet deposition (Gunawardena *et al.*, 2013).

2.1.3 Coincidental deposition

This form of deposition includes all particles that end up on roofs by chance. Overhanging trees, the presence of different animal species, faeces of overflying birds etc. all contribute to the pollution of rainwater. Animal faeces can be an important source of microbiological contamination while vegetal matter can be a source of natural organic material. This type of pollution is location specific and is not always present in case studies (Schets *et al.*, 2010). This is represented in Figure 2-1 as animal activities.

2.1.4 Material leaching/roof surfaces

Leaching is a process where substances are released from a solid phase by contact with a liquid. In the early stages, residues from the production and instalment process of roofing materials can be washed off. This gives high initial concentrations but quickly drops over time (Gonzalez, Lopez-Roldan and Cortina, 2013). Long term leaching is different as diffusion is the driving factor in this process. As the concentration of the leaching compounds is much higher in the solid phase than in the liquid phase, a concentration gradient is created. This gradient is responsible for the long term leaching of pollutants to the contacting water (Clark *et al.*, 2008). Other factors influencing leaching are the pH (the lower the pH, the more leaching occurs), redox potential of the material, water solubility, temperature, time etc. (van der Sloot and Dijkstra, 2004; Schoknecht and Töpfer, 2012). Different materials mean different compounds that are released into the runoff. As a general rule of thumb, the smoother the area of the collection surface, the smaller the concentration of the released chemicals as roughness leads to a longer hydraulic retention time and increases contact surface (Zdeb *et al.*, 2020). Rainwater samples were examined and it was found that leaching of pollutants increases with the age of roofing materials (Adeniyi and Olabanji, 2005). The construction material of gutters and pipelines also plays an important role in the release of pollutants. Traditionally, gutters are made out of zinc or copper so higher concentrations of these metals are detected in runoff from roofs with these gutters (Mendez *et al.*, 2010). Roof paint also contributes to leaching by releasing organic compounds such as triazines (algicides), carbamates (fungicides), isothiazolines (bactericides), phenylureas (algicides) and triazoles (fungicides). These compounds prevent the growth of micro-organisms on wet exterior surfaces. Paints generally contain a mixture of one to eight biocides with a content of 0.5-1% (Bester, 2014). It is subsequently strongly discouraged to apply such paint when implementing a RWH-system (Simmons *et al.*, 2001). The material of the storage tank is also an import variable in the leaching process. Most commonly, concrete tanks are installed underground and the lime of the concrete can leach into the water which can neutralize acidic rain or in its absence increase the pH of the water. Metals tanks are cheaper, lighter and easier to install but can corrode over time releasing metals in the water and soil. Plastic tanks can provide acceptable water quality provided that the tank is constructed with food grade plastics. Choosing the right tank is important as plastic often contains additives such as plasticizers (DIHP, DIDP), flame retardants (TCEP, TCPP), antioxidants (BPA, BHA) etc. All these compounds have the potential to leach into the water (Hahladakis *et al.*, 2018). Plastic tanks are usually installed underground to minimize algae growth which can deteriorate the water quality (Mosley, 2005). Besides, the presence of pesticides in rainwater was investigated and it was determined that agricultural use of these products was the main source of pollutants, especially during and right after application periods (in the form of deposition). Herbicides such as R-mecoprop and S-enantiomer on the other hand were found in much higher concentrations than in the collected open air samples indicating a leaching process occurred as these herbicides are often used in seals of flat gravel roofs (Bucheli *et al.*, 1998).



Source of runoff contamination	(1) Roof	(2) Vehicle related surface	(3) Other impermeable surface[f]	(4) Open space
Dry and wet atmospheric deposition[c]	Combustion processes - vehicle exhaust - industry – agriculture (pesticides, fertilizers)			
Release from solid materials (leaching, oxidation, wearing)	Roofing materials Roofing accessories (e.g. gutters, downspout)	Road construction materials	Wall paints (e.g. biocide leaching) Other construction materials (e.g. bricks, pavements)	Release of soil contaminants
Direct human activities	Roof maintenance and cleaning activities Removal of mosses, lichens and algae (Re)growth prevention Construction works	Tire and brake wearing Vehicle liquid spills (e.g. oil) Vehicle exhaust Fly-tipping	Use of pesticides for surface maintenance Various spills Fly-tipping	Use of pesticides for crop protection Use of fertilizers Contaminated soils Fly-tipping
Animal activities	Bird feces	Bird and mammal feces	Bird and mammal feces	Bird and mammal feces

Urban stormwater[d]

Figure 2-1: Summary overview of the different sources of pollution regarding precipitation (De Buyck et al., 2021)

2.2 Roof runoff quality

A brief summary of the typical contaminants in roof runoff is given in the following paragraphs. Table 2-1 gives the maximal absolute and the mean value of each basic water quality parameter. Table 2-2 does the same for the concentration levels of soluble metals and Table 2-3 gives the concentration ranges for the organic compounds. All these tables are reproduced from De Buyck *et al.* (2021).

When rainwater hits the roof, it drags along all the build-up of vegetal materials like twigs and leaves but also the majority of the dry deposition and faeces. A large part of these pollutants are washed off in the initial phase of the rainfall and the pollutant load, resulting in a high turbidity, conductivity and suspended solids in the beginning of the rainfall event (Leong *et al.*, 2017) which decreases towards the end of the event (Stenstrom and Kayhanian, 2005).

2.2.1 Turbidity

Turbidity indicates the relative clarity of a water sample and is an optical quality parameter. A high turbidity value means that the rainwater is contaminated physically, chemically or (micro)biologically. The primary pollutants are suspended solids, both organic and inorganic. Physical contamination can originate from animal waste or dust particles while chemical pollutants come from emissions of combustion processes from traffic or industry. Biological interference can originate from airborne viruses and bacteria (Khayan *et al.*, 2019).

2.2.2 Conductivity

Due to the presence of both cations and anions, water can conduct an electrical current. Therefore, conductivity is a non-specific measurement of water contamination by ions as the more ions are present, the higher the conductivity (US EPA, 1992). Ions in rainwater can originate from both natural phenomena such as dust and sandstorms as well as from anthropogenic activities such as industry or traffic (wet and dry deposition) (Sánchez, Cohim and Kalid, 2015). Furthermore, also mammal faeces can contribute to an elevated ion concentration through ammonium and phosphate compounds. In the special case of roof runoff harvested in the vicinity of the sea, a higher conductivity can be observed due to the deposition of salt water aerosols (deposition of NaCl, potassium, magnesium, calcium and sulphate) (Junge and Werby, 1958; Halstead, Cunninghame and Hunter, 2000). Conductivity can have a broad range as can be seen in Table 2-1(De Buyck *et al.,* 2021).

2.2.3 pH

The natural occurring pH of rainwater is controlled by the dissociation of dissolved CO_2 and can only go below 5.6 if there are emissions of acidic precursors in the atmosphere (Khadka, 2020). Acidity of precipitation is strongly correlated to the trajectory that the precipitation followed and to the possible passage over industrialized areas (Pye *et al.*, 2020). Acidic rain is mainly a consequence of human activities where SO_2 and NO_x, originating from exhaust gasses of industry and automobiles, are formed. They are precursors for the atmospheric formation of HNO_3 and H_2SO_4 by photocatalytic reactions (Sánchez, Cohim and Kalid, 2015). The presence of H_2O_2 (caused by the conversion of H_2O by lightning) or O_3 in the atmosphere also has an influence on the formation of acidic rain (Zuo and Deng, 1999). The peroxide is an efficient oxidant in the conversion of dissolved SO_2 to sulphuric acid. The presence of organic acids and aldehydes (which can further oxidize to carbon acids), originating from incomplete combustion processes with hydrocarbons, in the atmosphere of urban areas and their influence on the pH of precipitation has been observed by Peña *et al.* (2002). The most common acids that were found were formic acid, acetic acid and to a lesser extent oxalic acid, lactic acid and citric acid. The most common aldehydes were acrolein, formaldehyde and acetaldehyde.

Alkaline precipitation can also occur. Rainwater with a pH higher than 7 is the result of alkaline particles (NaOH and CaO) or industrial ammonium gasses that are emitted into the atmosphere which are absorbed into the rain droplets. In contrary to these anthropogenic sources, natural sources can also play an important role. Alkaline soil dust found in arid climates or sea components lead to a general higher pH (Zhang, 2019).

2.2.4 Nutrients

The average concentrations of nitrogen and phosphorus found in roof runoff are also presented in Table 2-1. The concentrations are regularly expressed as total nitrogen (TN) and total phosphorus (TP) present in roof runoff. Nutrients can end up in runoff by wet and dry deposition processes. Wet deposition means that gaseous or particulate nitrogen or phosphate are transferred to precipitation while dry deposition is caused by the presence of agricultural or animal activities (Karthikeyan, Balasubramanian and He, 2009).

2.2.5 Chemical oxygen demand as surrogate measurement

Chemical oxygen demand (COD) is a surrogate measurement related to the amount of organic compounds present in water. Runoff from roofs, courtyards and streets with low traffic was studied by Nolde (2007) and it was found that the water showed relative low levels of pollution. The COD of the runoff was on average 14 mg O_2/l (with a maximum of 36 mg O_2/l). Zhang *et al.* (2014) conducted research on what the effect of various roofing materials is on the quality parameters of the effluent. For the COD measurements, the research found that concrete and asphalt roofs gave similar values (around 100 mg O_2/l), ceramic roofs had lower values (50 mg O_2/l) and rainwater originating from green roofs had the highest concentration (150 mg O_2/l).

Table 2-1: Overview of the basic parameters and nutrients levels of rainwater. $c_{cmax,abs}$ stands for the maximal, absolute value that was reported, c_{mean} is the mean value of the summarized literature researched in the review (De Buyck et al., 2021).

Parameter	**Unit**	$c_{max,abs}$	c_{mean}
pH	-	11	6.9
EC	µS/cm	2300	51
Turbidity	NTU	98	46
COD	mg O_2/l	570	96
TN	mg N/l	35	5.4
TP	µg P/l	7660	215

2.2.6 Soluble metals

Sources for the presence of metals in rainwater can vary from dry and wet deposition to leaching of roofing and gutter materials (see 2.1.4 Material leaching/roof surfaces). Metals can be categorized into two classes based on their concentration: the major ions and trace elements. For rainwater calcium, sodium and potassium are considered major ions (mg/l range) while copper, iron, lead etc. are normally classified as trace metals (µg/l range) with the exception when leaching of gutters (copper or zinc) occurs. The presence of trace metals in rainwater can pose health problems, especially when the harvested water is intended for potable purposes as they are not metabolized and can accumulate in soft body tissue. Symptoms of chronic or acute toxicity are different for each metal but can include: reduced growth and development, cancer, damage to organs and nervous system etc. (Malassa *et al.*, 2014). Table 2-2 gives the range of concentration of metals found in roof runoff (De Buyck *et al.* (2021)).

Table 2-2: Concentration range of metals found in roof runoff. $c_{max,abs}$ stands for the maximal, absolute value that was reported, c_{mean} is the mean value of the summarized literature researched in the review in which the metal occurs (De Buyck et al., 2021).

Metal	**Unit**	$c_{max,abs}$	c_{mean}
Al	µg/l	6884	189
As	µg/l	8424	96
Ba	µg/l	2440	22
Ca	mg/l	120	21
Cd	µg/l	1270	20
Co	µg/l	0.76	0.23
Cr	µg/l	1120	17
Cu	µg/l	27177	532
Fe	µg/l	2050	192
K	mg/l	36	5.0
Mg	mg/l	62	1.6
Mn	µg/l	890	24
Mo	µg/l	1.3	3.4
Na	mg/l	390	6.5
Ni	µg/l	43	4.2
Pb	µg/l	2458	25
Rb	µg/l	0.7	0.3
Si	mg/l	36.7	5.2
Sr	µg/l	1700	83
Ti	µg/l	3.8	3.7
V	µg/l	13	2.8
Zn	mg/l	2123.3	1.5

2.2.7 Organic pollutants

When the presence of organic compounds in rainwater is discussed, important groups that deserve attention are the polycyclic aromatic hydrocarbons (PAHs), phthalates and pesticides/herbicides. Regarding PAHs, naphthalene, phenanthrene, benzo[ɑ]pyrene, anthracene, pyrene and fluoranthene can impact various aquatic species when present in concentrations below or equal to 100 ng/l and have a tendency to bioaccumulate. When ingested, these compounds can transform into intermediates with carcinogenic and mutagenic properties inflicting human health problems (De Buyck *et al.*, 2021). PAHs originate from incomplete combustion of fossil fuels in industry/traffic or biomass (Sánchez, Cohim and Kalid, 2015). A negative correlation was found between the presence of PAHs and the volume of the water sample from a rain event by Taguchi *et al.* (2012). The highest concentration is found in the beginning of the precipitation event, the rest of the sample volume (with lower concentrations) serves as dilution. The concentration of PAHs also varies seasonally: increased heating and more traffic during cold weather leads to an increased deposition while an increased exposure to UV-light during warm and sunny weather degrades PAHs (Motelay-Massei *et al.*, 2006; Gaga, Tuncel and Tuncel, 2009). Phthalates are used as plasticizers for plastics and are commonly found in rainwater samples. These compounds can have chronic and acute effects on aquatic organisms with effects reported for diethylhexyl phthalate (DEHP) at < 10 µg/l and for diisodecyl phthalate (DIDP) at < 100 µg/l (De Buyck *et al.*, 2021). Their presence in all sorts of products ranging from children’s toys to packaging materials and the fact that they can leach into the environment means that people are exposed to these compounds their whole life in various ways (dermal/oral contact and inhalation). As these compounds are endocrine disruptors, strict regulations on the presence and use of these compounds were implemented (Fernández-Amado *et al.*, 2017). DEHP followed by DIDP and diisononyl phthalate (DINP) are the most commonly used phthalates worldwide and can be expected to be present in the highest concentrations in roof runoff (Edser, 2017). Herbicides such as diuron can induce negative side-effects in non-target organisms because of the frequent use, persistent character and potential for bioaccumulation of pesticides. These effects were observed in ecotoxicity tests for such pesticides at similar concentration levels as found in roof runoff (e.g. diuron < 20 ng/l) (De Buyck *et al.*, 2021). The presence of different pesticides and herbicides in rainwater was investigated by Scheyer *et al.* (2007) in the Alsace region in France which is a very active agricultural region with crops such as corn, vineyard, vegetables etc. In this research it was found that diuron was detected all year round in contrary to some other compounds like mecoprop (MCPP) which was detected episodically. The high frequency of detection is caused by a low Henry’s constant ($5.1*10^{-5}$ Pa.m^3/mol), according to Scheyer *et al.* (2007), which leads to an easy transfer to water droplets. An important remark is the fact that wet deposition of diuron is in the same order of magnitude for urban and rural areas. Reasons for this are the frequent use of this herbicide in urban weed control and the persistent character of the molecule. Table 2-3 summarizes the maximal observed and mean concentrations of organic compounds found in roof runoff. The concentrations were compiled in a review by De Buyck *et al.* (2021). Table 2-3 also gives the theoretical chemical oxygen demand based on c_{mean} as these compounds are oxidized and make up for a part of the measured COD. Due to the long list of organic compounds that can be present in runoff, only a selection of compounds are presented in Table 2-3. The same selection will be used further in this study

Table 2-3: Maximum observed and mean concentrations of PAHs, pesticides and phthalates in roof runoff. $c_{max,abs}$ stands for the maximal, absolute value that was reported, c_{mean} is the mean value of the summarized literature researched in the review (De Buyck et al., 2021).

Compound	Unit	$c_{max,abs}$	c_{mean}	Molecular formula	Average oxygen consumption (µg/l)
Diethylhexylphtalate (DEHP)	ng/l	4200	266	$C_{24}H_{38}O_4$	0.688
Diisodecylphtalate (DIDP)	µg/l	100	33	$C_{28}H_{46}O_4$	88.8
Diuron	ng/l	5025	2914	$C_9H_{10}Cl_2N_2O$	3.80
Fluoranthene	ng/l	2900	55	$C_{16}H_{10}$	0.161
Fluorene	ng/l	232	23	$C_{13}H_{10}$	0.0687
Naphthalene	ng/l	129	13	$C_{10}H_8$	0.0390
Phenanthrene	ng/l	1600	46	$C_{14}H_{10}$	0.136
Pyrene	ng/l	2300	46	$C_{16}H_{10}$	0.135

Organic compounds are not frequently included in measuring campaigns because of their low concentrations and uncertain effects on health and treatment efficiency. Not much is known about their behaviour in long term treatment operations as they mostly pose chronic health risks. Some halogenated compounds are known as persistent organic pollutants (POPs) which are unsusceptible to photolytic, biological and chemical degradation. They have a high lipid solubility and can thus bioaccumulate in fatty tissue and biomagnify in food chains. POPs not only accumulate in solid matter but are also semi volatile which makes it possible for them to be transported over long distances in the atmosphere before they end up in deposition. This is the reason why POPs are measured all over the world, even in places where there is no production possible like the Arctic (Ritter *et al.*, 1996; Okonkwo, Okafor and Ofodum, 2019). Such POPs may be present in rainwater.

2.2.8 Pathogens

Micro-organisms (e.g. bacteria, viruses and protozoa) in rainwater can seriously deteriorate the water quality and can lead to illness (diarrhoea, vomiting, headache etc.) and even death. Pathogens like *Enterococcus*, *Legionella*, *Escherichia coli*, adenovirus etc. are frequently detected in roof runoff (Dobrowsky *et al.*, 2014; Bae *et al.*, 2019; Hamilton *et al.*, 2019). The sources for these micro-organisms can differ. Faecal indicator bacteria (FIB) are used to assess the microbiological quality when the catchment area is exposed to faeces of mammals and birds. Not only direct contact of faeces with the catchment surface plays a role, distribution by wind is also important (Lye, 2009). Human bacteria however can also be found (e.g. *Campylobacter*, *Legionella* and *Salmonella*). The type of roofing material and its texture also matters: metal roofs often give the lowest concentration of detected micro-organisms as they heat up quicker under the sun (Bae *et al.*, 2019). The smoother the surface of the material, the better the pathogens are washed off during a precipitation event (Egodawatta, Thomas and Goonetilleke, 2009). Table 2-4 gives an overview of some of the maximal observed concentrations of micro-organisms in rainwater according to the literature. Total viable count considers every colony that is cultivable on yeast extract agar when incubating plates at 22°C for 72 hours. Coliforms are part of the genera and species of the *Enterobacteriaceae* and *Escherichia coli* is part of the total coliform count. These organisms grow on a selective medium at 36°C and are incubated for 24 hours.

Table 2-4: Maximum observed concentrations of micro-organisms in roof runoff

Type	**Range of observed concentration (CFU/100 ml)**	**Reference**
Total viable count	$280 - 2.02*10^6$	(Ward, Memon and Butler, 2010; Kaushik, Balasubramanian and Dunstan, 2014)
Total coliforms	$1 – 2.6*10^3$	(Yaziz *et al.*, 1989; Ward, Memon and Butler, 2010; Chubaka *et al.*, 2018; Bae *et al.*, 2019)
E. coli	$0 – 10*10^3$	(Ahmed, Gardner and Toze, 2011; Ahmed *et al.*, 2012; Chubaka *et al.*, 2018; Hamilton *et al.*, 2019)

2.3 Legislative framework

As water scarcity increases in many countries, lawmakers try to encourage people to use alternative sources of water for toilet flushing, irrigation, cleaning, etc. One option is to use harvested rainwater in these applications. As there are no specific laws in Belgium concerning the quality of harvested rainwater, the most used legislation for the quality is the one for potable water. These quality parameters are compiled in Belgium in *Het besluit kwaliteit en levering van water, bestemd voor menselijke consumptie* more commonly known as *Drinkwaterbesluit*. These standards are based on the European framework which is in turn based on the WHO guidelines for potable water. Table 2-5 compares these different potable water quality standards. There are three types of parameters to consider: chemical, microbiological and indicator parameters. Values for microbiological and chemical parameters are quantitative and are explicitly listed in the legislative framework. Indicator parameters are parameters which indicate that there are problems with the production or supply of the potable water. These anomalies can have an effect on public health and should therefore be monitored closely. Drinking water quality standards are always the most strict of all water legislation as there will be a direct contact with the consumer. However, some applications (e.g. first washing water in the food industry, cooling water etc.) should not require drinking water quality as is it considered wasteful to use high-end water for low grade applications. Some examples of applications in the Flemish food industry are shown in Table 2-6. In this sector, alternative sources of water (e.g. harvested rainwater, recycled and treated process water, cooling water etc.) can be used if it is free of micro-organisms (bacteria, viruses, parasites, yeast and fungi) and harmful or toxic components which can have a direct or indirect influence on the quality of food products (Horion, 2005).

Table 2-5: Comparison of some selected quality parameters for potable water that are applied in this research

Compound		**Drinking water guideline WHO**	**EU drinking water standards**	**Flemish drinking water standards**
	Reference	WHO (2011)	EU (2015)	Vlaamse Overheid (2014)
	Unit			
pH	-	-	> 6.5 and < 9.5	> 6.5 and < 9.2
Turbidity	NTU	-	acceptable	acceptable
EC (20°C)	µS/cm	-	2500	2100
Al	µg/l	200	200	200
Cr	µg/l	50	50	50
Cu	mg/l	2	2	2
Fe	µg/l	-	200	200
Mn	µg/l	500	50	50
Ni	µg/l	20	20	20
Pb	µg/l	10	10	10
Sr		-	-	-
Zn	mg/l	3	-	5
PAHs	ng/l	700	100 (total)	100 (total)
Phthalates	µg/l	8 (DEHP)	-	-
Pesticides	ng/l	-	100 (individual) 500 (total)	100 (individual) 500 (total)
Total viable count (22°C)	CFU/ml	-	100	100
E. coli	CFU/100 ml	0	0	0

Although there are no strict quality requirements mentioned in legislation, rainwater harvesting systems have to comply with norms, standards and regulations in many different countries. For Flanders, these are bundled in the *Hemelwaterverordening* from the Flemish Environmental Agency (VMM). Every new house, building or impervious surface larger than 40 m² in Flanders has to comply with this decree since 29/09/2016. The first point is that the installation of a rainwater tank is mandatory when building a new house or renovating one bigger than 100 m² with a capacity of at least 50 l of rainwater per square meter of horizontal roof surface. The second one is that if the total of all impervious surfaces is larger than 250 m², an infiltration unit has to be installed. The goal is to minimize flood risks caused by heavy rainfall by maximizing own use and infiltration and delaying discharge in the sewers (Departement Omgeving, 2013). For the EU, the implementation of a RWH system is described in norm EN 16941-1:2018. This norm specifies the requirements and gives recommendations concerning all aspects of a RWH system (piping, materials, watertightness etc.) for the *in situ* use of rainwater as non-potable water (NBN, 2018). In France, the responsibilities and liabilities of rainwater application and implementation are declared in *Décret n°2008-652*. In Spain, a legislation for the reuse of reclaimed water was put in place with Royal Decree 1620/2007 which makes it possible to use rainwater and other sources of water for low grade applications such as urban, agricultural, industrial, recreational and environmental uses as shown in Table 2-7. Some examples are usage for fire hydrants, irrigation, cooling and cleaning water in the food industry. Every application requires a certain quality and comes with a minimal sampling and testing frequency for each parameter (Llopart-Mascaró *et al.*, 2010; Mujeriego and Hultquist, 2011). Strict quality controls on rainwater however are almost never carried out and the only tool governments have, is to recommend citizens to perform maintenance on their RWH system and to inform them of the benefits and dangers of using rainwater as an alternative source of water (Watergroep, 2021). A good example is that if a private person (less than 10 m³/day or less than 50 people in the building) will use rainwater for drinking water applications in Flanders, a check-up by the owner is required after the installation of the RWH system and after this every 5 (microbiological parameters) or 10 years (chemical parameters) (VMM, 2020b).

Table 2-6: Possible applications for non-potable water in the Belgian food industry as determined by the FAVV (Horion, 2005)

Intended use	**Examples**	**Remark**
Water that may come into indirect contact with a food product which is no basic product	- Cleaning of surfaces, installations, pipes etc. - Cleaning of room with unprotected food present	Final cleanse with potable water
Water that may come into direct contact with a basic product	- Cleaning of whole fishery products - Cleaning of plant based products directly from agriculture (beats, potatoes etc.) - Cleaning of carcasses directly from livestock - Cleaning of living clams before rinsing and transport	
Water that may not directly or indirectly come into contact with food products	- External cleaning of transport - Cooling water, fire hydrants, steam production	Kept in separated circuits

Table 2-7: Maximum acceptable values when reusing water for different purposes in Spain based on Royal Decree 1620/2007 (Mujeriego and Hultquist, 2011)

Intended use	**Intestinal nematodes (egg/10 l)**	***E. coli* (CFU/100 ml)**	**Suspended solids (mg/l)**	**Turbidity (NTU)**	**Other criteria**
Urban use					
Quality 1: Residential - Irrigation of private gardens - Supply to sanitary appliances	1	0	10	2	*Legionella spp.* 100 CFU/l
Quality 2: Services - Urban landscape irrigation - Street cleansing - Fire hydrants - Industrial washing of vehicles	1	200	20	10	
Agricultural use					
Quality 1: irrigation of crops with direct contact with edible parts if eaten raw	1	100	20	10	*Legionella spp.* 1000 CFU/l
Quality 2 - Irrigation of crops eaten after industrial treatment process - Irrigation of pastures for meat or milk producing animals - Aquaculture	1	1000	35	-	*Taenia saginata* and *Taenia solium* 1 egg/l
Quality 3 - Irrigation of tree crops with fruit not meant for direct human consumption - Irrigation of greenhouses, flowers etc. - Irrigation of industrial non-food crops, silo fodder, cereals etc.	1	10 000	35	-	*Legionella spp.* 100 CFU/l
Industrial use					
Quality 1 - Process and cleaning water except for use in food industry - Other industrial uses	-	10 000	35	15	*Legionella spp.* 100 CFU/l

- Process and cleaning water for use in food industry	-	1000	35	-	*Legionella spp.* 100 CFU/l
Quality 2: cooling towers and condensers	1	0	5	1	*Legionella spp.* 0 CFU/l
Recreational use					
Quality 1: golf course irrigation	1	200	20	10	*Legionella spp.* 100 CFU/l
Quality 2: ponds and lakes with prohibited public access	-	10 000	35	-	Total phosphorus 2 mg/l in standing water
Environmental use					
Quality 1: aquifer recharge by percolation	-	1000	35	-	Total nitrogen: 10 mg/l Nitrate: 25 mg/l
Quality 2: aquifer recharge by direct injection	1	0	10	2	
Quality 3 - Irrigation of green areas not accessible to public - Silviculture	-	-	35	-	
Quality 1: other uses (maintenance wetlands, minimum stream flow etc.	Case-by-case basis				

2.4 Treatment of rainwater

Based on the above mentioned information, it is clear that roof runoff can be physicochemically contaminated by a wide range of organic as well as inorganic substances (see 2.2 Roof runoff quality), originating from different sources (2.1 Sources of rainwater runoff contamination). Furthermore, rainwater and roof runoff can also be microbiologically contaminated. This in combination with the current legislative framework and quality requirements for certain applications (see 2.3 Legislative framework), leads to the fact that harvested rainwater needs to be treated prior to its higher-end application. Many options are available but some techniques are preferable, especially in an industrial context. Therefore, the mechanisms, advantages and disadvantages of the techniques are discussed in this section.

Some aspects have to be kept in mind when choosing a treatment process for the reuse of rainwater in an industrial setting. The first one is the removal efficiency for target compounds. It should be known what components could be removed by applying the proposed technique and up to what level. A sand filter is a good choice when the harvested rainwater has a high turbidity (Helmreich and Horn, 2009) whereas activated carbon is a better choice for the removal of metals and organics (Uzun and Güzel, 2000). The order in which units are placed also plays an important role: granular activated carbon (GAC) should always be used after a sand filter as it is wasteful to use expensive GAC as granular filtration medium. It is also important to keep in mind that rainwater is much cleaner than for example surface water. Techniques such as electrocoagulation-flotation (ECF) require a heavier pollutant load in order to work economically feasible and are already used for the treatment of landfill leachates (EMIS VITO, 2010a; Ricordel and Djelal, 2014). Full scale industrial operations are also rare and require a thorough pilot scale research (EMIS VITO, 2010b). Electrodialysis is a robust and simple technique but is mostly used for the separation of higher concentrations of ions and heavy metals. Some techniques (e.g. slow sand filtration, membrane filtration etc.) also achieve high removal percentages of micro-organisms and other components. If complete disinfection is the goal, advanced techniques such as ozonation, chlorination, UV-irradiation etc. could be considered. A second aspect is the robustness of the process. Techniques such as sand filtration and metal or ceramic membranes are easy to install and to maintain whereas polymer membranes are fragile and prone to fouling (Kim *et al.*, 2007). However, polymer membranes can have pores or work on the principle of diffusion which makes them suitable for drinking water production. Simple operation and maintenance are important in industrial applications. Lastly, costs are always a decisive factor in choosing a treatment system as will be discussed in 2.6 Cost analysis. ECF for example could be more costly in electricity consumption to reach the same level of removal efficiency as a sand filter followed by a GAC column and optionally a disinfection step. The latter also varies greatly in price: chlorination has always been a robust and cheap method whereas ozonation is more expensive and complex (Dong *et al.*, 2018). Ceramic membranes are pricier than metal membranes which are in turn more expensive than polymer ones. Table 2-8 gives an overview of some proposed treatment techniques with their advantages and disadvantages. These techniques will be discussed in detail in the following paragraphs.

Table 2-8: Comparison of different physicochemical treatment operations for rainwater treatment

Technique	Rate/ flux	Filtration mechanism	Advantage	Disadvantage	Reference
Slow sand filtration	0.1 m/h	Biological conversion by *schmutzdecke* and regular filtration	- Minimal maintenance - Removal of coliforms - Very low levels of nutrients in effluent - Low cost	- Maturity period - Only water containing low total suspended solids concentration - Need for space	a, b, c
Rapid sand filtration	5-15 m/h	Retention in filtration medium by adhesion, straining, interception etc.	- Conditioning step for disinfection - Combination of filter media	- Need for backwash - Not everything is removed	a, b
Activated carbon	5-20 m/h	Adsorption by Vanderwaals forces	- Selective removal of micropollutants - Reuse possible	- Expensive - Only as polishing step	a, d
Polymer membrane	100 l/m².h	Retention by pores or by diffusion (reverse osmosis)	- Cheaper - Modular construction - Small pores or RO possible	- Low flux - Fouling	a, b, e
Metal membrane	800 - 2000l/m².h	Retention by pores	- More robust - Higher flux - Slower fouling - Easy cleaning	- Expensive - Large pores only	a, f
Ceramic membrane	0.4 l/m².h	Retention by pores	- High flux - Slower fouling - Easy cleaning - Ultra- and nanofiltration possible	- Expensive	a, g

[a]Tchobanoglous *et al.* (2014) [b]Helmreich and Horn (2009) [c]Bauer *et al.* (2011) [d]Uzun and Güzel (2000) [e]Leong *et al.* (2017) [f]Kim, Lee and Kim (2005) [g]Ridder, Heijman and Hofman (2007)

2.4.1 First flush diverter

As discussed in 2.2.1, a large share of the settled dry deposition is concentrated in the first litres of precipitation. When the first flush quantity is diverted, a major improvement in the water quality can be obtained. First flush diverters should operate automatically and can reduce tank maintenance as they retain solid material (e.g. leaves and twigs) which could settle on the bottom of the tank (Helmreich and Horn, 2009). An example of such a device is shown in Figure 2-2. Leong *et al.* (2017) suggested that a device should always be installed after a coarse filter to avoid clogging. How much water should be deviated still remains a point of discussion. For example, a study conducted in Germany found that the optimal first flush was between 1 and 2 mm (Förster, 1999) whereas others concluded that a first flush of 0.11-0.13 mm was sufficient to significantly reduce the total suspended solids (TSS), Mg, Ca and Na concentrations in runoff (Gikas and Tsihrintzis, 2012). The wide range in proposed first flush volumes indicates that location specific factors, such as variation in rainfall intensity, duration and frequency might play a significant role. The diverting of a volume which is too low can lead to a reduced water quality, whereas the diverting of a higher volume has an impact on the available rainwater for (re)use. This should ensure a maximization of water quality without diminishing the water saving potential.

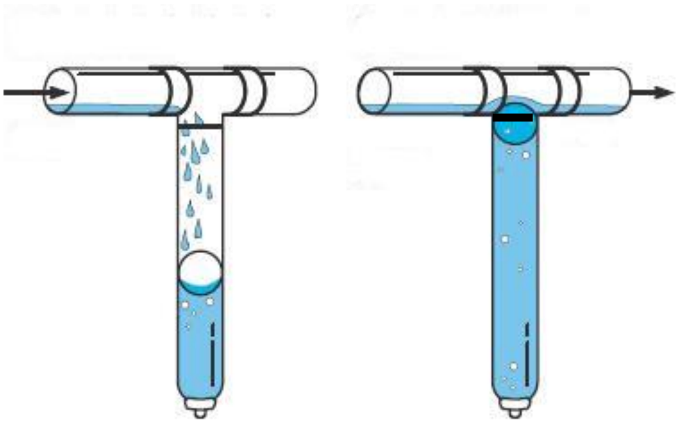


Figure 2-2: Example of an automatic first flush diverter

2.4.2 Physicochemical treatment

2.4.2.1 Sand filtration

Two forms of sand filtration exist: slow and rapid sand filtration. Both are a physicochemical way of treating water by removing solids from the influent. The difference is that a slow sand filter has a lower filtration rate (0.1 m/h) and better bacterial removal rather than a rapid sand filter. A thin biofilm on the top of the filter medium, called *schmutzdecke,* is responsible for the microbiological filtration efficiency (Tchobanoglous *et al.*, 2014).

Rapid sand filtration is more fixed on withholding solids within the filter medium. The name also indicates that the surface loading rate can be much higher than a slow sand filter. Due to the accumulation of solids in the granular medium, a pressure build-up can be observed leading to a backwash. The purpose of the backwash is to wash out the retained particles in the medium. Slow sand filters do not need a backwash, the layer of *schmutzdecke* is scraped off when a pressure build-up is noticed (Barnes, Collin and Ziff, 2009).

2.4.2.2 Activated carbon (AC)

An activated carbon filter is a treatment option for the removal of dissolved substances by adsorption to a solid phase. In this case the solid phase will be activated carbon, this can be powdered activated carbon (PAC) or granular activated carbon (GAC). An advantage of GAC is that it can be regenerated (chemical or thermal) which makes reuse possible. The principle of the removal via AC is based upon adsorption phenomena. These are mass transfer operations as the compounds are transferred from the liquid phase to the solid phase and are bound by weak physicochemical bonds (e.g. London forces). Treatment with AC is mostly considered a polishing step for water that has already gone through various treatment steps. In this polishing step, removal of refractory organic constituents, nitrogen, sulphides and heavy metals as well as odour compounds in water can be obtained. A general rule of thumb for organic compounds is the higher the molecular weight, the better and more efficient the removal will be (Tchobanoglous *et al.*, 2014).

Table 7-1 in the supplementary materials shows the adsorption of both metals and organic compounds to activated carbon. By comparing Langmuir isotherm parameters (especially K_f and 1/n), it becomes clear that organics are better adsorbed than metals. The reason for this is the combination of the charge of metals and the fact that normal AC has no charge. The adsorption of metals can be enhanced by using a more negatively charged carbon (Rivera-Utrilla *et al.*, 2001). Neutrally charged organics are better adsorbed and could be removed from the influent more efficiently. The size of the component besides the size of the pores also matters: smaller metal ions are retained better than ions with a big ionic diameter and organics with a high molecular weight are also better retained than those with a low molecular weight (Uzun and Güzel, 2000; Tchobanoglous *et al.*, 2014).

2.4.2.3 Membrane filtration

A membrane is a physical barrier which allows the passage of certain compounds (permeate) while retaining others (retentate) that are present in a liquid. Three major classes of membranes will be discussed here: polymer, metal and ceramic membranes.

Polymer membranes are categorized into four classes in order of decreasing pore sizes: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reversed osmosis (RO). The mechanism of separation for MF (pore size 0.1 µm) and UF (pore size 0.01 µm) is mostly sieving while NF (pore size 0.001 µm) also operates on the principle of differences in charges. RO is considered a dense membrane with no real pores and should theoretically withhold every compound. Polymer membranes are fragile and should always be placed on a support layer to obtain a high flux. Helmreich and Horn (2009) supported the idea of filtration of rainwater with a MF polymer membrane (pore size 0.1 µm). Protozoa, algae and bacteria can be removed efficiently when low pressure membranes in linear arrays of modules are used. However, viruses still remain an important pollutant and a thorough disinfection is recommended if the rainwater is to be used for potable water applications. Hofman-Caris *et al.* (2019) also states that a RO-filtration requires a remineralization with a filtration over calcite to add minerals to make water suited for drinking water purposes. A subsequent UV-disinfection step is necessary to remove micro-organisms that can start to grow on the calcite filter. A disadvantage of using membranes is the creation of a concentrated brine stream that is considered waste (Anumol *et al.*, 2015).

Metal membranes are a more recent technology and holds a few advantages over polymer membranes. Firstly metal membranes are more resistant to higher pressures (up to 1 MPa) and temperatures (up to 350°C), to outer shocks and chemical oxidation processes e.g. ozonation. In addition, these membranes have a minimal maintenance cost because of their prolonged lifetime. Furthermore, they can be conserved in a dry environment which is an interesting feature considering the fact that precipitation occurs irregularly. Metal membranes are usually submerged in a storage tank and an aeration or ozonation installation can be implemented to improve the filterability and to minimize membrane fouling. A 1 and 5 µm metal membrane was compared by Kim, Lee and Kim (2005) to a hydrophilic hollow fibre polymer membrane with pore size 0.1 µm. They concluded that the polymer membrane (0.1 µm) and the metal membrane (1 µm) had about the same removal of total coliforms (> 98%) while the 5 µm metal membrane had a lower removal efficiency (78%). The transmembrane pressure profiles showed that, although the flux of the polymer membrane was less than 9% of the flux of the metal membrane, the polymer one fouled more rapidly because of the smaller pore size. This indicates that a metal membrane could be a better treatment option because they can operate with a higher flux and have a slower fouling. Another reason is the easier cleaning of these types of membranes by using strong oxidizing cleaning products like chlorine, ozone, hydrogen peroxide and strong acids and bases. In case a more polluted water stream needs to be filtered e.g. green roof runoff, ozone bubbling can be introduced to reduce the transmembrane pressure. The reason is that ozonation, unlike aeration, destroys organic matter and destabilizes colloids leading to less fouling (Kim, Lee and Kim, 2005). Pore blockage was the major fouling mechanism for metal membranes.

Ceramic membranes are membranes made from inorganic compounds (e.g. Al_2O_3, TiO, ZrO_2 and SiO_2) and are used in small scale industrial applications. This type of membrane can be used for the treatment of high temperature, heavily polluted and aggressive wastewaters. Some features of this membrane are a high mechanical strength (up to 90 bar), high chemical resistance (usable in pH 1-4), heat resistance (up to 400°C), long life cycle (more than 15 years), high selectivity and permeability. Recoveries of 99% are possible and fluxes can vary from 50-400 $l/m^2.h$ whereas polymer membranes have a maximal flux of around 100 $l/m^2.h$ so less area is needed when using ceramic membranes. The only downside is the price (€80-100/m^2), which can be up to almost 1.6 times higher than their polymer variants (€50/m^2). Ceramic microfiltration membranes are a good alternative for polymer membranes while ceramic ultrafiltration membranes offer a complete removal of pathogens making the production of drinking water possible. Ceramic nanofiltration can be used to remove solids, viruses and micropollutants and lower water hardness. Ceramic nanofiltration membranes can be combined with ozonation or advanced oxidation processes and can obtain higher fluxes leading to a more compact installation (Ridder, Heijman and Hofman, 2007). The costs of ceramic ultra- and nanofiltration membranes to produce drinking water are high but these can replace the need for sedimentation or disinfection processes in a drinking water treatment plant (Asif and Zhang, 2021).

2.4.3 Disinfection

Disinfection is normally one of the last steps in the treatment process to drinking or process water and has the purpose of removing all pathogens (bacteria, protozoa, viruses etc.) from the rainwater before the intended use. Some of the aforementioned physicochemical treatment options can already remove a significant amount of pathogens but an advanced disinfection step is needed in order to use the rainwater as potable water or for other applications (see Table 2-6 and Table 2-7). These aqueous organisms can pose serious health threats to humans (Liu *et al.*, 2020). The majority of disinfection processes is based on damaging the DNA-structure or by destroying the cell material. There are many different options to disinfect water: chlorination, ozone, UV, H_2O_2, pasteurization, solar disinfection (SODIS) etc. The associated costs to use a certain disinfection technique differ. A way of comparing the effectiveness of treatment options where a chemical compound is added or a technique is applied, is by using CT-values for a certain removal. CT-values are the product of the residual concentration of the disinfectant (C) and the contact time (T) according to the Chick-Watson expression and use mg/l*min as unit (Tchobanoglous *et al.*, 2014). Table 2-9 compares the CT-values required for a certain log removal of pathogens by using different disinfection techniques.

Table 2-9: Comparison of CT-values required for a certain removal of pathogens using various disinfection techniques (National Research Council (U.S.) Safe Drinking Water Committee, 1980; Tchobanoglous et al., 2014; Haq, 2017)

Disinfectant	**Pathogen**	**CT-value**	**Removal (log units)**
Chlorine (mg/l*min)	Bacteria	1.6-2.4	4
	Virus	3-3.5	4
	Cryptosporidium	4000-5000	2
	Giardia lamblia	70-80	3
Ozone (mg/l*min)	Bacteria	0.02-0.04	4
	Virus	0.5-0.6	4
	Cryptosporidium	12-13	3
	Giardia lamblia	0.75-0.8	3
UV-radiation (mW/cm²*min)	Bacteria	2400-3600	4
	Virus	4800-60 000	4
	Cryptosporidium	720-780	3
	Giardia lamblia	660-780	3
Pasteurization (°C*s)	Bacteria	< 130 – 20 880	3
	Virus	2250	3
	Cryptosporidium	18 000	4
	Giardia lamblia	33 600	2
Hydrogen peroxide (mg/l*min)	Bacteria	900	2

2.4.3.1 Chlorination

Chlorination is the addition of chlorine compounds (e.g. NaOCl, $Ca(OCl)_2$, Cl_2, ClO_2) to water. Disinfection by chlorine is obtained as chlorine compounds enter the cell and substitute hydrogen atoms in proteins, effectively changing the whole molecule by converting enzymes until the cell dies. Neutrally charged hypochlorous acid can better penetrate the negatively charged cell membrane of micro-organisms than the negatively charged hypochlorite ion (Tchobanoglous *et al.*, 2014). Although chlorination is the most frequently used and cheapest disinfection method to date, the creation of toxic disinfection by-products (DBPs) such as trihalomethanes pushes the industry to turn away from chlorination towards other methods of disinfecting water. DBPs are created when chlorine reacts with natural organic material (NOM). NOM consists of lots of different compounds but humic and fulvic acids seem to be the major reagents. The formation of DBPs is dependent on many variables: type of disinfectant, dose, residual concentration, contact time, temperature, pH and the concentration of NOM in the water (Kim, Shim and Lee, 2002). Especially with rainwater, where organic material is almost always present, a removal step for these DBPs is necessary. This can be done by placing an activated carbon filter before (to remove organic material) or after the disinfection step (to remove DBPs). Not only should the trihalomethanes be removed, the residual chlorine concentration in the water should also be controlled as chronic overexposure may result in bladder, colon or other types of cancer (Villanueva *et al.*, 2015). This is also the reason why many countries have adopted other methods of disinfection although the costs may be substantially higher. Addition of chlorine to water will not result immediately in a removal of pathogens. A breakpoint chlorination must therefore be performed to determine the amount of chlorine needed. An advantage worth noting, is the fact that residual disinfection is possible and so the regrowth of pathogens in pipelines or storage tanks can be kept to a minimum. The residual chlorine concentration should be between 0.2 and 0.5 mg/l as this is the optimal concentration for residual disinfection without excessive odour or taste complications (WHO, 2011b). Keithley et al. (2018) determined that the immediate chlorine demand of rainwater is around 0.5 mg/l. Around 1 or 2 mg/l of chlorine are typical dosages for disinfection of rainwater. The World Health Organization recommends a guide value of 5 mg/l for the chlorination of drinking water (Fawell, Lund and Mintz, 2003).

2.4.3.2 Ozonation

Ozonation is another method for the disinfection of rainwater and is the strongest oxidant in comparison with all the different disinfections agents. Because ozone is a very unstable gas, it must be generated *in situ* via a corona discharge mechanism. Advantages for the use of ozone are that pathogens are more susceptible to ozone than chlorination, the possible removal of taste and odour components and the faster reaction than chlorine. Disadvantages are the point of use generation and thus higher operational costs, the fact that residual ozone must be removed and that it has no residual disinfection properties (Haq, 2017). Another disadvantage is the formation of DBPs such as bromate, haloacetaldehydes and halonitromethanes that can be formed during disinfection (Li and Mitch, 2018). The disinfection mechanism is caused by either the creation of a hydroxyl radical which has a strong oxidizing potential or by the oxidizing effect of ozone itself. Bench or pilot set-ups are required to determine the immediate ozone demand of the water, the transfer of ozone to the water and the ozone decay profile (Tchobanoglous *et al.*, 2014).

2.4.3.3 UV-disinfection

UV-disinfection is an effective method for the removal of bacteria, viruses and protozoa while not forming any toxic by-products or introducing disinfectant-resistance to micro-organisms. UV-radiation is the area of the electromagnetic spectrum between 100 and 400 nm. The germicidal portion ranges from 220 to 320 nm and the most effective wavelengths for inactivation are between 255 and 265 nm (Tchobanoglous *et al.*, 2014). Photochemical reactions are the cause of the germicidal effect on biomolecules resulting in inhibition of growth or complete inactivation dependant on the applied dosage. These photochemical reactions have free radicals as intermediates and cause chain reactions if an absorbing target is present. In living cells these targets are DNA, RNA and proteins which strongly absorb in the 260 nm region of the spectrum (Meulemans, 1987). Various setups can be introduced depending on the volumes that need to be treated: this can go from a simple single low pressure UV-lamp for point-of-use systems (POU) to systems of hundreds of lamps in an open or closed configuration for the disinfection at a potable water plant. LED UVC is a more recent alternative to traditional mercury lamps. It is a technique that is more cost effective, compact, has a longer lifetime and does not contain toxic mercury. Wavelengths between 210 and 365 nm are available, depending on the used semiconductors (Song, Mohseni and Taghipour, 2016). The quality of water also plays a major role in the removal efficiency. UV-radiation cannot reach micro-organisms if a high concentration of solids is still present as they are shielded from the UV-light by these solids (Tchobanoglous *et al.*, 2014). An important advantage of using UV and the reason this technology has gained renewed interest is its ability to extensively inactivate two chlorine resistant protozoa: *Cryptosporidium parvum* and *Giardia lamblia* (Naddeo, Scannapieco and Belgiorno, 2013).

2.4.3.4 Pasteurization

Pasteurization, which is the oldest form of disinfection, uses heat to inactivate micro-organisms. By heating water to temperatures below the boiling point (70-80°C), many indicator bacteria such as *E. coli* and heterotrophic bacteria can be inactivated. Micro-organisms are killed because their functional proteins (e.g. enzymes) are inactivated by denaturation caused by high temperatures. This technique was introduced in developing countries as the sun is a free and natural source of heat. These systems, the so-called solar pasteurization (SOPAS) systems, are robust and require no or little maintenance which is ideal in rural areas. SOPAS is different from solar disinfection (SODIS) as SODIS uses both the heat and the radiation of the sun for inactivation of the pathogens. Systems become even cheaper when thermo-siphoning is used instead of pumps. As warm water has a lower density than cold water, a system can be designed that heats up entire storage tanks without the need for pumps (Dobrowsky *et al.*, 2015). Again, as with disinfection using chlorine, ozone or UV-radiation, the performance of a pasteurization is dependent on both the temperature and the holding time. The temperature is needed for the denaturation of the enzymes in the cell protoplasm and to change the structure of the cell wall. The holding time is essential for the completion of the reaction (Haq, 2017).

2.4.3.5 Hydrogen peroxide

Mostly used as an oxidant for the treatment of wastewater containing cyanides, formaldehyde, hydrogen sulphide and phenols, hydrogen peroxide is a well-known compound in the paper and textile industry as bleaching agent. It has a lower redox potential than ozone (2.1 V) but higher than chlorine (-derivatives) (1.4 – 1.5 V). When a catalyst (typically Fe^{2+} or Cu^{2+}) is added, the process is called the Fenton process and the possibility to form hydroxyl radicals increases the redox potential to 2.8 V. A disadvantage of this process is the amount of sludge that is formed. The required concentration for an inactivation of 99% of *E. coli* was 90 mg/l with a contact time of 30 minutes (National Research Council (U.S.) Safe Drinking Water Committee, 1980). Advanced oxidation processes are applied when degradation of synthetic organic compounds (e.g. pharmaceuticals) is required. These potentially harmful substances can be successfully destroyed by using a combination of UV and H_2O_2. Hydrogen peroxide is split up in hydroxyl radicals due to a photolysis reaction caused by UV-radiation. The radiation can thus work simultaneously as disinfectant and catalyst by breaking down hydrogen peroxide (Mierzwa, Rodrigues and Teixeira, 2018).

2.5 Case studies and associated removal efficiencies

2.5.1 Review of the case studies

In this section a review of different state-of-the-art strategies for rainwater treatment and their removal efficiency is discussed. When looking at the summary presented in Table 2-10 it can be seen that all studies apply robust, cost efficient and proven water filtration and disinfection techniques. The main reason for this is to reduce purchase and maintenance costs in order to substitute a part of the water demand at an affordable cost that can compete with tap water. Furthermore, most studies used a combination of different techniques to treat rainwater.

When looking at the reported parameters and removal efficiencies in the studies presented in Table 2-10, it becomes clear that the majority of the research in the field of rainwater treatment focusses on standard water quality parameters and microbiological quality. Information regarding inorganic micropollutants on the other hand is only limited available while no information regarding organic micropollutants was found in the literature. From the consulted literature, only Rasid *et al.* (2009) and Khayan *et al.* (2019) offered information on the removal of Fe and Pb respectively.

The sand filter combined with GAC investigated by Khayan *et al.* (2019) showed a high pH adjustment. This was attributed to the use of mollusc sand and the associated carbonate groups provided by the mollusc. In this study, also the removal of Pb was investigated as the used zinc roofing materials were treated with a Pb containing coating. The reported removal efficiency for Pb was high (>99.4%) and the rainwater, after filtration with mollusc and GAC, met the local drinking water quality standard for Pb. Also in the study by Rasid *et al.* (2009) the removal of a metal (Fe) during rainwater treatment was investigated. Here, the removal of Fe was attributed to the presence of biofilm on the GAC where the metals are used as metabolites or are neutralised in the biofilm to prevent toxic growth conditions for the micro-organisms. Because of the specific configuration of the system (up-flow fluidized bed reactor with GAC and biofilm, followed by a down flow slow sand filter), the set-up could be operated for 390 days, without the need to replace the GAC.

As mentioned above, the other studies mainly focus on standard water quality parameters and microbiological quality of harvested rainwater. For example, the objective of the study of Moreira Neto *et al.* (2012) was on the removal of micro-organisms to obtain bacterially safe water to use for toilet flushing and cleaning of public areas (airport). Microbiologically safe water is also important when rainwater is used for drinking water purposes. Another study by Barnes, Collin and Ziff (2009) investigated the removal of micro-organisms by several techniques in order to have microbiologically safe drinking water for local communities in Ghana. Here, it was found that the slow sand filter had better removal efficiencies for micro-organisms than the tested siphon ceramic filter. A siphon filter is a household water filter using a ceramic membrane. The filter is placed in a container with water to be filtered and by using a rubber bulb, which act as pump, the water is pulled through consecutively a cloth prefilter and the ceramic membrane. Another example where rainwater was treated in order to obtain microbiologically safe water for communities or households, was studied by Naddeo, Scannapieco and Belgiorno (2013). In this study, a combination of microfiltration (MF), followed by GAC, which in turn was followed by a UV disinfection step was used. This combination is also known as a filtration, adsorption and disinfection (FAD)set up which is shown in Figure 2-3. In contrast to the previously mentioned studies, also some physicochemical water quality parameters were considered next to the microbiological parameters.

The other studies presented in Table 2-10 mainly focussed on the effects of the applied treatment on standard water quality parameters. For example, Teixeira and Ghisi (2019) compared the removal of several standard water quality parameters from rainwater by a sand and GAC filter with a polyester membrane filter (see Figure 2-4). It was found that the sand filter had the highest removal of turbidity and phosphate while the membrane filter had the biggest pH adjustment and nitrate removal. Both set-ups were equipped with a first flush device (discarding the first 2 mm) and the membrane filter could be backflushed automatically by using a system of floats. The membrane filter had to be replaced by the end of the operation period (approximately 4 months) while the sand filter could run continuously. The sand filter was thus considered the cheapest and more robust option.

Next to traditional rainwater harvesting systems, some studies also investigate the potential of greywater in combination with rainwater. For example, Kim *et al.* (2007) mixed greywater and rainwater (1:1) and treated it with a wood fibre filter and a metal membrane. The recycled wood fibre was modified with Al_2O_3 for enhanced removal of phosphate and heavy metals and was meant for treating first flush volumes. It succeeded in removing a part of nutrients and turbidity. This effluent was not good enough for non-potable applications (e.g. laundry or shower). In order to improve the quality of the water, different metal membranes with varying pore sizes were used to filter the effluent of the wood fibre filter. The removal or adjustment was dependable on the pore size: the larger the pore size, the lower the rejection but also a slower fouling occurred. Small adjustments in pH and EC were observed with higher removals of turbidity and COD. Aeration and ozonation were tested to reduce fouling: ozonation had a higher observed effect than aeration as ozone bubbling not only creates turbulence around the membrane but also causes degradation of solids and colloids.

2.5.2 Application areas and challenges for rainwater utilization

Applications of rainwater after treatment consist mainly of domestic use (showers, irrigation, laundry etc.) or as replacement of potable water (Barnes, Collin and Ziff, 2009; Khayan *et al.*, 2019). The study by Moreira Neto *et al.* (2012) and Kim *et al.* (2007) focusses more on large scale rainwater treatment such as airports or office buildings. However, the applications of the rainwater still remain the same as in the households like toilet flushing, floor cleaning etc. Not one industrial application for the harvested rainwater was found in the literature.

Furthermore, the ease of use can also be a major constraint, especially in households where RWH systems are used by people who generally have no technical background. Metal membranes investigated by Kim *et al.* (2007) are difficult to operate on household level especially in the case where the membranes are combined with aeration or ozonation for enhanced removal. The siphon ceramic membrane and the bio-sand filter (Barnes, Collin and Ziff, 2009) on the other hand can be operated with minimal technical knowledge in places with no basic facilities (e.g. running water, electricity etc.). The other treatments discussed above, lay somewhere in between these extremes. Membrane and sand filtration are easy to operate as backflushing or replacements of membrane can be performed easily when guidelines are provided (Khayan *et al.*, 2019; Teixeira and Ghisi, 2019). A more difficult and delicate technique is the GAC biofilm combined with slow sand filtration by Rasid *et al.* (2009) as long maturation periods and optimal process conditions are required to guarantee optimal removal efficiencies. However, some of these challenges can be resolved with the introduction of remote control and online monitoring (Kasmin, Bakar and Zubir, 2016; Chandrika Kota *et al.*, 2020).

Another hurdle which was discussed in the study by Moreira Neto *et al.* (2012), is the fact that the water supply is not guaranteed, due to fluctuations in water supply during dry and wet months. Harvesting of runoff from other surfaces (e.g. parking spaces and runways in the case of an airport), in combination with larger storage capacity, are one option to guarantee a constant supply of rainwater. This is why the correct sizing of the rainwater tank is paramount as it is one of the biggest components of the capital costs. Sizing a tank varies from location to location as differences in meteorological data, the area of the catchment surface and the water demand are the three most important parameters (Campisano and Modica, 2012). However, these streams are of a poorer quality and thus require more thorough treatment (Moreira Neto *et al.*, 2012). As an alternative, also grey water recuperation, in combination with rainwater can be used to overcome the dry periods. The last alternative is a top-up function by mains water supply. But this function also has negative consequences besides being a reliable back-up. The first one is the long residence times of mains water in the central water supply in times of intense rainfall when RWH is the most active. The second one is the requirement of a second, separated, local distribution net for treatment and transport in the case of treatment to potable water as this type of water cannot be transported through the tap water distribution net (Hofman-Caris *et al.*, 2019).

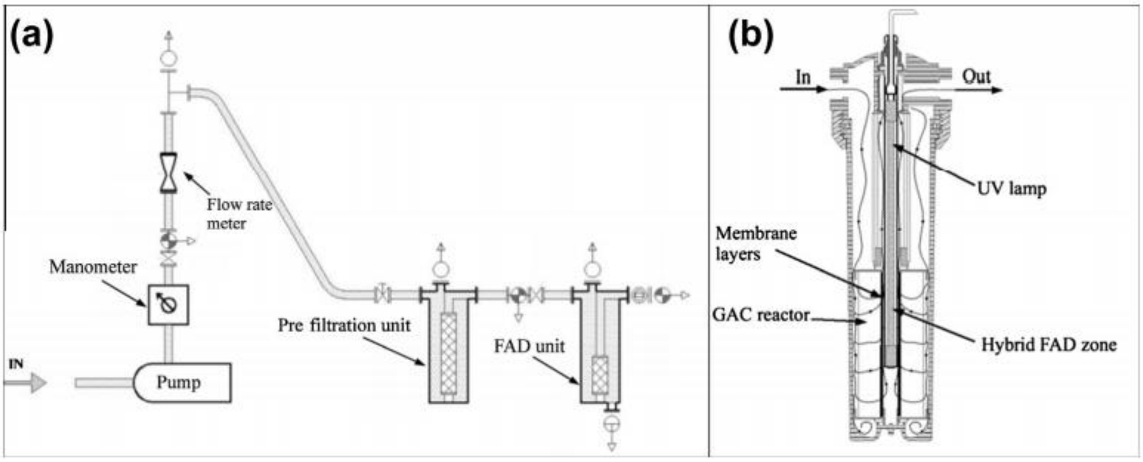


Figure 2-3: Set-up of the FAD-unit (Naddeo, Scannapieco and Belgiorno, 2013)

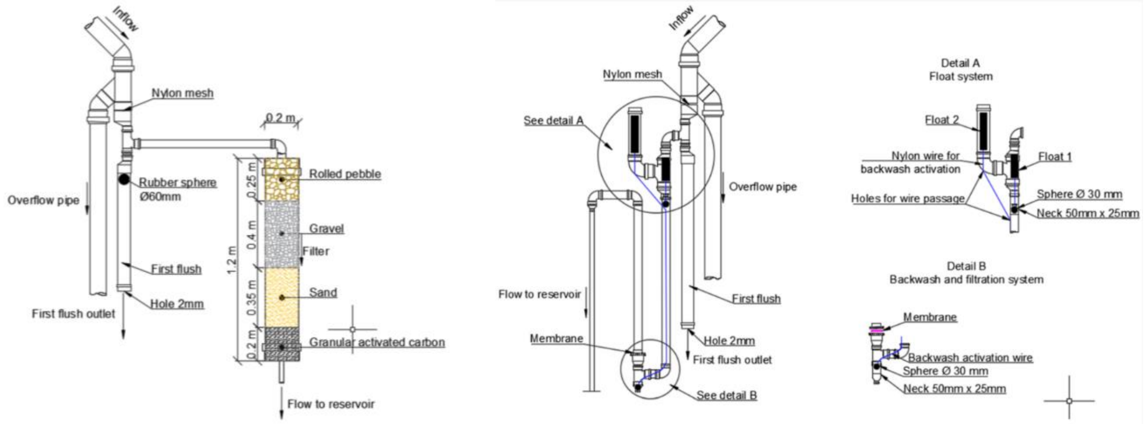


Figure 2-4: Comparison of sand (left) and membrane filter (right) (Teixeira and Ghisi, 2019)

Table 2-10: Removal efficiencies for state of the art rainwater treatment systems found in the literature

Technique	**Scale – Catchment surface**	**Removal efficiencies**			**Reference**
		Standard parameters (%)	Metals (%)	Micro-organisms (log-units)	
Slow sand filtration - chlorination	Airport Brazil Runway runoff Pavement runoff	-	-	*E. coli*: 3-4 Total coliforms: 3-4	(Moreira Neto *et al.*, 2012)
MF (0.5 μm) – GAC - UV	Rooftop of small building in Salerno, Italy	COD (GAC): 22.8 ± 8.7 COD (FAD): 49.9 ± 7.5	-	*E. coli*: 4 Total coliforms: 4	(Naddeo, Scannapieco and Belgiorno, 2013)
GAC biofilm – slow sand filtration (UFEB)	Household roof runoff in Kuala Lumpur, Malaysia	COD: 37.2 – 92.6 NO_3^-: 51.8 – 83.0 NO_2^-: 2.5 – 65.9	Fe^{2+}: up to 75.49	-	(Rasid *et al.*, 2009)
Mollusc sand filter – GAC	Household roof runoff in Indonesia (galvanised steel roofing)	Turbidity: 72 pH before: 5.2 pH after: 7.0	Pb: 99.47	-	(Khayan *et al.*, 2019)
Siphon ceramic filter	Household and community roof runoff in Ghana	-	-	Total coliforms: 1.03 *E. coli*: 1.25	(Barnes, Collin and Ziff, 2009)
Bio-sand filter	Household and community roof runoff in Ghana	Turbidity: 96	-	Bacteria/viruses: 3 Protozoa: 4	(Barnes, Collin and Ziff, 2009)
Down flow gravel, sand and GAC filter	Building roof runoff close by the sea + highway in Brazil	pH before: 6.5 pH after: 6.8 Turbidity: 13.0 NO_3^-: 9.2 PO_4^{3-}: 37.5	-	-	(Teixeira and Ghisi, 2019)
Polyester membrane	Building roof runoff close by the sea + highway in Brazil	pH before: 6.5 pH after: 6.9 Turbidity: 8.7 NO_3^-: 12.6 PO_4^{3-}: 27.1	-	-	(Teixeira and Ghisi, 2019)
Recycled wood fibre filter	Office building roof runoff - Modified with Al_2O_3 for treatment first flush	TN: 22 TP: 32 Turbidity: 33	-	-	(Kim *et al.*, 2007)
Metal membranes (5, 1 and 0.5 μm)	Office building roof runoff	pH influent: 7.3 pH effluent 5, 1, 0.5 μm: 7.4, 7.4, 6.8 EC: -5.3 – 0.5 Turbidity: 17.7 – 53.8 COD: 55.7 – 67.2	-	-	(Kim *et al.*, 2007)

2.6 Cost analysis

2.6.1 Basic economic principles of investment analysis

The financial cost of implementing a RWH system is ultimately the most important argument in persuading enterprises or households to install a system. Investment analysis tools have been developed to determine if a system is economically feasible under certain conditions. These tools also make it possible to compare different scenarios. Investments are 'solidifying' cash into assets hoping that it will generate future profits. These profits can be seen as increased revenue (e.g. machines in factory) or in the form of less costs. In this case a reduction in water bills could be achieved by substituting tap water with harvested rainwater. The feasibility of investments is always compared on the basis of cash flows which are divided into cash-out (purchase and costs) and cash-in. From now on, cash-in will be considered as reduction in tap water bills in the case of rainwater harvesting rather than actual incoming cash flows. A short introduction into various financial investment approaches required for this thesis will be given in the next paragraph. A referral to specialised literature is given for more details and in-depth discussion of these methods.

Net present value (NPV) assesses the profitability of an investment project based on the time value of money (cost of opportunity). The goal is to determine the present value of future cash flows above the initial investment cost. Discount times and rates are needed when calculating todays value of future cash flows (Poelaert, 2019). Discounting is the process of converting costs and benefits at different points in time into a single time dimension using a discount rate which denotes the change in the money value per time period (Jawad and Ozbay, 2006). This principle acknowledges the fact that €1 today is worth more than €1 received in one year as the cash-in that is received today can generate more interest, if placed in the bank, than the cash-in received in 1 year (Poelaert, 2019). The discount time is thus the lifecycle of the project (in RWH systems this can go up to 50 or 60 years) and discount rates are the expected rates of return of the investment. The applied discount rate is dependent on the stakeholder. Private individuals choose higher rates and shorter periods while institutions tend to go for lower rates and longer discount periods especially if they have social responsibilities (e.g. government, local authorities, social institutions) (Roebuck, Oltean-Dumbrava and Tait, 2011). The determination of discount rates and times is highly significant and should be calculated by specialists (Jawad and Ozbay, 2006). Another factor to implement is the progressive trends of prices. The rate of the trend can be determined by giving a historic overview of prices and applying a linear extrapolation. This allows for a simple price prediction method commonly used for analyses of RWH systems (Farreny, Gabarrell and Rieradevall, 2011). Using the NPV method leaves a substantial room for error as this method relies heavily on assumptions and speculations (Gallo, 2014). A major disadvantage is that two projects with a different time span cannot be compared. Only positive NPVs are considered worthy investments (Ross, 1995).

Whole life costing (WLC) gathers all the costs of an investment over its lifespan into one value, the total cost of ownership (TCO). This is not only the purchase costs but also includes planning and design costs, operational costs, maintenance costs, decommissioning costs etc. WLC is made up of technical and financial input. Technical input is the installation, maintenance, materials, energy consumption etc. while the financial input consists of the discount rates, discount times and progressive trends of prices (e.g. price of tap water or electricity prices over time). The more accurate the financial input is, the more correct the WLC will be. A low WLC value is desired for an investment to be deemed financially feasible (Słyś and Zelenakova, 2012).

The payback time of an investment can be calculated as the ratio of the initial purchase and installation costs and the reduction in tap water bills. The investment has reached break-even point when payback time has passed. However the time value of incoming cash flows in neglected. This method is thus less sensitive for external factors (Poelaert, 2019). The shorter the payback time, the more interesting it is to make the investment.

Benefit cost ratio (BCR) are the total cash benefits divided by the total proposed costs of a project. Again, the cash benefits are calculated by discounting them over the lifespan of the project using an appropriate discount rate. Many assumptions and uncertainties are incorporated as is the case with NPV analysis. BCR is mostly used as a rough estimation on the viability of a project as a wide range of outcomes can be obtained. BCR should have a value greater than 1 to be considered a good investment (Shively, 2012).

2.6.2 Examples of financial studies concerning RWH systems

To date, some investment analysis studies have been conducted concerning the financial benefits of a RWH system, but no definitive conclusion of this topic has been reached as different authors use a different set of parameters (number of occupants, water consuming activities, household versus neighbourhood implementation, new versus retrofit construction etc.) and the heavy geographical dependence of the efficiency. Table 2-11 summarizes different studies conducted on this topic, gives some important parameters regarding the implemented RWH systems such as scale, time period, discounting factors etc. and also the outcome of the conducted investment analyses.

Costs were defined as crucial factors in investment analyses in the previous paragraph. RWH systems are not free and require capital, operation and maintenance costs. Capital costs include the purchase of every component of the system, going from the concrete tank to the pump and the valves, plus the installations costs. Decommissioning costs also are part of the capital costs. Buildings where an extra RWH system was installed, start with a larger financial disadvantage than properties with only a tap water supply. Operating expenses include the use of electricity for the pump and the use of filtration media or disinfection techniques (e.g. activated carbon, chlorine tablets, UV-irradiation etc.). In some studies, volumetric sewerage charges are also included, but in most cases these are neglected as the charges are the same for rainwater and mains water or these are incorporated into the drinking water bill. The costs of mains water should also be included if the system consists of a RWH system with a mains top-up function. Maintenance costs consists of every repair and replacement to keep the system up and running. Comparing this type of costs is hard as every literature reference uses different timetables and prices. Table 7-2 in the supplementary materials gives a summary of the maintenance costs that can be expected after a certain amount of time for a RWH system in the UK with taxes included. Table 7-3 gives a detailed overview of the price ranges for the components and labour when installing a RWH system. These prices are averages and differ in every country. Table 7-4 gives a complete list of the three types of costs (capital, operation and maintenance) for a RWH system in Barcelona, Spain. It also takes in account the decommissioning costs.

Table 2-11: Review of financial analyses of RWH systems

Location + average rainfall (mm/year)	**Scale**[a]	**Time**[b]	**Financial assessment approach**	**Discount rate (%)**	**Discount time (years)**	**Outcome of investment analysis**	**Reference**
West Yorkshire, UK (± 700)	S	N	WLC of 3840 different scenario's	3.5 5 10 15	5 10 25 50	Not feasible (**Fout! Verwijzingsbron niet gevonden.**)	Roebuck, Oltean-Dumbrava and Tait (2011)
Florianopolis, Brazil (1706)	M	N	Determination of payback period + NPV for combination of greywater and rainwater	1	15	Payback time: 94 -165 months NPV: € -636 - 25	Ghisi, Tavares and Rocha (2009)
Amsterdam (856)	NB, S	N	Determination of TCO/m³ for 6 scenarios	/	/	TCO/m³: €2.71 – 85.24	Hofman-Caris *et al.* (2019)
Greater Sydney, Australia (743 – 1376)	S	N	BCR for 3 tank sizes in 10 locations	3	40	BCR: 0.52 -0.90 BCR: 1.00 – 1.82 (with governmental tank rebate)	Rahman, Keane and Imteaz (2012)
Sydney, Australia (1200)	M	N	BCR for 36 scenarios with multi storey buildings	5 7.5 10	60	BCR: 0.26 – 0.68	Rahman (2010)
Granollers, Spain (665)	NB, M	R, N	Determination of NPV and payback period of 4 scenarios	0 3	60	Payback period: - NPV (0%): € -14076 – (-217 046) NPV (3%): € -9511 – (-324 201)	Farreny, Gabarrell and Rieradevall (2011)
Barcelona, Spain (640)	S, M	N	NPV and payback time for 8 different scenarios	1.75	50	Payback time: 3.31 – 26.27 years NPV: € -3735 – 983 227	(Vargas-Parra *et al.*, 2014)

[a] S = single household, M = multi storey building, NB = neighbourhood level

[b] N = new construction, R = retrofit action

Despite some problems concerning the non-uniformity of the investment analyses, some general conclusions can be drawn when comparing the data provided in Table 2-11.

Based on the available literature, an RWH-system is financially not viable for a single household but should be implemented on a neighbourhood level. This means however that less water is available per inhabitant, certainly when the neighbourhood consists of apartment buildings (Farreny, Gabarrell and Rieradevall, 2011). If the water should reach potable quality, expensive analyses further increase the costs to a point where it is no longer interesting for a single household to install a system as can be seen in the study by Hofman-Caris *et al.* (2019) where the TCO for a rural single household is upwards of €80 for a household RWH system and only €2 for a city district.

When comparing the outcomes of the studies summarized in Table 2-11 capital costs are almost never recovered by savings in water bills. Operational and maintenance costs however can be covered. This is shown in Figure 2-5 where the WLC of a RWH system starts from £2500 (€2900) where the WLC of a tap water installation is still £0. This price difference is caused by the capital costs of installing a system. Profit might be obtainable from using an RWH system if the capital costs would be carried by a third party (e.g. government, local authorities etc.). This was also the outcome in a study by Rahman, Keane and Imteaz (2012) where a scenario with and without a government rebate was calculated. Only the BRC of the projects with a rebate were greater than 1. The present rebate given by the Australian government however was not enough for the installations to reach a BRC of 1. Tank rebates had to be between €738 and €1117, this would mean an increase of the current rebates by 24-38%. Comparing the implementation costs of a RWH system is difficult as research spans over almost 20 years and most studies are conducted in countries that historically have a high level of water stress e.g. Australia, Italy and Spain. The closest resembling case study that was conducted to the author’s knowledge is the one from Hofman-Caris *et al.* (2019) in The Netherlands. As the climatological parameters are very much alike, costs of labour and components were similar and the research was published very recently, this study is a good starting point to verify whether or not an RWH system would be financially feasible in Flanders. The study concluded that, at the moment, rainwater harvesting used as an alternative for drinking water production is not economically feasible. The cost for a single, rural household would be too high (€60-110/m³) while RWH harvesting in a city district covers only about 50% of the demand.

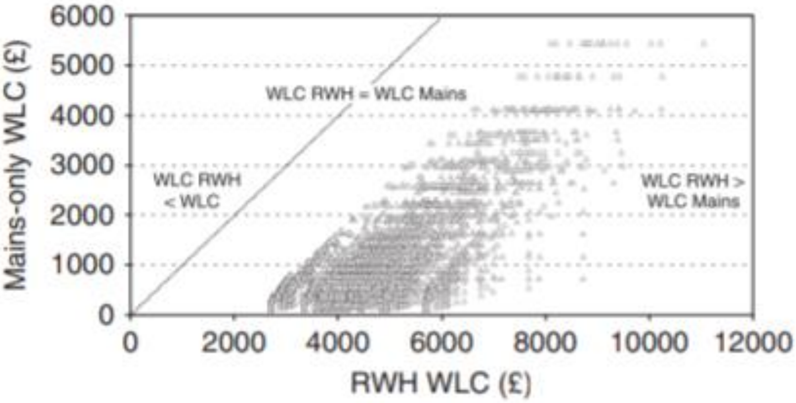


Figure 2-5: Representation of WLC of a RWH system in function of WLC of a tap water installation for 3840 scenarios (Roebuck, Oltean-Dumbrava and Tait, 2011)

All these studies investigated the short term costs and gains but failed to implement the long term, less tangible gains in the form of reduction in downstream water conveyance and treatment systems, alternative water infrastructure such as dams, desalination installations and storm flood drainage and flood prevention infrastructure (Roebuck, Oltean-Dumbrava and Tait, 2011; Vargas-Parra *et al.*, 2014; Hofman-Caris *et al.*, 2019). Furthermore, rainwater storage provides insurance against water scarcity and has a positive environmental impact on the surrounding waterways. The benefit cost ratio would improve drastically and the payback time would be reduced significantly if these benefits would be incorporated in the calculation. For example, in Berlin, a tax of €1.84 is levied per square metre of paved surface area. The tax is to compensate the costs of nuisance caused by storm flooding and water treatment. If the water is harvested however, this cost can be cut and can be added to savings. This is an example how government policy directly influences the economic feasibility of RWH-systems (Hofman-Caris *et al.*, 2019).

Another problem is the low costs of tap water production and supply that have a detrimental effect on the financial viability of a rainwater harvesting system. The study by Farreny, Gabarrell and Rieradevall (2011) added another scenario in their paper keeping in mind the progressive price trend of tap water. Here, the average water price would increase from 1.12 €/m³ in 2011 to 4 €/m³ in the coming years. This price was chosen as the average water price has risen over the past years at an annual rate of 5%. Given the location where the study was conducted, the city of Granollers in Spain, where there is a high level of water stress and an increasing demand of potable water, this price could be an underestimation. Only in this case would the implementation of an RWH system on household level be feasible. This would either be retrofitted (with a discount rate of 0% and a payback time of 31 years) or newly constructed with a discount rate of 0% and 3% which would give a payback period of 27 and 51 years respectively. At the current water price, none of the four scenarios would give a positive NPV as shown in Table 2-11. Rahman, Keane and Imteaz (2012) also investigated the effect of the water price in Sydney, Australia. A sensitivity analysis was conducted on a BRC simulation that showed that an increase of 39% (from €1.16 to €1.61/m³) would be necessary to get the BRC up to 1.

2.7 Conclusion of the literature study

Rainwater originating from water vapour in the air ends up as roof runoff after following a pathway of processes and reactions. Many sources of pollution, from wet and dry deposition to leaching to coincidental deposition, deteriorate the quality of pure rainwater. The quality of runoff can therefore vary a great deal as these processes are dependent on various factors (e.g. meteorological data, used construction materials, location etc.). Standard water quality parameters, which are already well studied and discussed in the literature, are regularly used to get an overview of the runoff quality. But information about micropollutants, both of metallic and organic nature, that can end up in rainwater is limited.

The legislative framework does not support the reuse of rainwater in Flanders at the moment. Only the quality of drinking water and the quality requirements for the laundry sector can be used as a comparative framework as these standards express clear quality parameter values that should be met. Other frameworks for the use of rainwater in other sectors such as the food industry only allow the water to be used in low-grade applications. Spain was cited as an example where progressive legislature was put in place for the reuse of reclaimed water. This clear and incentivising framework, if extended to a European level, may persuade households and the industrial sector from embracing rainwater harvesting as a potential alternative to tap water.

After assessing the numerous sources of pollution and the requirements set on a legislative level, it became clear that treatment of roof runoff was necessary. Rainwater treatment is extensively discussed in the literature but mainly focusses on the improvement of standard water quality parameters. Very few sources have focussed on the removal of micropollutants such as heavy metals, PAHs, biocides and phthalates from roof runoff. Not only the efficiency of different treatment systems was compared, costs were also deemed of the upmost importance especially in an industrial setting. The financial viability of an RWH-system can be calculated using various investment analysis tools. However, as rainwater harvesting is very specific to the location, use of the treated water, prices etc. no one-size-fits-all technique can be proposed and the viability of the project should be determined on a case-by-case basis. Only cost effective treatments were considered. Rapid sand filtration is a proven technique that withholds solid particles while activated carbon is well known for the removal of dissolved metals and organic pollutants. The choice for this treatment set-up is based on the research by Khayan *et al.* (2019) who determined that sand filtration followed by GAC filtration had a high removal efficiency for a metal (Pb), was robust and simple and cheap to operate. Disinfection techniques were also compared: UV-disinfection was chosen to be used during the long-term filtration tests based on the results by Naddeo, Scannapieco and Belgiorno (2013) (see Table 2-10) as the ease of use and effectiveness were the most important advantages.

3 Materials and methods

3.1 Preparation of glassware

Every piece of reusable glassware used for the determination of the standard parameters or taking samples for organic analysis was thoroughly cleaned and rinsed by the following procedure:

1) Double rinsing with distilled water
2) Rinsing with a 0.1% (v/v) NH_3-solution (Chemlab, 25%)
3) Rinsing with a 0.1% (v/v) HNO_3-solution (Chemlab, 65%)
4) Rinsing with acetone

In case of immediate use, to prevent the non-evaporated acetone from interfering with the organic compounds, the glassware was rinsed with double distilled water.

Every piece of glassware for the determination of metal concentrations was rinsed with demineralised water, a 10% HNO_3 solution and double distilled water.
Samples for microbiological tests were taken in sterile flasks of 1 l. These were autoclaved at 121°C for 15 minutes. These flasks were stored at 4°C and should be processed within 24 hours of sampling.

Samples taken for the determination of metal concentrations were stored in new polypropylene falcon tubes of 50 ml. The tubes could be stored at -18°C for multiple weeks. Samples for the determination of the calibration curve were also stored in falcon tubes of 50 ml and were kept at 4°C.

Samples for the determination of organic concentrations were stored in dark brown glass flasks of 0.5 l to minimize degradation of organics due to light. The flasks were stored at 4°C and should be processed as soon as possible to obtain the most accurate results.

3.2 Preparation of stock solutions

The runoff used for this experiment originated from a house, equipped with a clay tile roof with zinc gutters. The house was located in a residential area. The runoff was filtered before it entered the tank (15 000 l) by a lava rock filter (bed volume = 108 l). On a weekly base, approximately 300 l of roof runoff was collected from an outdoor tap. The runoff was pumped from the tank to the tap by a submersible pump and was filtered by a fibre filter with a pore size of 100 µm.

To be able to detect the effluent concentrations and to calculate removal percentages, the influent was spiked with metals, PAHs, phthalates and a herbicide. To spike the influent (fresh roof runoff), a multi-element standard solution of metals was made in distilled water. This multi-element standard solution contained Fe, Ni, Cu, Mn, Zn, Al, Pb, Sr and Cr. Stock solutions and multi-element standard (made up from the stock solutions) were stored in 0.5 l plastic jars at 4°C. Table 3-1 gives more detailed information about these solutions and the desired concentration levels in the influent based on the concentrations given in Table 2-2. The concentrations in the stock solutions were calculated based on the fact that 10 ml of multi-element standard should be added per 100 l of influent.

Table 3-1: Expected concentrations for each metal in the influent rainwater

Element (salt)	**Manufacturer and purity (%)**	**Concentration in stock solution (g/l)**	**Concentration in multi element standard solution (g/l)**	**Desired concentration in influent (µg/l)**
Fe ($FeCl_3.6H_2O$)	Chemlab, > 99	20	2	200
Ni ($NiCl_2.6H_2O$)	Sigma-Aldrich	2	0.2	20
Cu ($CuSO_4.5H_2O$)	Unknown	50	5	500
Mn ($MnSO_4.H_2O$)	Merck, > 99	2	0.2	20
Zn ($ZnCl_2$)	Sigma-Aldrich, > 98	150	15	1500
Al ($AlCl_3.6H_2O$)	UCB N.V.	20	2	200
Pb ($PbCl_2$)	Unknown	2	0.2	20
Sr ($SrCl_2.6H_2O$)	Merck	10	1	100
Cr ($K_2Cr_2O_7$)	Sigma-Aldrich, > 99.5	2	0.2	20

In addition, a multi-component standard of PAHs, phthalates and diuron was prepared in methanol from stock solutions of these compounds in methanol (VWR Chemicals, HiPerSolv Chromanorm) due to the low solubility of these compounds in water. A volume of 1 ml of multi component standard should be added per 100 l of influent. Stock solutions and multi-component standard were stored in dark brown flasks of 0.1 l at 4°C to prevent photodegradation of the organics. Table 3-2 gives more information about these solutions and the desired concentration levels of each organic compound in the influent based on the concentrations present in roof runoff given in Table 2-3. If 0.01 ml of standard was spiked per litre of influent, then the added COD because of the methanol would be 11.9 mg/l.

Table 3-2: Expected concentrations for each organic in the influent rainwater

Compound	**Manufacturer and GC area%**	**Concentration stock (g/l)**	**Concentration multi-component standard (mg/l)**	**Desired concentration in influent (ng/l rainwater)**
Diuron	Sigma-Aldrich, > 98	20	1000	10 000
Diisodecyl phthalate	Sigma-Aldrich, > 99	20	1000	10 000
Bis(2-ethylhexyl) phthalate	Sigma-Aldrich, > 99.5	0.2	10	100
Phenanthrene	Merck	0.2	10	100
Naphthalene	VWR Chemicals, > 99	0.2	10	100

Fluoranthene	Sigma-Aldrich, 98	0.2	10	100
Pyrene	Merck, > 96	0.2	10	100
Fluorene	Merck, > 95	0.2	10	100

Table 3-3 summarizes some parameter values (water solubility, log K_{ow} and molecular weight) of the organic compounds which will be used later to explain the obtained results.

Table 3-3: Water solubility at 25°C, log K_{ow}-values and molecular weights for the organic compounds of interest in this research (European bureau of chemicals, 2003; WHO, 2003; Mackay et al., 2006; Krieger, Doull and Vega, 2010)

Group	**Compound**	**Water solubility at 25°C (µg/l)**	**Log K_{ow}**	**Molecular weight (g/mol)**
PAHs	Naphthalene	31 000	3.37	128
	Fluorene	1900	4.18	166
	Phenanthrene	1100	4.57	178
	Fluoranthene	260	5.22	202
	Pyrene	132	5.18	202
Herbicide	Diuron	42 000	2.68	233
Phthalates	DEHP	25-50	4.88	391
	DIDP	0.13-1000	8.80	447

The influent, consisting of spiked roof runoff, was stored in two 300 l polypropylene containers to ensure a constant filtration, taking into account weekends and holidays. The exact volume of roof runoff transferred to these containers was measured so accurate spiking could be performed. The containers were placed besides the set-up and were kept at room temperatures (± 18°C) in the lab.

3.3 Column tests

3.3.1 Experimental set-up long term tests

To determine the long-term effects of the filtration of rainwater, a system consisting of a sand filter followed by an activated carbon filter and an optional UV disinfection was installed. To get a complete overview of the rainwater quality, following parameters were measured: pH, EC, turbidity, COD, TN, TP, UV-VIS, metal concentrations, organic concentrations, total coliforms (37°C) and total viable count (22°C). This was performed on both the influent and the effluent of the two filters. This way, a clear overview of the changes in removal efficiency for each filtration technique could be obtained. Coliforms were used as indicator organisms for the presence of pathogens in the rainwater and the efficiency of the applied disinfection technique, UV-irradiation.

Furthermore, the set-up was duplicated: one was used for the continuous filtration of rainwater (24 hours per day, 7 days per week) while the other one was operated to simulate the average water consumption of a family (2 parents, 1 child). This was done based on the research by Santos *et al.* (2020) who collected this data in both summer and winter time. The pump was operated for 6 hours every weekday and for 9 hours every day of the weekend.

3.3.2 Column build-up

Each set-up consisted of two glass columns with an internal diameter of 23 mm. The top and bottom were closed off with perforated, elastomer stoppers. To ensure easy connections with the tubes, plastic connectors were placed in the stoppers. The stoppers were taped thoroughly with duct tape to withstand the pressure build-up of the water. To support the filtration medium and to prevent it from washing out, a layer of approximately 10 cm gravel was inserted. Afterwards, sand (particle size 0.4-0.8 mm, density 2.50 g/cm³) and GAC (particle size 0.60-2.36 mm, density 1.16 g/cm³, organosorb 10-AA from Desotec) were prewashed with distilled water until a clear supernatant was obtained. This suspension was added onto the gravel until the desired bed height was reached (30 cm for both sand and GAC). The columns were then loaded with at least 10 bed volumes of distilled water to clean them and to flush out the small particles that would increase turbidity. To make sure that the filter medium would always be submerged, a siphon with a de-aerator was installed so that there was always around 70 cm of water on top. Figure 3-1 gives a schematic overview of the set-up. Each column was numbered with a roman numeral: the sand filter and the GAC of the continuous set-up were I & II respectively, the same was done for the identical discontinuous set-up (III for sand filter and IV for GAC). The supply of rainwater was adjusted using peristaltic pumps (two in total) that were kept at a constant flow rate. Because the pump that supplied the water for the continuous column had a parallel pump functionality, the parallel one was used to pump the water from the second storage tank to the first one. This way, there would always be enough influent and once the second container was fully transferred to the first container, new runoff was added in storage container 2 and spiked.

The applied flow rate was determined by using the linear filtration rate and the cross-sectional area of the column. Referring back to the typical filtration rates used in rapid sand filtration in (see 2.4.2) and given the diameter of the column, 23 mm, the filtration volumetric rate was set at 40 ml/min which is equal to 5.8 m/h.

With this information, empty bed volumes can be calculated. For example, after 1 hour, a volume of 60 min * 40 ml/min = 2400 ml had passed through the filter. Using the diameter of the column and the height of the filtration bed, the volume of filtration medium can be calculated (e.g. column I = 117.8 ml). Then the amount of bed volumes is (2400 ml/h)/(117.8 ml/BV) = 20 BV/h.

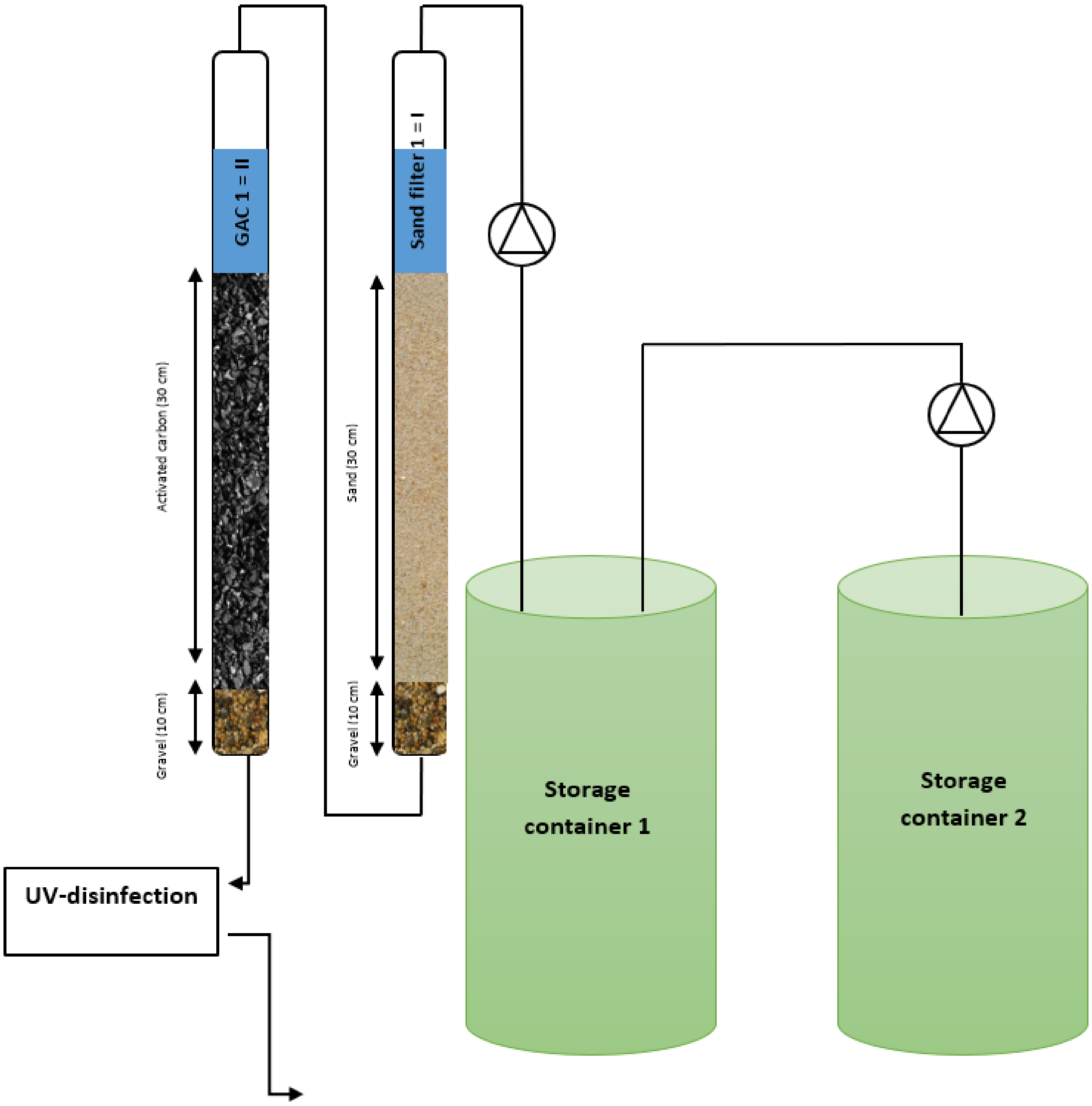


Figure 3-1: Schematic overview of the filtration set-up for the long-term column tests

3.3.3 Determination of the stability of spiked components

As discussed in 3.2, the metals were not acidified when the multi-element stock solution was prepared. This means that the ions can adhere to the container wall which decreases the available concentration of the metal in the influent (Asakawa, 2021). Organic compounds on the other hand can easily degrade to different, non-target compounds especially when micro-organisms are present in the water (Banerjee *et al.*, 2021).

The influence of both phenomena was investigated by spiking 30 l of roof runoff in a polypropylene storage tank with the desired concentration of metals and organics (as described in Table 3-1 and Table 3-2) and taking four samples over time. This way, a possible decrease in concentration over time could be accounted for and explained.

3.4 Laundry case study

3.4.1 Description of the situation

A case study was selected in collaboration with an industrial laundry company aiming at the optimization of their existing RWH system. The goal was to partially substitute the washing water, which is tap water, with treated rainwater that met certain requirements. The collected rainwater should meet certain quality requirements, but drinking water quality is not required (i.e. water fit-for-use). A reduction in costs and the implementation of a buffer against the dry summer months due to climate change was the aim. The requirements of the rainwater that should be met before using in the washing process are shown in Table 3-4.

Table 3-4: Quality requirements for process water used in the laundry industry

Parameter	Requirements for laundry industry process water
Conductivity (µS/cm)	< 1800 but preferably as low as possible
pH	Between 6.5 and 8.5
Hardness (mg/l CaO)	0 - 40
Metals (mg/l)	Fe < 0.1 Cu < 0.05 Mn < 0.05
Total viable count	< 100 CFU/ml
Total coliforms	0 CFU/ml
Other requirements	Clear water with no suspended particles No odour

The current RWH system available to the company consists of a buffer made from a flexible membrane (capacity 10 000 l), a rapid sand filter and chlorinator. There is also a pre-treatment train that consists of a fine screen filtration (mesh size 1 mm) and three sedimentation tanks (polymer containers with a capacity of 1 m³ each). A first flush technique was applied by discharging the first 20 l into the nearby river. The whole process is shown in Figure 3-2. Because this system counters the recurring droughts that have occurred in Belgium for the last five years, subsidies were granted for the installation and optimization of this project as part of the Blue Deal of the Flemish Government. This aims to combat droughts, water scarcity and strives for sustainable water consumption (VMM, 2021b).

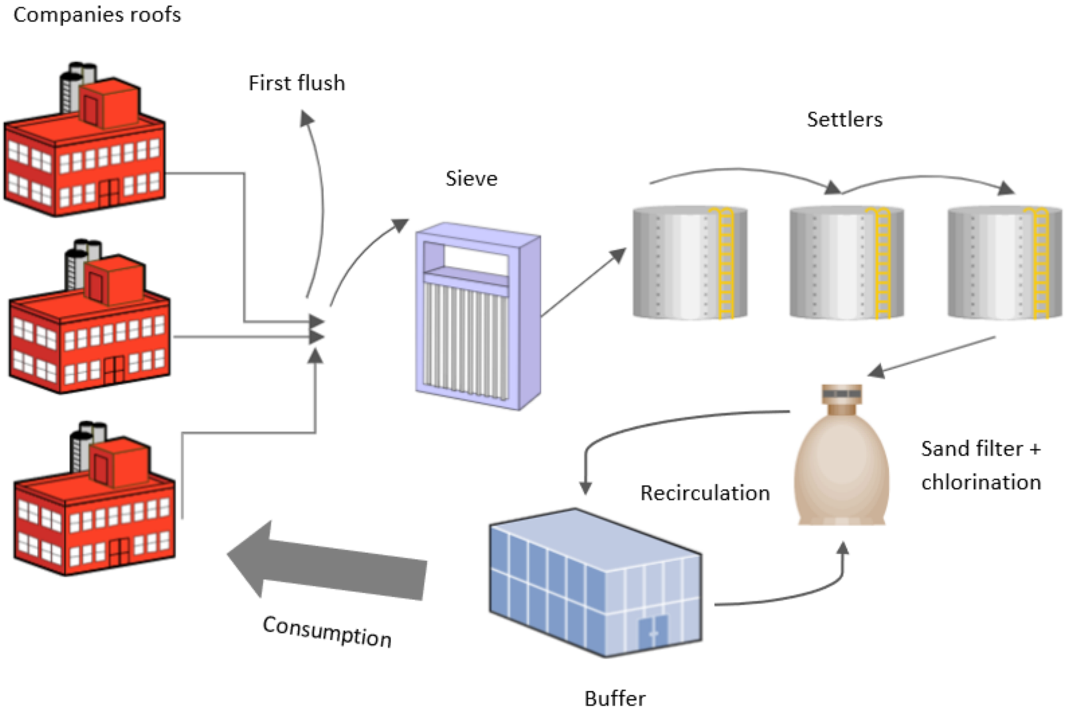


Figure 3-2: Schematic overview of the treatment train provided by the laundry company

The collection capacity is going to expand in the near future to around 6 buffers (60 000 l in total) and to harvest rainwater from three different roofs. The roofs would consist of the company itself and the two neighbouring buildings. An estimation was made for the collection surfaces based on aerial images via AGIV generic viewer (Vlaanderen, 2021). Table 3-5 gives the surface per roof and their percentual share in the total catchment surface. The average yearly precipitation in 2020 for the area was consulted from a local weather station and was 655.7 mm (Weerstation Denderstreek, 2020). So a volume of around 5649 m³ can be expected on a yearly base.

Table 3-5: Surfaces of the catchment areas and their percentual share

Roof location	**Surface (m²)**	**%**
Roof A: laundry company	2190	26
Roof B: commercial building	2790	33
Roof C: industrial building	3635	42
∑	8615	100

3.4.2 Experimental set-up laundry case

A treatment train consisting of a lava rock filter, a sand filter, a GAC column and several disinfection options was proposed in order to achieve a water quality fit-for-use in the laundry industry. An optimal balance between costs, ease of use and removal efficiencies was obtained this way. To better estimate whether each component of the train is necessary, tests were performed on each filter unit separately. Because strict requirements for the microbiological parameters were imposed, a robust disinfection process was coupled at the end of the physicochemical treatment train. The different strategies for disinfection that were tested, were ozonation, hydrogen peroxide, chlorination and UV-irradiation.

Before the filtration tests, each influent stream from the different roofs was examined separately for the imposed set of quality parameters. The filtration tests however were performed with a mix of the three different runoffs as a simulation of what would happen during real application in the near future. A container with a volume of 70 l was filled according to the collection surface area distribution given in Table 3-5.

The columns that were used for sand filtration and GAC were identical to the ones used in the column test (see 3.3.2 Column build-up). For the lava rock filter (particle size 3-5 cm), a plexiglass column with an internal diameter of 125 mm was used. The bed height of the rocks was 60 cm. All filtration media (activated carbon, sand and lava rocks) were washed with demineralised water until a clear supernatant was obtained. De-aerators were attached on each column so that the glass columns had a hydraulic head of around 70 cm of water and the plexiglass column around 30 cm of water. A peristaltic pump was used to feed the lava rock filter with a flowrate of 600 ml/min (2.9 m/h). The effluent was then collected and pumped to the top of the sand filter using a peristaltic pump with a flowrate of 40 ml/min. The effluent of the sand filter was gravitationally pumped to the GAC column. Figure 3-3 gives a complete overview of the filtration set-up.

Samples were taken at different locations: the effluents of the lava rock, sand filter, sand filter + GAC and lava rock + sand filter + GAC. The analyses that were performed on each effluent were: pH, EC, turbidity, UV-VIS, COD, TN and concentrations of soluble metals. This was done according to the methods described in 3.6.4 Microbiological analyses comprised of the determination of total viable count (22°C) and total coliforms (37°C). Therefore, samples were taken in sterile recipients and disinfected in accordance with 3.6.4.

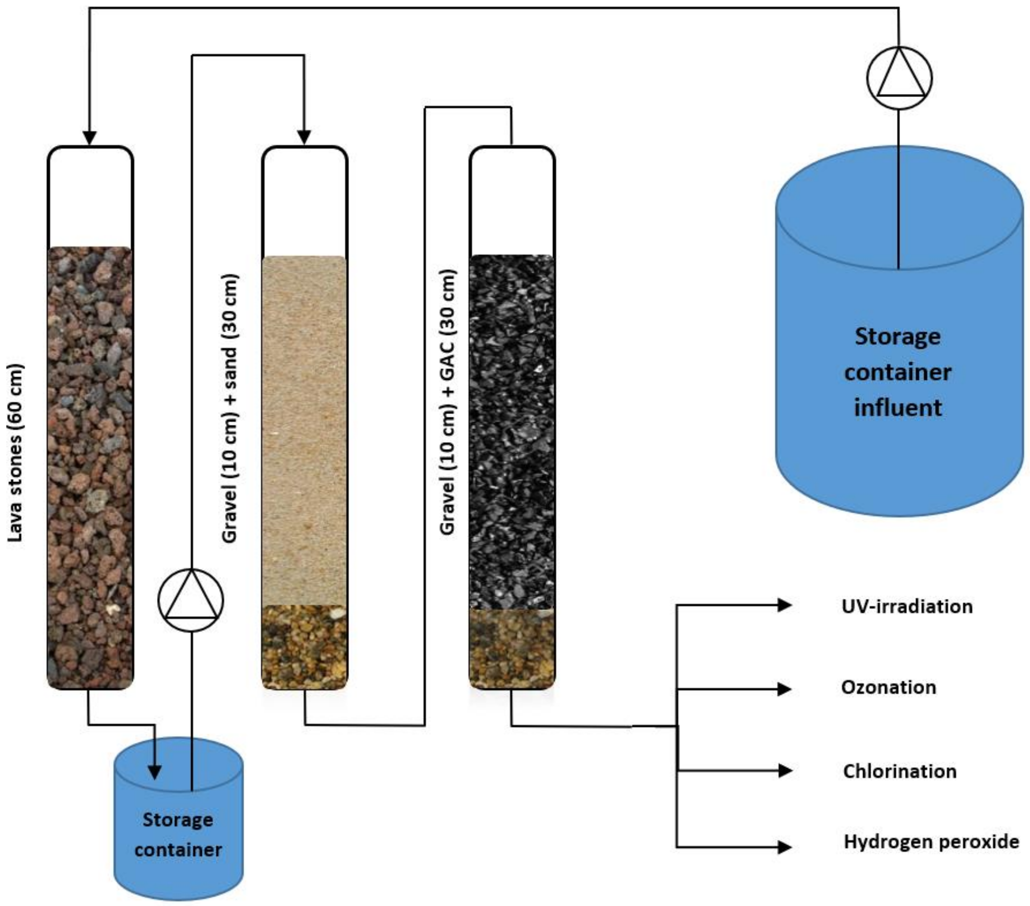


Figure 3-3: Schematic representation of the filtration set-up of the laundry case (each column was tested separately and finally all together in series)

3.5 Disinfection strategies

When using ozonation, chlorination or hydrogen peroxide for disinfection, 500 ml of effluent from the laundry case study was taken in a sterile recipient and disinfected with the appropriate dose. Effluents (both laundry case and long term filtration tests) were disinfected with UV-irradiation by pumping the sample through the UV-reactor and 500 ml of the disinfected stream was collected in a sterile recipient.

3.5.1 Ozone

An ozone stock solution was prepared according to the method described in 7.3 (Supplementary material). It was made by bubbling ozone for 20 minutes through a cooled wash-bottle filled with double distilled water using sintered glass. High purity oxygen gas (AirLiquide) was transformed into ozone using an ozone generator (Ozomat COM-AD-02, Anseros) (Chys, 2017). A saturated ozone solution was obtained this way and could be used to disinfect the effluents. The concentration of the stock solution was determined and the immediate ozone demand (IOD) of the rainwater was determined alongside the disinfection, following the protocol in 7.3, 7.4 and 7.5 in the supplementary materials. The determination of the IOD is necessary as ozone will react quick with any oxidizable compound present in the rainwater. Thus a part of the oxidizing potential is lost before it can be applied as disinfectant (Xu *et al.*, 2002). When the concentration of the ozone stock solution was known, a concentration of 5 mg/l is added to 500 ml sample taken in a sterile recipient for microbiological analysis. A contact time of 10 minutes is recommended by Tchobanoglous *et al.* (2014).

A screening test for the disinfections agents was performed before the filtration tests of the laundry case were carried out to determine the right concentrations of each medium and its effectiveness on the removal of micro-organisms. A concentration of 2 mg/l ozone was tested along with other disinfectants as this is a value found in the literature (see 2.4.3.2). However, after reviewing the results from the tests, a concentration of 5 mg/l was used due to its increased effectiveness.

The IOD for the influent of the laundry case was also determined according to 7.5. The immediate ozone demand of the water was 0.34 mg/l.

3.5.2 UV irradiation

A UV lamp (Oase Vitronic 18W UV-C) was used to provide the radiation to disinfect the rainwater. The internal volume of the lamp was 1.5 l and the flow rate was 35 ml/min. This gives a residence time of 43 minutes, which is high. The lamp was turned on 15 minutes before sampling to ensure a disinfected effluent. A 5 mg/l chlorine solution was pumped through the reactor interior before pumping the sample through to disinfect the effluent tubing and the lamp sleeve. The interior of the reactor was afterwards flushed with demineralised water. A 500 ml sample was taken for microbiological analyses in a sterile recipient.

3.5.3 Chlorine

3.5.3.1 Determination of chlorine concentration

A commercial bleach solution was used to disinfect the water. Because the commercial bleach solution (4.05%) is subject to an auto redox reaction, a redox titration is performed to determine the exact concentration of hypochlorite present in the bleach. The method is described in 7.6 in the supplementary materials and based on the method described by Shearer (2020).

3.5.3.2 Determination of the required chlorine dose

The determination of the residual concentration of disinfectant can be calculated using the Chick-Watson equation. Aiming for a 4-log reduction in micro-organisms, applying a contact time of 10 minutes (as recommended by Tchobanoglous *et al.* (2014)) and knowing the specific lethality coefficient for total coliforms using free chlorine (2 l/mg.min), the residual concentration should be 0.2 mg/l according to Equation 3-1.

$$C = \frac{\log\frac{N}{N_0}}{\Lambda * t} = \frac{4}{2\frac{l}{mg.min} * 10\ min} = 0.2\ mg/l$$ *Equation 3-1*

N = amount of organisms at time t
N_0 = initial amount of organisms
Λ = coefficient of specific lethality (l/mg.min)
C = residual concentration of disinfectant (mg/l)
t = contact time (minutes)

As discussed in 2.4.3.1, the immediate chlorine demand of rainwater is typically 0.5 mg/l. Adding the residual concentration and immediate chlorine demand together leads to a typical dosage of around 1 or 2 mg/l. To disinfect with 2 mg/l , a volume of 50 µl of a 40.5 g/l solution was added to 500 ml sample, taken in a sterile recipient for microbiological analysis.

Concentrations of 1 and 2 mg/l chlorine were tested in de screening for the laundry case based on values found in the literature (see 2.4.3.1). The decision was made based on the obtained results to use a concentration of 2 mg/l in the filtration tests of the laundry case.

3.5.4 Hydrogen peroxide

A 30 w/v% H_2O_2-solution (Chemlab Analytical, phosphate stabilised) was used to disinfect 500 ml of rainwater that was collected in a sterile recipient. Keeping in mind the proposed concentration levels discussed in 2.4.3.5, 90 mg/l was added.

Three concentrations were tested in the screening test of the laundry case as sources in the literature on this subject are scarce (see 2.4.3.5): 30, 60 and 90 mg/l H_2O_2. From these three concentrations, 90 mg/l had the highest effectiveness on microbial removal for the laundry case.

3.6 Chemical and microbiological analyses

3.6.1 Analysis of standard parameters

The method and used devices for the determination of the standard parameters are shown in Table 3-6.

Table 3-6: Measurement method of the standard parameters

Quantity	**Unit**	**Appliance**	**Method**
pH	-	Hach HQ 30d flexi portable meter	Electrometric: ISO 10523
Electrical conductivity (EC)	µS/cm	Hach HQ 30d flexi portable meter	Electrometric: ISO 7888
Turbidity	NTU	Hanna HI 98703 Turbimeter	Nephelometric: ISO 7027
UV-VIS	-	UV-1280 Shimadzu (range 190-1100 nm, 0.5 nm interval)	ASTM D2008
Chemical oxygen demand (COD)	mg O_2/l	Hach test kits LCK 1414 (range 15 – 150 and 5-60 mg O_2/l)	(Laurent, 2014)
Total nitrogen (TN)	mg/l N	Hach test kits LCK 138 (range 1-16 mg/l N)	ASTM D8083-16
Total phosphate (TP)	mg/l PO_4	Hach test kits LCK 349 (range 0.15-4.5 mg/l PO_4)	ASTM D8001-16
Chlorine free	mg/l Cl_2	Hach test kit LCK 410 (range 0.05 -2.00 mg/l Cl_2)	ISO 7393-2

3.6.2 ICP analysis – determination of dissolved heavy metals

The procedure is based on US EPA method 6010D (US EPA, 2014). Samples for ICP analysis were filtered with a 0.45 µm filter (Chromafil® Xtra RC-45/25 from Macherey-Nagel), acidified to 1% with HNO_3 (Chemlab, 65%) and stored in a plastic falcon tube. They were stored at -18°C, the calibration standards were stored at 4°C. The calibration standards were prepared from a stock solution of 1000 mg/l (Certipur® ICP multi-element standard solution IV) and had a range from 1 to 1000 µg/l.

The concentrations of dissolved metals were measured using an ICP-OES (iCAP 7200 Series from Thermo Scientific) with autosampler (CETAC ASX-560). Due to the low concentrations, the axial mode was used. The limit of detection (LOD) was calculated by determining the standard deviation on the lowest standard for six repeated measurements. The LOD was set to be equal to three times this standard deviation. The limit of quantification (LOQ) was set at 6 times the standard deviation (Virgilio *et al.*, 2020). Table 3-7 gives the used wavelength, the LOD and the LOQ for each metal measured.

Table 3-7: Wavelengths, LOD and LOQ of ICP-OES

Element	**Wavelength (nm)**	**LOD (µg/l)**	**LOQ (µg/l)**
Al	309.271	5.4	10.8
Cr	267.716	1.5	3.1
Cu	324.754	0.8	1.5
Fe	259.94	1.1	2.1

Mn	257.61	0.6	1.2
Ni	231.604	0.7	1.5
Pb	220.353	4.0	8.1
Sr	407.771	0.4	0.7
Zn	202.548	0.4	0.8

3.6.3 GC-MS analysis – determination of organic compounds

To be able to determine the organic concentrations with gas chromatography (GC), a transfer from the aqueous phase to an organic volatile solvent is necessary. Solid phase extraction (SPE) is designed to retain compounds of interest on a solid packing and transferring them to a solvent by elution. A rapid volatilization occurs when injecting the sample in the GC-injection port with a temperature around 280°C. The compounds are then separated in the GC-column. Detection was performed by a mass spectrometer (MS) which can identify compounds on their mass/charge ratio. The analysis by GC-MS preceded by SPE is based on the method described by ISO 21253-1:2019 method (ISO, 2019).

3.6.3.1 SPE-extraction

Due to the presence of biofilm and other particles in the samples, a filtration with glass fibre filters (Whatman, particle retention 1 µm) was performed using a vacuum pump. Exactly 400 ml of the filtered sample was transferred to an Erlenmeyer flask. A volume of 100 µl of a surrogate standard (consisting of deuterated acenaphthene, phenanthrene and chrysene in methanol) was added to the flask and thoroughly shaken.

The SPE-cartridges (octadecyl-modified silica gel (endcapped), 500 mg/6 ml, Macherey-Nagel) were conditioned with respectively 4 ml of dichloromethane (DCM, Carl Roth, > 99.9% purity, GC ultra-grade), 4 ml of methanol (VWR Chemicals, HiPerSolv Chromanorm) and two cartridge volumes of double distilled water. It is important to note that the cartridge should never be dry when applying the vacuum. Next, the sample was loaded onto the cartridge. The vacuum was adjusted so that one drop of sample per second was loaded onto the cartridge. After the whole sample had passed through, the cartridge was dried by leaving the vacuum on for 20 minutes. A volume of 4 ml DCM was then used for the elution and the eluate was stored in a dark glass flask with ± 1 g of anhydrous sodium sulphate (Riedel-de Haën AG) to trap the remaining water in the salt. To be able to inject the sample in the GC-MS, 1 ml of the eluate was pipetted into a dark vial and sealed with a PTFE cap.

3.6.3.2 GC-MS analysis

The vials were then placed in the autosampler (Agilent 7683 Series) and injected in splitless mode into the GC (Agilent 6890 Series, Agilent 19091S-433 capillary column). The oven program is described in Table 3-8. Detection was performed by a mass spectrometer (MS) (HP MS 5973) in SIM mode. Chromatograms were integrated using Chemstation® software (Agilent).

Table 3-8: Temperature program for GC oven

Ramp	**Rate (°C/min)**	**Target temperature (°C)**	**Runtime (min)**
Initial	/	45	2
1	12	250	20.08
2	5	310	32.08
3	12	315	34.50

Table 3-9 shows the two qualifier ions, the LOD and LOQ for each organic compound measured with the GC-MS. The LOD was calculated by determining the signal/noise ratio for each component on the lowest standard. The concentration corresponding with a signal/noise ratio of three was used as the LOD. The concentration corresponding to a signal to noise ratio of 10 was used as LOQ (AOAC, 2011).

Table 3-9: Overview of target and qualifier ions for each compound as well as the LOD and LOQ

Compound	**Target ion (m/z)**	**Qualifier ion 1 (m/z)**	**Qualifier ion 2 (m/z)**	**LOD (ng/l)**	**LOQ (ng/l)**
Naphthalene	128	127	129	15	50
Diuron	72	232	234	830	2767
Fluorene	166	165	82	9	31
Phenanthrene	178	176	179	13	43
Fluoranthene	202	200	203	3	11
Pyrene	202	200	203	4	13
DIDP	149	307	167	15 784	52 615
DEHP	149	279	167	16	54

3.6.4 Microbiological analysis

The method used for *E. coli* and total coliform analysis is based on the ISO 9308-1:2014 standard (ISO, 2014). *E. coli* and total coliforms were determined by filtration and plating using RAPID'*E.coli* 2 Agar (Biorad) as medium. The plates were incubated at 37°C for 24 hours.

Total viable count was based on the ISO 6222:1999 standard (ISO, 2021). A volume of 1 ml was plated on plate count agar (CMO325 (ISO), Oxoid). The plates were incubated at 22°C for 72 hours.

4 Results and discussion

4.1 Stability of spike solutions

As discussed in 3.3.3, tests were carried to determine interactions between the environment and compounds. Both organic compounds and metals were spiked in runoff.

4.1.1 Soluble metals

Figure 4-1 and Figure 4-2 show the changes in concentrations over time for the metals. Samples were taken at the start and after 1, 16 and 72 hours. This period of time was considered as a batch of spiked influent used for the long term filtration tests typically lasted 3 days. For some elements, such as Pb, Al, and Fe, a strong decrease over time was observed. This can be caused by adsorption onto the walls of the container or adherence onto particles present in the influent which will be removed from the influent by sedimentation (Kaushal, 2017). For other elements, such as Ni, Mn, Cr and Zn, the concentrations in the spiked runoff were found to increase over time. This can potentially be explained by leaching of these metals from the storage container. Metals can be used in polymers as pigments, biocides, flame retardants, fillers etc. These compounds are added for their specific functionality and their concentrations can amount to several percent on weight basis (Turner and Filella, 2021). The leaching of zinc, as shown in Figure 4-2, is also mentioned by Stephen *et al.* (2014) who showed that the leaching process was pH dependant with a lower pH leading to higher leaching rates. Nickel and manganese showed a significant increase over time while chromium only exhibited a small peak at the start. For Cu and Sr, only a small variation over time was observed. The theoretical spiked concentrations and the measured concentrations of each metal are presented in Table 4-1. From this data, it becomes clear that measuring, dilution and spiking errors cannot be ruled out from having an impact on the results of these tests. For example, lead was only measured in a concentration which was half of what the intended concentration should be after spiking. This is probably due to the low solubility of $PbCl_2$. These results indicate that the removal percentages calculated for the long-term filtration tests are subject to a number of side-effects which may influence the accuracy of the removal results. Measured concentrations were used for the calculation of the removal percentages for the long term filtration tests as insufficient data is provided in these tests to determine a structural behaviour of leaching or adsorption by these metal compounds.

Table 4-1: Theoretical and measured concentrations of each metal at the start of the experiment

Metal	**Spiked concentration (µg/l)**	**Concentration at start (µg/l)**
Ni	20	19.6
Mn	20	26.2
Pb	20	10.3

Cr	20	19.6
Fe	200	191.3
Cu	500	446
Zn	1500	1848.8
Al	200	222.6
Sr	100	172.7

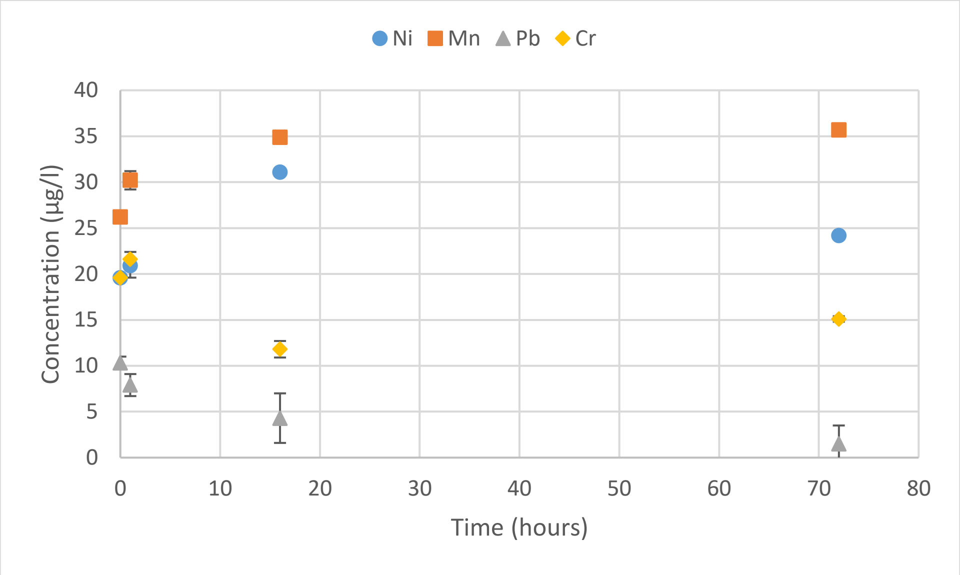


Figure 4-1: Changes in concentrations in the storage container over time due to adsorption/leaching for the lower concentration range of metals

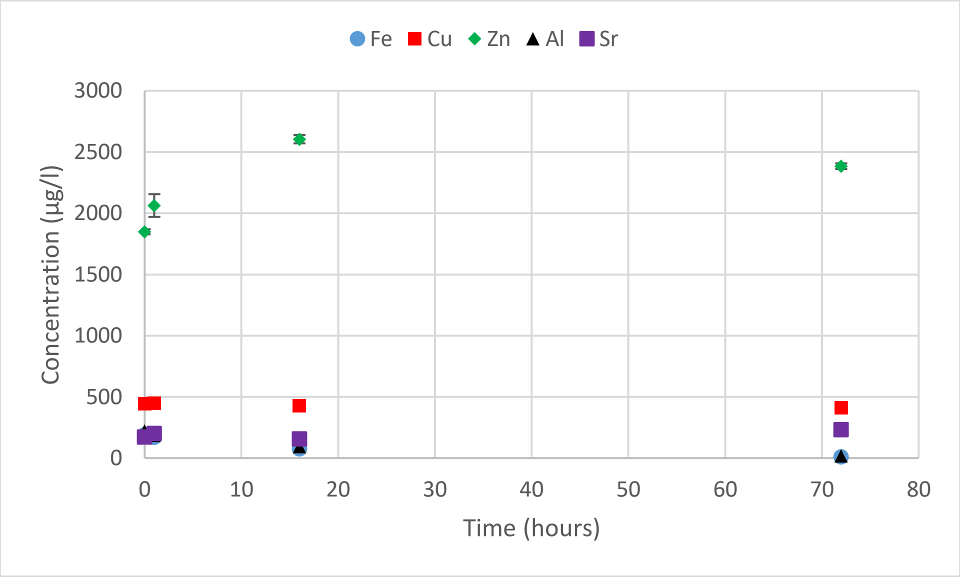


Figure 4-2: Changes in concentrations in the storage container over time due to adsorption/leaching for the higher concentration range of metals

4.1.2 Organic compounds

Leaching of organic compounds from plastic containers can also occur. When looking at Figure 4-3 and Figure 4-4, it becomes clear that plasticizers such as DEHP or DIDP and diuron, which functions as a herbicide, show an increase in concentration over time followed by a decrease while the concentrations of the PAHs remain quite stable throughout the testing period. The theoretical spiked concentrations and the measured concentrations of each compound are presented in Table 4-2.

Table 4-2: Theoretical and measured concentrations of each organic compound at the start of the experiment (: < LOD)*

Compound	**Spiked concentration (ng/l)**	**Concentration at start (ng/l)**
Diuron	10 000	12 056
Naphthalene	100	111
Fluorene	100	121
Phenanthrene	100	103
Fluoranthene	100	119
Pyrene	100	124
DIDP	10 000	14 391*
DEHP	100	133

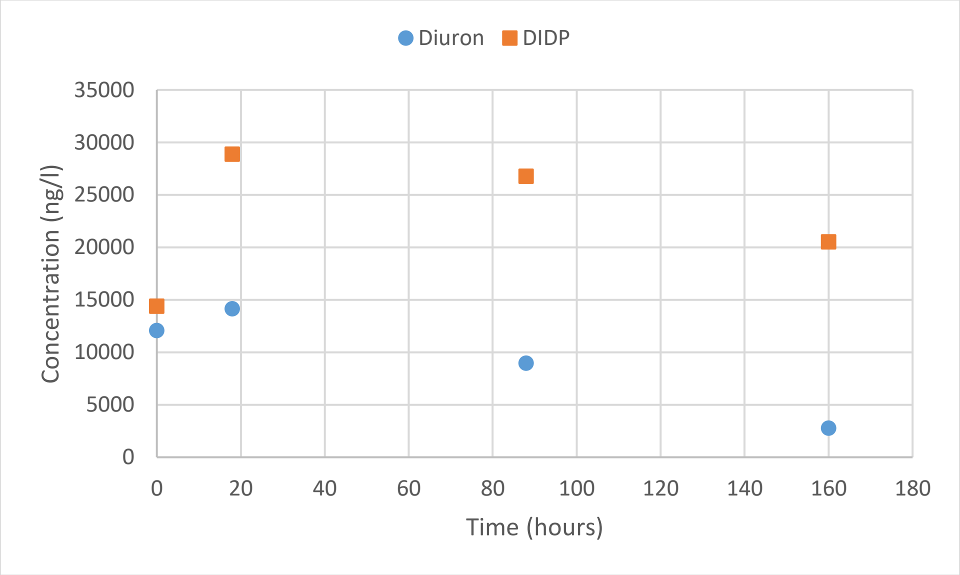


Figure 4-3: Changes in concentrations of organic compounds over time due to side-effects for the higher concentration range

Possible explanations for the increase in concentrations can be leaching of plasticizers present in the polypropylene storage tanks however, the very low solubility of DEHP and DIDP in water make this scenario unlikely. The decrease in concentrations can be explained by the degradation of the organic compounds by micro-organisms or by adherence onto the walls. Various sources in the literature have discussed this for diuron (Giacomazzi and Cochet, 2004), phthalates (Boll *et al.*, 2020) and PAHs (Baboshin and Golovleva, 2012). Degradation by sunlight was unlikely as the container was closed off during the testing period. Not only side-effects such as microbial activity or adsorption are a factor in these tests, also sufficient mixing at the start of the experiment can influence the results.

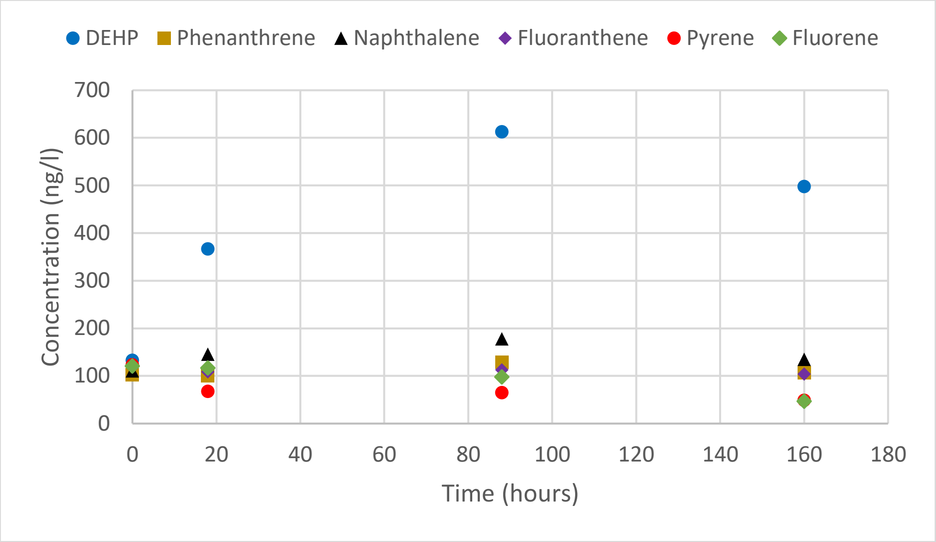


Figure 4-4: Changes in concentrations of organic compounds over time due to side-effects for the lower concentration range

4.2 Column optimisation

During the long-term rainwater filtration test, some problems arose that were mainly originating from the presence of micro-organisms in the water. One of the key reasons for the development of a biofilm is that the storage containers were kept at room temperature (around 18°C) whereas rainwater is normally stored outside and underground subject to the temperature of the soil (around 10°C) which slows microbiological growth drastically. Another reason is the fact that the tubing and columns were transparent so additional phototrophic growth could be possible.

The presence of biofilm caused clogging of the sand filter during the continuous filtration. This pressure build-up resulted in an increase of the water level inside the column and ultimately, the filter would overflow. The sand filter of the discontinuous unit had the same problem but to lesser extent: overflow of the column never occurred. This issue resulted in a number of optimizations that were applied:

1) Regular backwashing of the sand filter was the first step. At first, a backflush with only water was applied (bed volume 1-4000). Later on, a more aggressive cleaning method consisting of a combined air and water backflush was used (bed volume 4000 – 18 000). This helped control the pressure build-up in the column whereby the filter could operate for a longer time. However, it was ineffective in fully unclogging the sand filter.

2) Installing an automatic sensor based on conductivity was the second attempt at resolving the issue. Based on data acquisition software (Arduino), the peristaltic pump was linked with a data acquisition board which acted as a transmitter as well as two stripped wires placed in the top cork which functioned as conductivity sensor (see Figure 4-5). If the water in the column rose to the level of the wires, a current would flow between the wires resulting

in a signal from the micro controller which then switched off the pump. This system was successful at first but as the biofilm kept on growing, the pressure build-up would be faster and still pushed the stopper out of the column.

3) To break up the clog of sand particles and biofilm, a magnetic stirrer bar was inserted in the column which was manually operated by using a magnet on the outside. This way the column did not need to be opened every time to manually break up the clogging with a stirring bar. In combination with backflushing this system gave fairly good results in preventing overflow of the column.

4) The next step was to implement an outflow conductivity meter which was placed after the GAC column and a bit lower than the columns aerator. This one worked with a reverse mechanism of the inflow sensor. The pump was switched off when the internal volume of this conductivity sensor (see Figure 4-5) was empty. This could only happen if the sand filter was overflowing and no water was led to the GAC filter. No current was then measured. The two conductivity meters combined solved the problem for the most part.

5) A shock chlorination with a bleach solution was carried out to remove biofilm from the whole installation (containers, tubing, connectors and columns). Adding 5 mg/l of chlorine removed all the biofilm from the filtration unit. This was performed after 13 000 and 6000 bed volumes for the continuous and discontinuous filtration set-up respectively. A drastic drop in the amount of backflushing and overflowing was observed. However, an increased removal of dead biofilm by backflushing in the first day after the chlorination was also noticed.

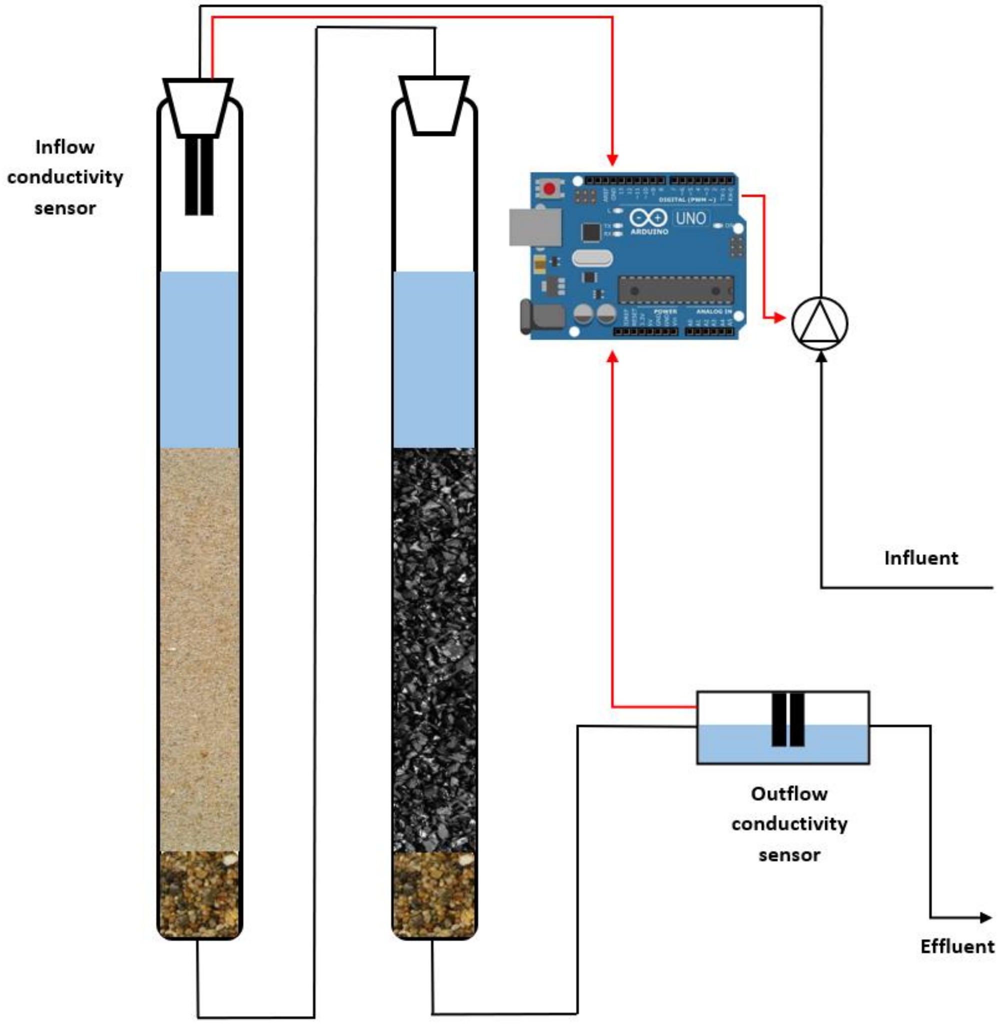


Figure 4-5: Schematic representation of the automated filtration set-up using two conductivity sensors and a data acquisition controller as automatic transmitter connected with the peristaltic pump

4.3 Long-term continuous treatment

The results of the long-term column tests are discussed in this section. The filtration lasted around 2 months. The set-up is divided into two parts, a sand filter and a GAC column. Filtered bed volumes (BV) were used as a time indicator. Therefore, the flow rate of the peristaltic pump was kept constant throughout the whole duration of the experiment (see 3.3.2) and the time was also registered. The complete system can be reviewed in Figure 3-1. Column I indicates the sand filter and column II represents the GAC filter.

4.3.1 Standard quality parameters of the continuous filtration

Before discussing the results of the standard quality water parameters, it should be made clear that the runoff used for these tests is different from freshly harvested runoff from a roof (as is the case for the values presented in Table 2-1 from the review of De Buyck *et al.* (2021)) as this runoff had already undergone some (rudimentary) treatment. As discussed in 3.2, treatment consisted of a sediment and lava rock filter and the water was also allowed to settle in the storage tank. The water quality can therefore be deemed better and the values measured are almost always lower than required for drinking water (see Table 2-5).

The standard water quality parameters were measured every time a sample was taken to obtain a clear overview of the changes that might occur over time. The pH of the influent ranged from 6.5 to 7.3. The pH of the influent showed some natural variations due to the fact that fresh roof runoff was added to the storage containers on a regular basis. As no acids were used to store the spike solutions and the influent was not acidified, the obtained pH of the influent was as realistic as possible and corresponds with the natural pH of the harvested roof runoff. Typical pH values for the runoff originating from a ceramic tile roof are around 6.5 on average with a maximal observed value of 8.5 (calculated in the review of De Buyck *et al.* (2021)). The pH of the sand filter and GAC column showed no significant changes over time (Figure 7-1, supplementary materials). All GAC effluent samples conformed the Flemish drinking water standards (6.5 < pH < 9.2) as reported in Table 2-5.

The turbidity of harvested rainwater is regularly discussed in the literature and is on average around 46 NTU (see Table 2-1). This average value also includes scenarios where no first flush diverters are installed. The turbidity starts off low after 500 BV with values between 0.50 and 0.85 NTU and gradually increases over time to an absolute maximum of 4.27 NTU after 14 000 BV due to the performed chlorination after 13 000 BV. After this, the turbidity drops again to values comparable to the starting situation (see Figure 4-6).

The turbidity values are low in comparison with the proposed values in Table 2-1. The reason for this is that a filtration system consisting of a sediment and lava rock filter was present at the location where the rainwater was collected. Another reason is that the harvested rainwater was poured into a second storage container. It was then spiked and pumped to the first storage container from which it was then loaded onto the columns. The first container acted therefore as a settler which removed turbidity.

The turbidity also varies as the different batches of rainwater used as influent were susceptible to different meteorological circumstances. As discussed in 2.1.2, the length of the antecedent dry period influences the amount of dry deposition on the catchment roof. Extended periods of drought can therefore increase the turbidity as dry deposition starts to pile up and is then washed off during the next precipitation event. After chlorination, turbidity increased as the biofilm was detached from the surfaces.

Very low turbidity values for the effluents were measured which leads to the conclusion that these effluents were conform the drinking water quality norms for turbidity. When comparing these values with the prescribed quality norms for reuse of reclaimed water in Spain (shown in Table 2-7), it can be concluded that this water, in terms of turbidity, can be used for all proposed applications with the exception for usage in cooling towers and condensers (requirement for turbidity is < 1 NTU), however the obtained turbidity came very close to the requirement.

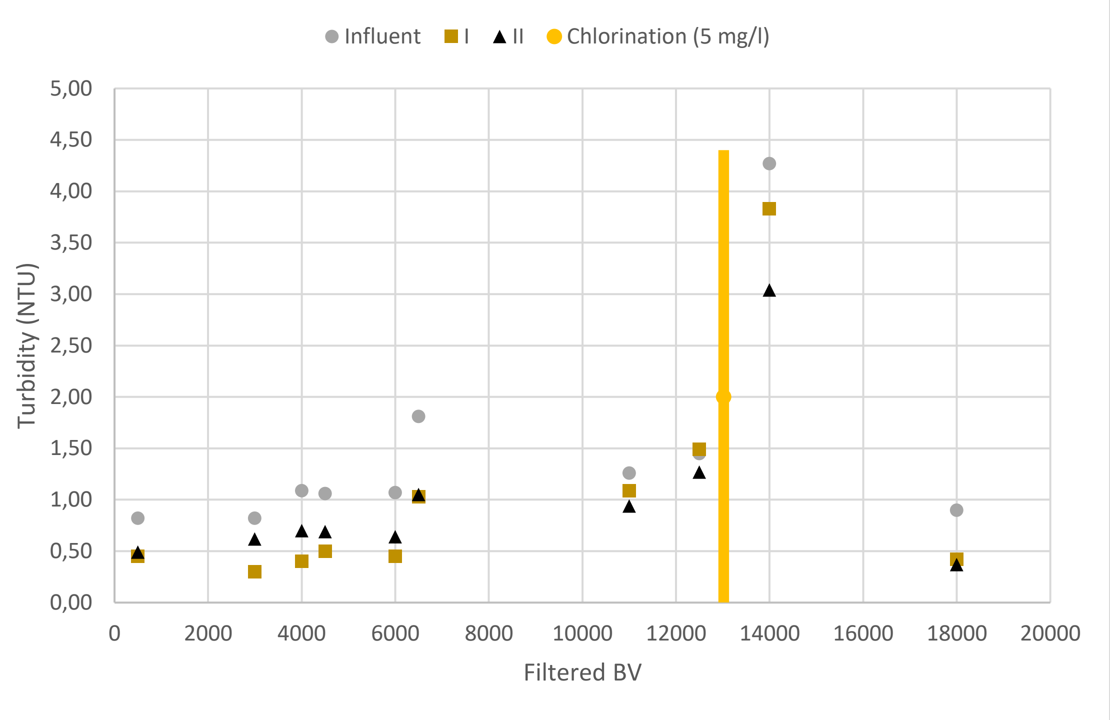


Figure 4-6: Evolution of turbidity over the course of time for the continuous filtration system (yellow line = performed chlorination, I = sand filter, II = GAC)

The EC of untreated rainwater is on average 51 µS/cm as reported in Table 2-1. The conductivity for the influents ranged from 69 to 103 µS/cm as shown in Figure 4-7. The roof runoff was also once again susceptible to variations in meteorology resulting in differences concerning the build-up of dry deposition and variations in wet deposition as discussed in 2.1.1 and 2.1.2.

The higher conductivity at 4500 BV for the effluent of the GAC filter (column II) is the result of the presence of tap water which was used to backflush the sand filter. The presence of tap water residues in the GAC filter effluent was confirmed by the detected calcium levels: the levels in the effluent of this filter (approximately 12 mg/l) were twice as high as the calcium concentrations in the influent and effluent of the sand filter (approximately 6 mg/l). The conductivity of all influents and effluents was lower than the Flemish drinking water quality standards (EC_{max} = 2100 µS/cm) presented in Table 2-5.

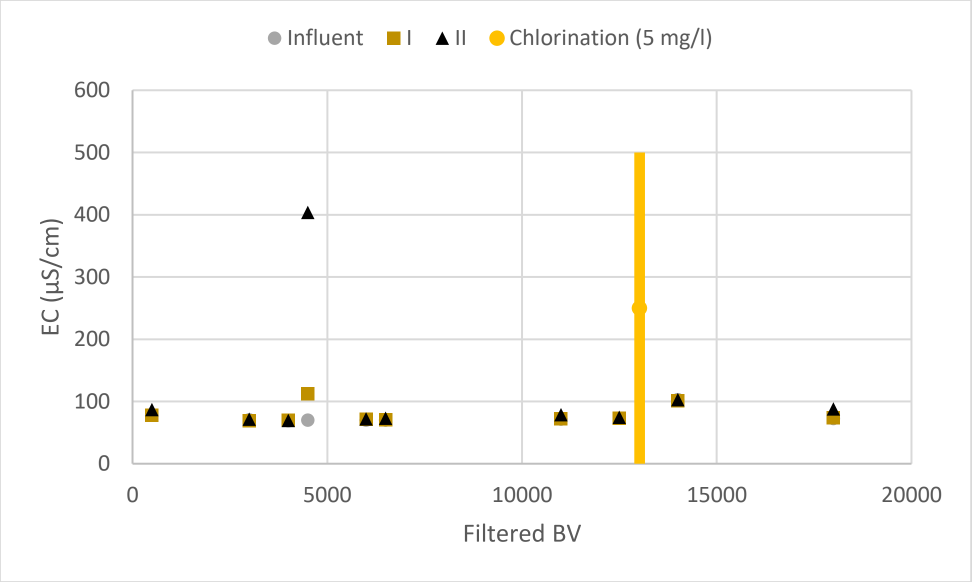


Figure 4-7: Evolution of EC over the course of time for the continuous filtration (yellow line = performed chlorination, I = sand filter, II = GAC)

The changes in COD over the course of the experiment are shown in Figure 4-8. As calculated in 3.2, the presence of methanol caused an increase of COD by 11.9 mg O_2/l by spiking 1 ml of methanol per 100 l of influent. Not only the methanol but also the dissolved organic compounds added to an increase in COD in comparison with freshly harvested rainwater, although only in a very small amount as calculated in Table 2-3. The COD of the influents ranged from < 15 mg/l (LOD of test kit) to 37.1 mg O_2/l. When comparing these concentrations with the value reported in Table 2-1, which gives an average of 96 mg O_2/l, it can be concluded that these concentrations are low, even with the addition of extra organic molecules during spiking. However, this average concentration was calculated by taking into account all measured CODs with no distinction between different roofing materials. When looking at the concentrations gathered from ceramic tile roofs only, the average concentration reduces to 57 mg O_2/l with a maximal observed concentration of 160 mg O_2/l.

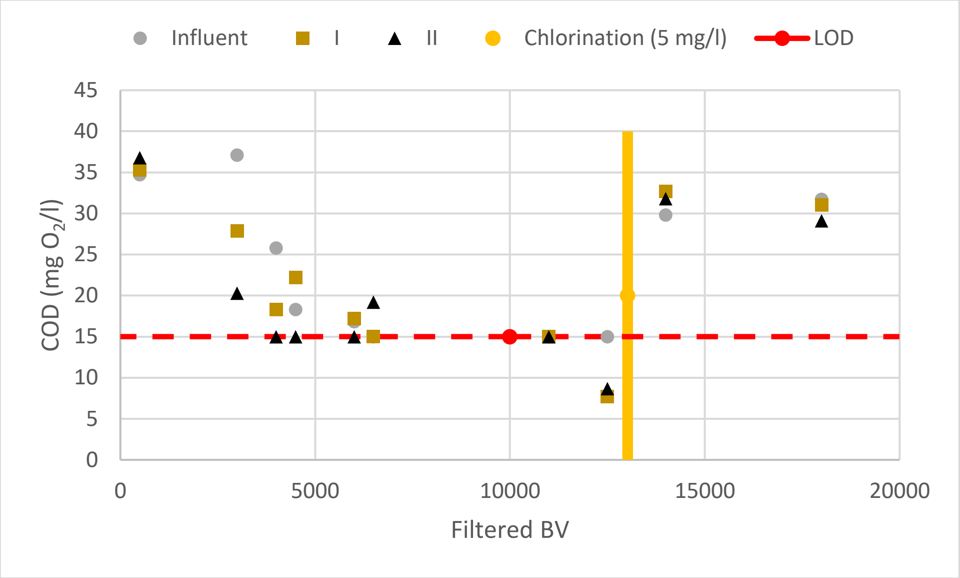


Figure 4-8: Changes in COD over the course of time for the continuous filtration (LOD (red dotted line) = 15 and 1 mg O_2/l, yellow line = performed chlorination, I = sand filter, II = GAC)

After 3000 BV, the expected behaviour for the removal of COD using a sand filter followed by GAC was obtained. The GAC effluent should therefore have the lowest concentration while the sand filter effluent should have a lower concentration than the influent.

As discussed in the research of Naddeo, Scannapieco and Belgiorno (2013), which is summarised in Table 2-8, organic material can be present in the form of particulate organic matter (POM) and dissolved organic matter (DOM). In that research a MF membrane was used to filter POM while GAC effectively removed DOM. A COD removal of 50% was obtained starting from an influent with a concentration of 4.8 mg O_2/l and ending with an effluent concentration of 2.4 mg O_2/l. In this research a sand filter was used for the removal of POM instead of a MF membrane. The removal percentage calculated after 3000 BV, which represents the ideal filtration behaviour, is 45%. This value is in accordance with the removal efficiency of Naddeo, Scannapieco and Belgiorno (2013) which indicates that a sand filter followed by GAC is effective in removing COD from harvested rainwater. The amount of analysed samples that were above the LOD are too small to make a definitive conclusion on the removal of COD.

When reviewing the changes in COD in Figure 4-8, a decrease in organic content can be observed until after 13 000 BV where the chlorination happened. This is probably because the organic matter present in the influent was biologically degraded and absorbed by the biofilm for its growth (Boltz, 2005). The biofilm had more optimal growth conditions as the temperature in the lab was higher than in a normal outdoor scenario where the storage tanks are underground. When the biofilm was killed off, COD concentrations were once again similar to levels in the beginning. Chlorination has thus a positive effect on the operation of a sand filter but when the dosage is applied directly in an underground storage tank, an increase in COD can be observed. However the compounds that cause this increase are not identified. These can either be desorbed organic compounds from the GAC which poses health risks in the long term or they can be degradation products of the biofilm. After 18 000 BV, the GAC filter is once again capable of removing COD after performing a chlorination however only in small amounts. The decrease of COD levels after 4500 BV leads to unusable results until 12 500 BV. Long periods of heavy rainfall lead to lower concentrations of organic compounds as deposition happens less (see 2.1.2). The reason why the samples after 12 500 BV are below the LOD indicated in Figure 4-8 is that the sand filter effluent (I) and GAC effluent (II) were measured using kits with an LOD of 5 mg O_2/l, the influent was measured using a kit with an LOD of 15 mg O_2/l.

The absorbance over time at wavelength 254 nm (UV_{254}) is presented in Figure 4-9. The organic compounds in the influent were partially removed by the sand filter and the activated carbon. The sand filter removed the largest part of the present absorbing organic content in most cases, the GAC only removed a smaller portion. The total organic content after chlorination increases significantly due to the releasing biofilm and here, only a small part is removed by the sand filter and the largest part by the GAC.

The correlation between COD and the spectrophotometric absorbance at 254 nm (UV_{254}) has been frequently reported in literature (Mooney, 2000; Albrektiene *et al.*, 2012; Serajuddin *et al.*, 2018). The reason for the use of UV_{254} is that is can be used online to quickly asses the water quality in contrast with manual COD measurements. At 254 nm, double bounds which are present in organic molecules, absorb the UV-radiation (Blondeel *et al.*, 2014). Naddeo, Scannapieco and Belgiorno (2013) did not only measure the removal in COD but also the removal in UV_{254}. The removal in absorbance, using their FAD-unit discussed in 2.5, was 38% while the COD removal was 50%. A correlation between the two parameters was not discussed in the research.

To determine whether a correlation existed between the COD removal and UV_{254} removal, removal percentages were calculated for both the sand filter and the GAC and were then plotted (see Figure 7-2, supplementary materials). Sample points lower than the LOD are not included in the dataset. No significant correlation can exist as only three sample points of both the sand filter (I) and GAC (II) were situated in the first quadrant of the plot. Interference of methanol could have occurred as this compound does not possess chromophores which absorb in the 254 nm region.

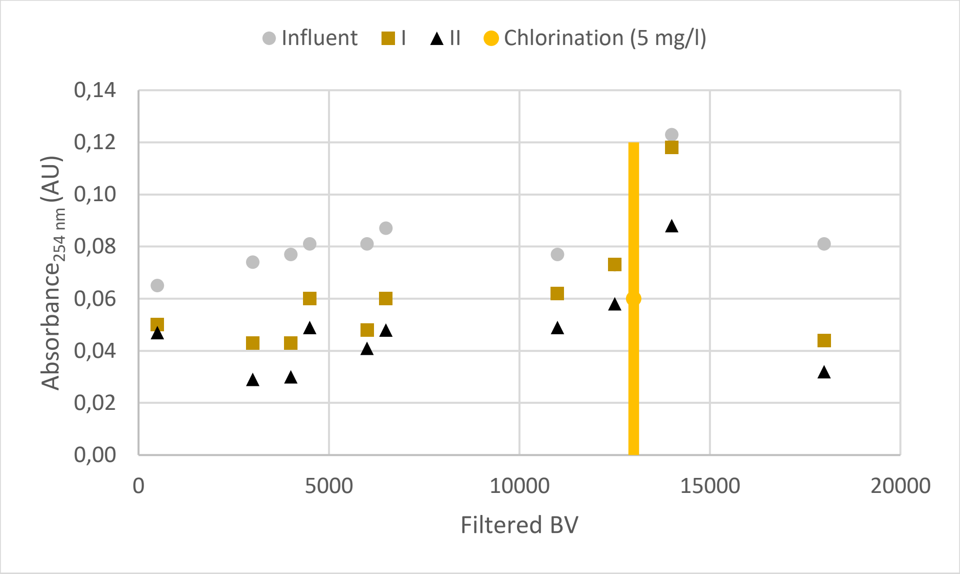


Figure 4-9: Changes in absorbance over time of UV-irradiation at 254 nm during continuous filtration (yellow line = performed chlorination, I = sand filter, II = GAC)

The changes in total nitrogen are shown in Figure 7-3 (supplementary materials). At the start of the experiment a value of the influent of 1.51 mg/l was measured. The sand filter and GAC both removed a portion of nitrogen to end up with an effluent concentration of 1.19 mg/l. A sand filter will withhold a portion of biomolecules containing nitrogen groups (vegetal matter, insects, biofilm etc.) while nitrate is partially removed by fresh GAC (Shahmoradi, Zade and Torabian, 2015). According to Table 2-1, an average value for TN of 5.4 mg/l for fresh roof runoff can be expected. However, when looking per type of roofing material, an average value of 4.5 mg/l is found with a maximal observed concentration of 20 mg/l for a ceramic tile roof (De Buyck *et al.*, 2021). These values are high when comparing them with the influent concentrations. This can be attributed to sedimentation in the storage tank and filtration by sediment filter as discussed with the COD.

Total nitrogen concentrations did not vary much throughout the duration of the experiment. Small changes are attributed to the natural variation of the influent. The only outlier is the GAC effluent (II) sample after 4500 BV which had a concentration of 1.91 mg/l. This was most likely due to the presence of tap water residues in the GAC column which was confirmed by an increased conductivity (see Figure 4-7). The average concentration of TN in tap water for 2019 was 5.2 mg TN/l (VMM, 2019). It is therefore worth noting that the concentrations of total nitrogen in rainwater are notably lower than the concentration in tap water.

4.3.2 Soluble metals of the continuous filtration

The first group of investigated micropollutants were soluble metals. As discussed in 2.1.2 and 2.1.4, these compounds can end up in roof runoff through deposition and leaching processes. The choice to follow up on these 9 metals is based on Table 2-2 where the metals with the highest concentrations found in roof runoff were selected while the frequency of detection in samples was another reason why these were specifically chosen.

The removal of metals by activated carbon is more difficult in comparison with organic compounds as GAC is not charged and has a better affinity for the neutral organic compounds (Uzun and Güzel, 2000). This results in a competition between organic compounds and metals (Tchobanoglous *et al.*, 2014). Metals are thus expected to be less adsorbed and show less consistent adsorptive behaviour. As shown in Table 4-3, chromium, iron and lead had influent and effluent concentrations which were lower than the LOQ. Only few GAC effluent samples where chromium and iron were analysed, had concentrations higher than the LOQ. The concentration of lead in every sample was lower or equal to the LOQ. These metals were spiked in accordance with the concentrations given in Table 3-1, but these metals were not detectable in the samples. The samples were also filtered (0.45 µm filter) before being acidified so metals adhering to particles could not be measured (see 3.6.2). This can be explained by the occurrence of sedimentation: these metals adhere to particles which are present in roof runoff. This theory is confirmed by the presence of a red sludge layer at the bottom which could be observed when a storage container was empty. This either indicates the presence of algae or mosses where metals could have adhered to or the precipitation of certain metals. The red sludge present at the bottom of an empty container is shown in Figure 4-10.

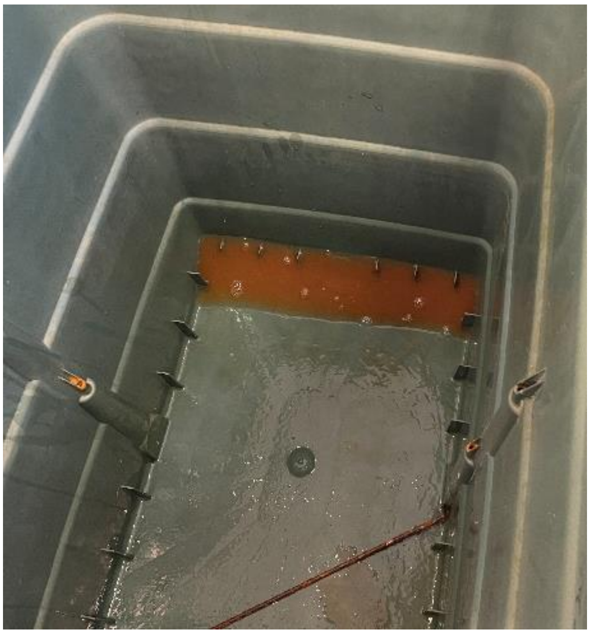


Figure 4-10: Red sludge present in the filtration system

After analysing the results of the metal analyses, extra iron and lead were spiked to make sure that an error in spiking quantity could be ruled out as the reason for the low observed concentrations. This increase in concentration in iron and lead was not detected in the influent or effluent when these were analysed so it was concluded that sedimentation was the major reason why the concentrations of some metals were lower than their respective LOQ. This behaviour is also reported in the literature (Nguyen *et al.*, 2005; Magyar *et al.*, 2007, 2008). The addition of acids would increase the solubility of metals but was not carried out to keep the experiment as realistic as possible.

Table 4-3: Influent and effluent concentration range for the sand filter and GAC column during continuous filtration (= limit of quantification)*

Metal	Influent concentration (µg/l)	Concentration range effluent sand filter (µg/l)	Concentration range effluent GAC (µg/l)	Flemish drinking water quality norms for metals (µg/l)
Al	35.9 – 57.5	33.2 -52.4	12.7 – 51.6	200
Cr	< 3.1* – 15.5	< 3.1* – 13.1	< 3.1* – 5.3	50
Cu	132.7 – 495.4	48.9 – 309.8	50.0 – 183.1	2000
Fe	< 2.1* – 20.4	< 2.1* – 15.2	< 2.1* – 22.6	200
Mn	6.1 – 35.9	6.1 – 30.7	7.1 – 46.4	50
Ni	7.6 – 26.8	7.9 – 23.6	5.2 – 26.1	20
Pb	< 8.1*	< 8.1*	< 8.1*	10
Sr	39.3 – 134.6	35.9 – 113.1	77.4 – 228.4	-
Zn	774.2 – 2596	775.9 – 2200.1	1401.1 – 2414.7	5000

Results of the removal of metals by sand filtration are shown in Figure 4-11 and Figure 4-12. The only metal that was not included was lead because even the concentration in the influents was lower than the LOQ. It can be concluded that removal was quite high before 5000 BV of the experiment. After 10 000 BV and before the chlorination, only copper showed high removal (56% and 42%). The rest of the metals had low removal percentages or even exhibited some leaching (from the filtration medium, i.e. sand), for example manganese, zinc, iron and strontium. This last metal also showed some leaching behaviour after 4500 BV. The removal behaviour immediately after chlorination did not change much. The removal after 18 000 BV was at the same level as the start of the experiment.

A sand filter can thus be useful to withhold soluble metals from the influent. However, the amount of backflushes throughout the experiment was high (almost every day) to keep the biofilm growth under control. Under real life circumstances (colder environment which limits biofilm growth), backflushing would not occur as regular so the filtration efficiency of a sand filter might deviate. The uptake of metals for the growth of biofilm could also have played a part but only to a certain amount. However this cannot be concluded with certainty as the biofilm was not analysed for the presence of metals. Sorption of metals to the container wall can also have influenced the removal as discussed in 4.1.1.

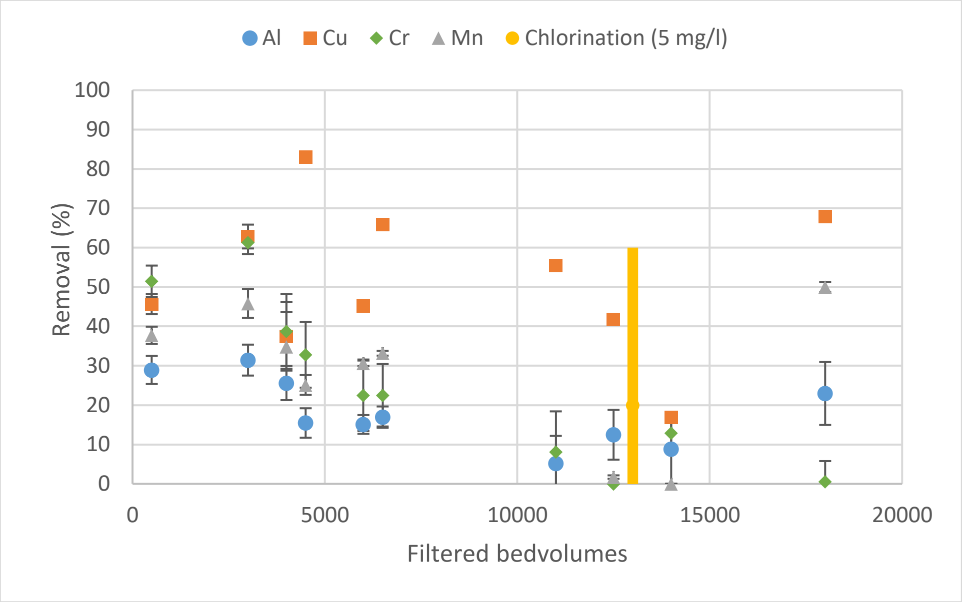


Figure 4-11: Removal of aluminium, copper, chromium and manganese by sand filtration during continuous filtration (yellow line = performed chlorination)

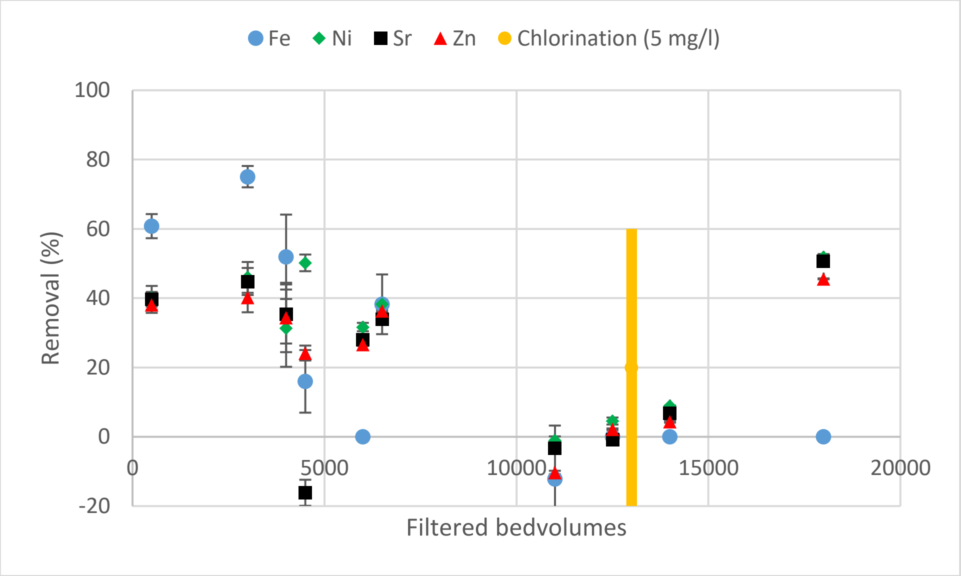


Figure 4-12: Removal of iron, nickel, strontium and zinc by sand filtration during continuous filtration (yellow line = performed chlorination)

The removal of soluble metals by combining sand filtration with GAC is presented in Figure 4-13 and Figure 4-14. Lead and iron were not included in the dataset for this reason as the effluent concentrations were for the majority of samples lower than the LOQ. The data for chromium is limited to the first four samples points as the rest of the influent concentrations were below the LOQ. A minimal removal for these four points was determined as the effluent concentrations were below the LOQ. The removal was at least 77% after 500 BV, 91% volume 3000 BV, 80% after 4000 BV and 49% after 4500 BV.

The removal behaviour of the metals fluctuates a lot. The expected behaviour would be a descending curve as the removal of metals should decrease over time as there is a depletion of adsorption locations. However, ion exchange reactions also play a part in the removal of metals from the influent. The only metal that somewhat shows a decreasing evolution in Figure 4-13 is copper. At the start of the filtration experiment, new activated carbon was used. The negative removal percentage of Al, Mn, Ni, Sr and Zn after bed volume 500 is probably caused by the presence of aluminium and other transition metals (e.g. manganese and iron) in the fresh activated carbon according to the manufacturer (Desotec, 2021). This leaching process only takes places when fresh activated carbon is used and only during the first 500 BV. It should be kept in mind that the first 500 BV can deviate from the predetermined water quality when applying GAC for the removal of pollutants.

Another mechanism that plays an important role in the adsorption is the overloading of GAC. If high concentrations of soluble metals are present, all the available adsorption locations will quickly be filled and competition between metals can take place which leads to replacement reactions. The competition will be won by elements who are more readily adsorbed than others. Presence of tap water (used for backwashing) which has high concentrations of calcium also has a negative effect on the lifetime of GAC. Calcium easily forms complexes with organic material and GAC (Lambert, Miguel and Graham, 2002). This leads to less available adsorption spots for trace elements. Full-out leaching of metals from GAC can only happen with low levels of pH which did not occur during this experiment (see Figure 7-1). Further tests should be conducted to determine the leaching behaviour and conditions to accurately indicate the reason for the leaching of soluble metals throughout the experiment.

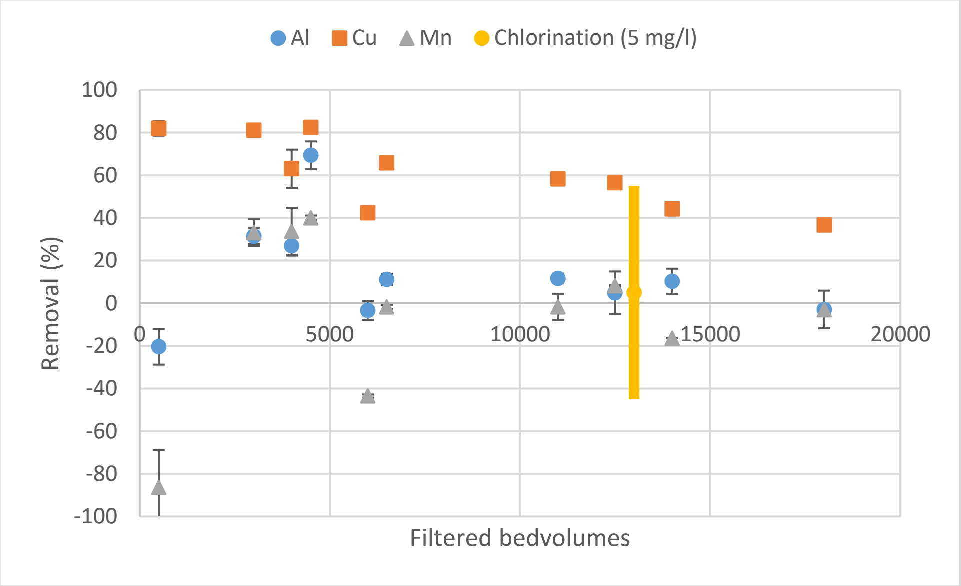


Figure 4-13: Removal of aluminium, copper and manganese by a combination of sand and GAC filtration over time during continuous filtration (yellow line = performed chlorination)

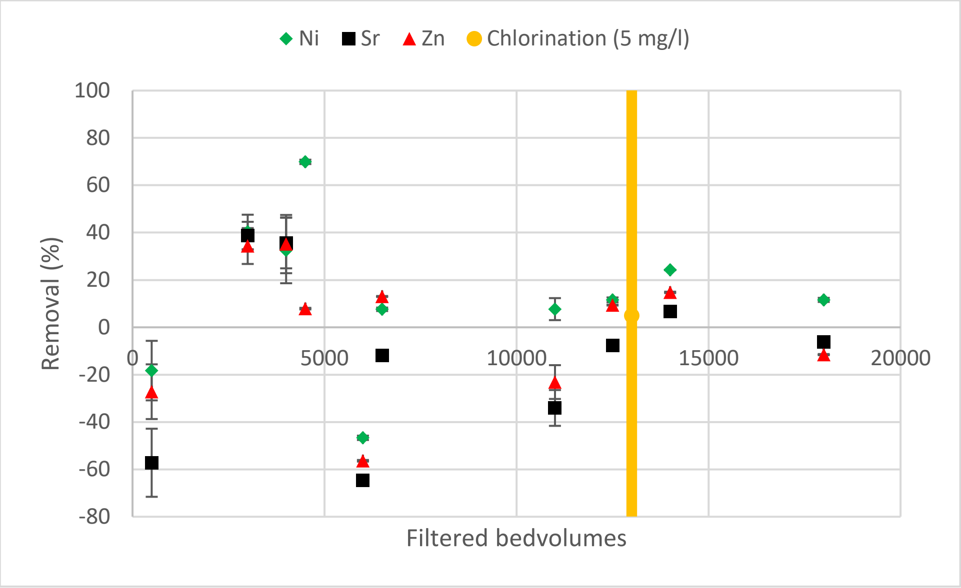


Figure 4-14: Removal of nickel, strontium and zinc by a combination of sand and GAC filtration over time during continuous filtration (yellow line = performed chlorination)

The allowed concentrations of heavy metals in treated water effluents are strongly regulated when using this water for consumption purposes (see Table 2-5). Some observations can be made when comparing the concentration range of the GAC effluent for each metal with the maximum allowed concentration in Flemish drinking water as shown in Table 4-3. All metals with the exception of nickel are below the legally allowed concentration. The average concentration of nickel in the effluent of all samples taken was 17.3 µg/l and the concentration was only for 20% of the samples taken, higher than the legal limit. An enhanced removal could be obtained by using a negatively charged type of activated carbon on which metals can adsorb better or by using other techniques such as ion exchangers (IEX).

4.3.3 PAHs, phthalates and herbicide of the continuous filtration

The second important group of micropollutant are the organic compounds. Rapid sand filtration has been described in the literature as a possible pathway to remove organic micropollutants besides activated carbon, advanced oxidation processes or membrane filtration techniques. Removal mechanisms by sand filtration that are reported are (a combination of) adsorption onto granular material, oxidation by iron and manganese oxides, biodegradation by autotrophic and heterotrophic bacteria. Of these four mechanisms is biodegradation the most significant in the literature for longer retention times. The potential of organic micropollutant is still largely unexplored due to the variable removal. Organic compounds can also be changed during biodegradation into transformation products which complicates the follow-up of the removal (Wang *et al.*, 2020).

Figure 4-15, Figure 4-16 and Figure 4-17 show the removal of the organic compounds by sand filtration over time. The removal of PAHs by sand filtration under normal conditions is partially dependent on their molecular weight. PAHs with a high molecular weight tend to dissolve less and adhere more to particles (Kim, Kabir and Kabir, 2015). The results obtained after sand filtration do not follow this theory as shown in Table 4-4. Not only the molecular weight but also the water solubility of organic compounds can play a role. Solubility in water is expressed in a concentration at 25°C while hydrophobicity is expressed by a log K_{ow}-value. A higher log K_{ow}-value, i.e. the difference in concentration of the solute between a non-polar (n-octanol) and a polar liquid (water), means that the solute is present in a higher concentration in n-octanol and is thus less soluble in water. The values for each organic compound that was spiked in this experiment are given in Table 3-3.

When discussing the results of the sand filtration, it can be concluded that only a partial removal was observed and it was variable for most components. Only phenanthrene, fluoranthene and pyrene show a similar evolution over time. The removal starts off at around 40% and drops to around 0% after 5000 BV. The high water solubility and low log K_{ow}-values for naphthalene and diuron can explain the ineffectiveness for removal of these compounds by sand filtration. Low recoveries at higher concentrations of phthalates can be caused by the formation of colloids, aggregates and micro drops. Sorption onto glasswork and SPE equipment besides sorption onto particles could have also played a role in the fluctuating removal behaviour of both PAHs and phthalates. These effects were reported by Wang *et al.* (2007) and Jara *et al.* (2000).

Some observations can be made when comparing the removal of organic compounds from the influent by sand filtration with the concentration of COD (in column I) during the continuous filtration (see Figure 4-8). The COD concentration of the sand filter effluent after 500 BV is higher than the influent concentration while all removals were positive. The COD concentration of the sand filter effluent for the next sample points was constantly lower than the influent but leaching op PAHs and phthalates occurred. No correlation between COD and organic removal after 4500 BV could be found as the COD concentrations were lower than the LOD. The COD concentration of the sand filter effluent after chlorination was higher than the influent concentration and leaching of multiple PAHs was observed. The COD concentration of the sand filter effluent after 18 000 BV is lower than the influent and the removal of all organic compounds was once again positive.

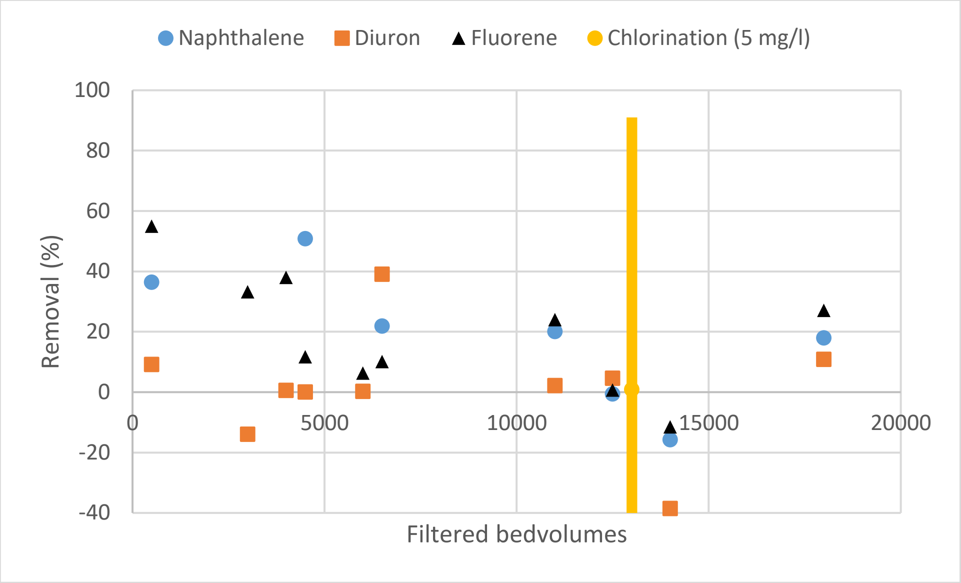


Figure 4-15: Removal of naphthalene, diuron and fluorene by sand filtration during continuous filtration (yellow line = performed chlorination)

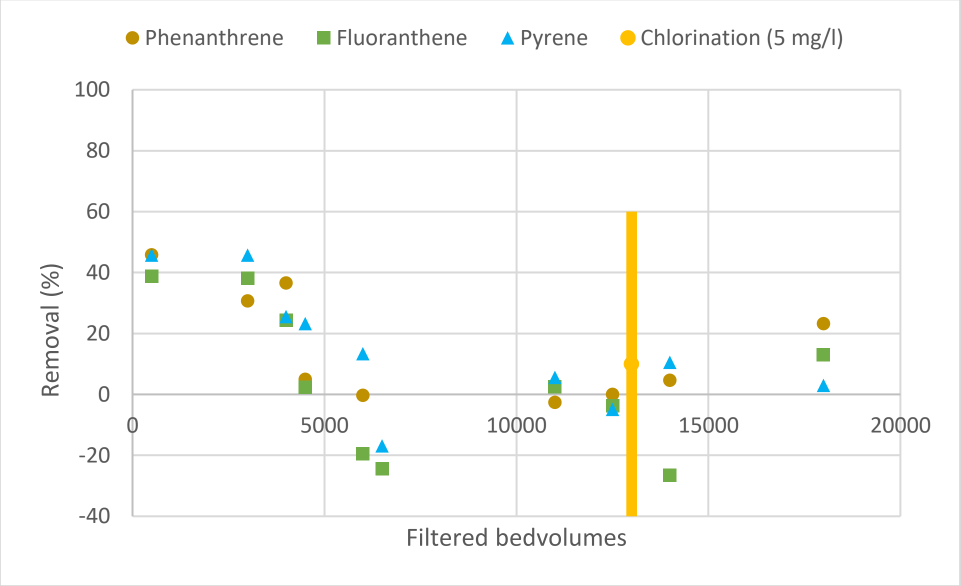


Figure 4-16: Removal of phenanthrene, fluoranthene and pyrene by sand filtration during continuous filtration (yellow line = performed chlorination)

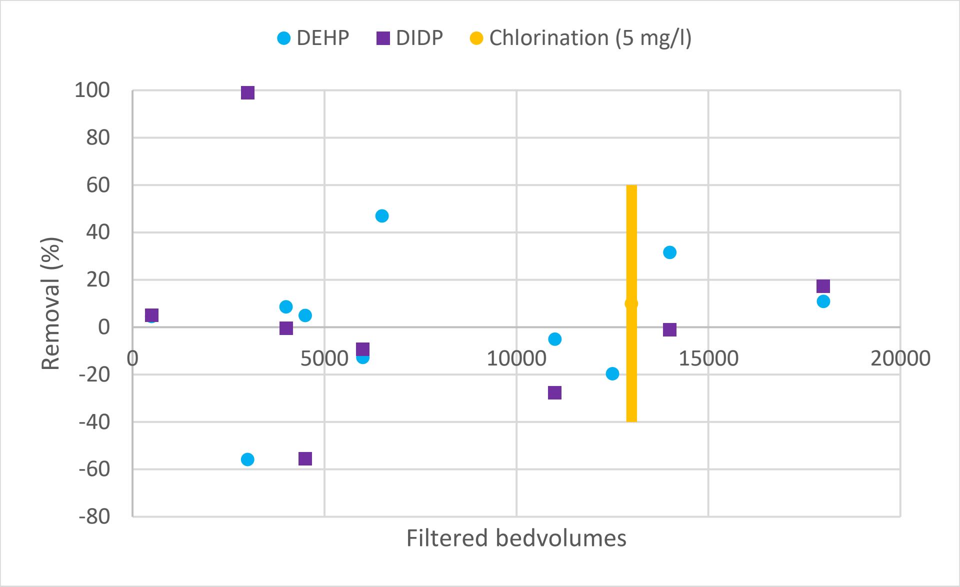


Figure 4-17: Removal of DEHP and DIDP by sand filtration during continuous filtration (yellow line = performed chlorination)

The removal percentages of organic micropollutants by a combination of sand filtration and GAC are presented in Figure 4-18, Figure 4-19 and Figure 4-20. What immediately stands out when comparing these removals with the removals of metals discussed in 4.3.2 is that better results are obtained.

Some general observations can be made: high removal efficiencies for PAHs were observed as could be expected with this particular type of GAC (see 3.3.2). The lower removal of naphthalene can be caused by the low molecular weight in comparison with other PAHs. As discussed in 2.4.2.2, organic compounds with a high molecular weight are better retained by AC (Uzun and Güzel, 2000). Diuron is also removed in high concentrations from the influent. Finally, after 18 000 BV, all compounds had a higher removal than before the chlorination. The performed chlorination had thus an immediate effect on the increase in removal of organic micropollutants. The addition of chlorine resulted in the oxidation of components that were adsorbed onto the GAC. The compounds were released from the GAC because of this oxidation reaction and new adsorption sites became therefore available (Evans *et al.*, 1998). This explains the improved removal behaviour of the organic pollutants which can be observed some bed volumes after the chlorination (see Figure 4-15, Figure 4-16 and Figure 4-17).

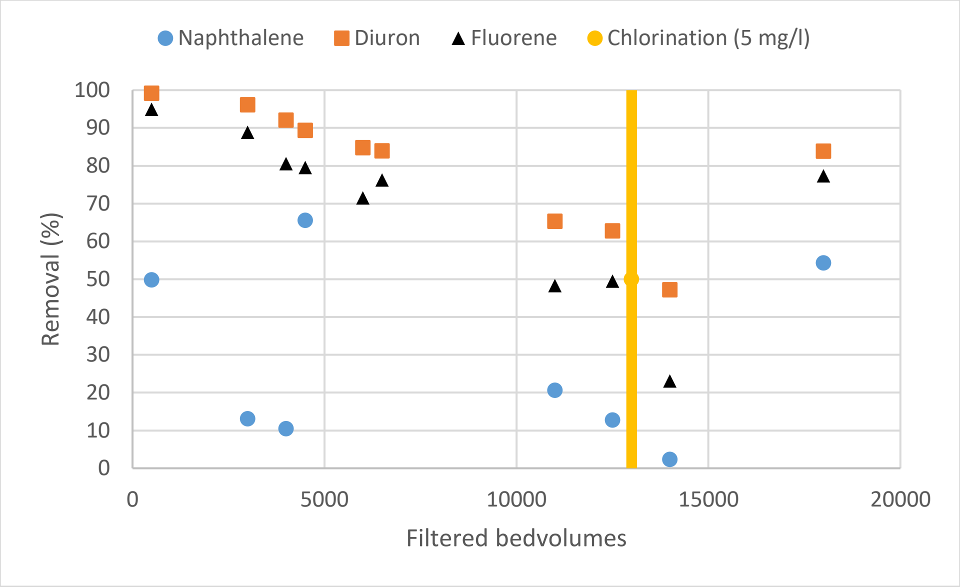


Figure 4-18: Removal of naphthalene, diuron and fluorene over time by a combination of sand filtration and GAC during continuous filtration (yellow line = performed chlorination)

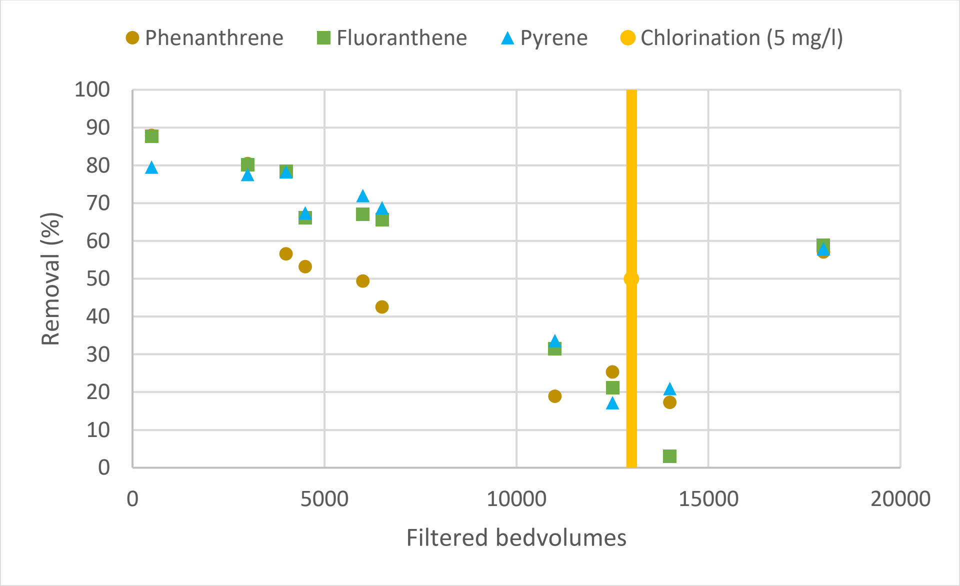


Figure 4-19: Removal of phenanthrene, fluoranthene and pyrene over time by a combination of sand filtration and GAC during continuous filtration (yellow line = performed chlorination)

The removal of phthalates is presented in Figure 4-20. The removal of both compounds fluctuates a lot. There are several reasons for this. The first one is that the sensitivity of the GC-MS for DIDP was low. A second possibility is that contamination happened during sample preparation. The third reason is the solubility of the phthalates as discussed previously (see Table 3-3). The last reason is that DEHP is a widely used plasticizer in the industry. A lot of lab equipment, such as storage containers, tubing etc. were made of plastic. DEHP is also mentioned in the WHO drinking water guidelines with a maximum recommended concentration of 8 µg/l as this component can be toxic and is widely used in plastics (see 2.2.7). Only 2 samples did not conform with this guideline. An increase in concentration of DIDP after sand filtration was observed for 70% of the samples. A maximum concentration of 100 ng/l for all PAHs combined is allowed in drinking water in Flanders (see Table

4-4). Half of the samples taken from the continuous set-up had a GAC-effluent concentration which exceeded this limit. Lastly, diuron is also mentioned in the drinking water norm: 100 ng/l is allowed in drinking water. Around 80% of the samples had a concentration which was higher. Effluent concentrations which exceeded the LOD for diuron (830 ng/l) were considered due to the high LOQ of diuron.

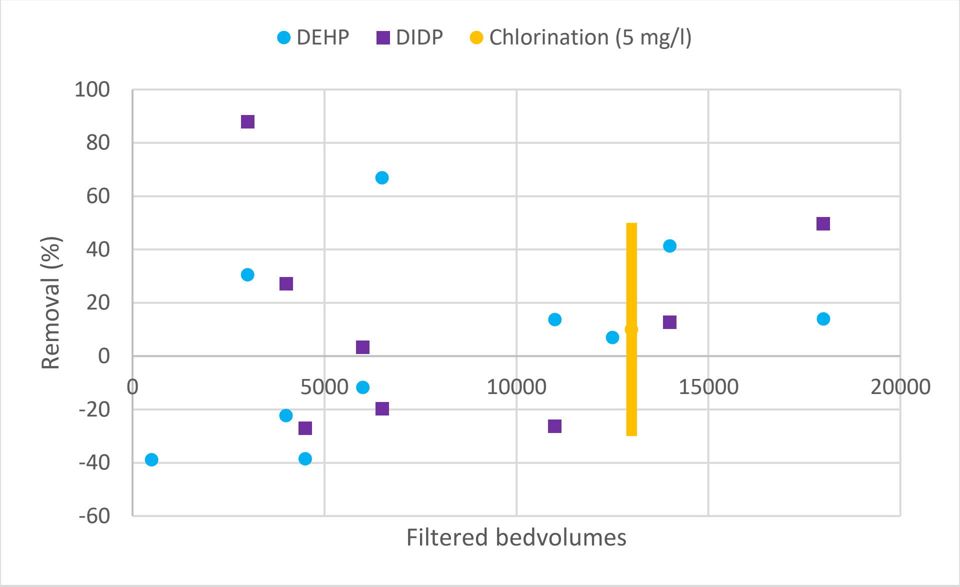


Figure 4-20: Removal of DEHP and DIDP over time by a combination of sand filtration and GAC during continuous filtration (yellow line = performed chlorination)

Table 4-4: Summary of the removal by sand filtration and GAC separately of the target organic compounds and their median effluent concentration for the continuous filtration (= limit of detection)*

Compound	**Maximum removal by sand filter (%)**	**Median sand filter effluent concentration (ng/l)**	**Removal range GAC (%)**	**Median GAC effluent concentration (ng/l)**	**Flemish drinking water quality norms for organics (ng/l)**
Naphthalene	51	66	-77 – 47	72	100 (total PAHs)
Diuron	39	9301	61 – 99	1451	500 (total pesticides) 100 (individual)
Fluorene	55	13	38 – 88	9*	100 (total PAHs)
Phenanthrene	57	23	13 – 82	13*	100 (total PAHs)
Fluoranthene	40	7	15 – 77	3*	100 (total PAHs)
Pyrene	89	4*	26 – 72	4*	100 (total PAHs)
DEHP	46	168	-92 – 58	16*	8000 (guideline WHO)
DIDP	78	25 435	2 – 100	16 297	-

4.3.4 Microbiological quality

Microbial quality is besides the physicochemical quality an important parameter to take in consideration when reusing runoff. Rainwater is almost always contaminated with micro-organisms (see 2.2.8) and must therefore be treated by disinfection (see

2.4.3) to comply with legislative standards (see 2.3) for the various, potential uses of this type of water.

Samples were taken to determine the presence of micro-organisms in the (dis)continuous filtration set-up. A disinfection step was also included to determine the removal by UV-irradiation. The UV lamp was connected to the effluent of the GAC column of the continuous filter (II). The effluent of the GAC column of the discontinuous set-up was not disinfected as no major changes were expected between both set-ups for the microbiological parameters.

4.3.4.1 Total viable count (22°C)

Figure 4-21 shows the result of the count for each column and for the UV-disinfection step after 10 000 BV. The sample of the sand filter of the discontinuous set-up (III) showed an increased presence of micro-organisms in comparison with the rest of the samples. When comparing these results with the values found in the literature for total viable count (reported in Table 2-4), it can be concluded that these concentrations are in accordance with the reviewed literature. The maximum observed concentration for roof runoff in the reviewed literature was 2.02 x 10^6 CFU/ml, the influent concentration for this experiment was 8.23 x 10^4 CFU/ml. The GAC column of the discontinuous set-up (IV) had a high standard deviation. This is likely caused by a dilution error.

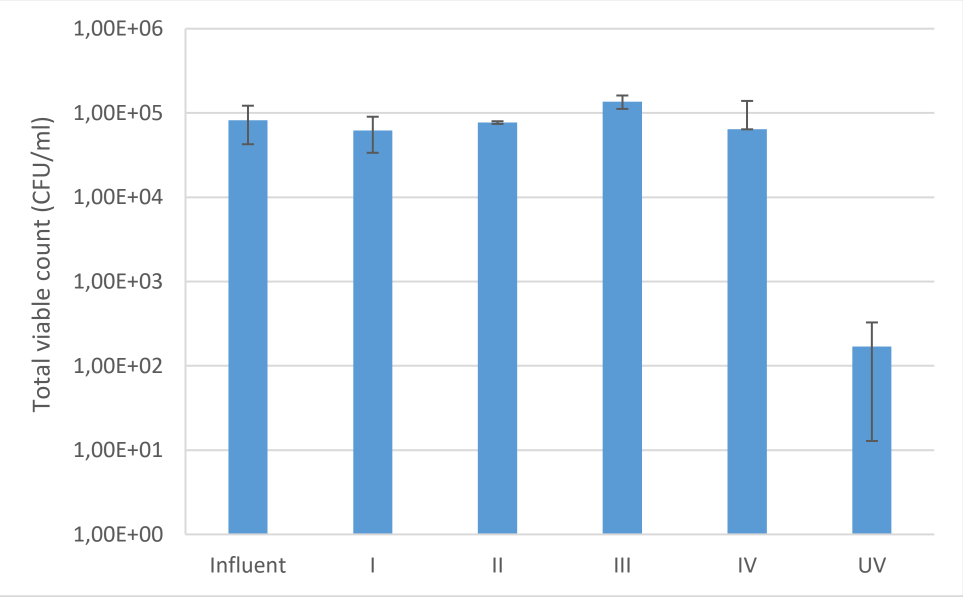


Figure 4-21: Results of the total viable count (22°C) after 10 000 BV (I: continuous sand filter, II: continuous GAC filter, III: discontinuous sand filter, IV: discontinuous GAC filter, UV: UV disinfected sample)

Table 4-5 shows the concentration for each effluent and their standard deviation. The log reduction was also calculated using the influent/effluent of the previous column and the effluent of the column/lamp. A negative log reduction of column III was calculated as the effluent of the sand filter had a higher concentration than the influent. Microbiological pollution thus occurred in this column. A small removal was obtained by the physicochemical treatment steps of the continuous set-up (I & II) but the bulk of the removal (2.68 log units) was obtained by using UV-irradiation.

Table 4-5: Concentrations of micro-organisms present in each effluent along with their respective log reduction (I: continuous sand filter, II: continuous GAC filter, III: discontinuous sand filter, IV: discontinuous GAC filter, UV: UV disinfected sample)

	Average concentration (CFU/ml)	**Absolute logarithmic change**
Influent	(8 ± 4) x 10^4	-
I	(6 ± 3) x 10^4	0.123
II	(7.7 ± 0.3) x 10^4	0.0289
III	(1.4 ± 0.3) x 10^5	0.221
IV	(6 ± 8) x 10^4	0.109
UV	(2 ± 2) x 10^2	2.68

The maximum allowed concentration using total viable count (22°C) is given in the Flemish drinking water quality norm and is shown in Table 2-5. A concentration of only 100 CFU/ml is legally allowed for drinking water while the effluent of the UV-lamp contained 170 CFU/ml. The effluent of the column test can therefore not be used for drinking water applications.

After sampling, the sand filter (I) was backflushed. A sample was taken from the backflush water as visible pieces of biofilm were washed out of the sand. This sample along with the other samples were processed to check if these pieces were indeed biofilm. The backflush had a concentration of (5 ± 2) x 10^4 CFU/ml. This concentration was in the same range as the influent concentration (see Table 4-5) so the microbial growth in the sand filter cannot be considered relevant.

4.3.4.2 Total coliforms (37°C)

The same samples, taken after 10 000 BV, which were plated for the determination of total viable count, were therefore used to determine the concentration of total coliforms and *E. coli* in the influent and effluents. Values found in the reviewed literature suggest a range of 0-10 x 10^3 CFU/100 ml for *E.coli* and 1-2.6 x 10^3 CFU/100 ml for total coliforms. Two methods were used for the determination of the coliform concentration: water filtration (100 ml of sample) and plating (1 ml of sample), both used the same medium for growth (see 3.6.4).

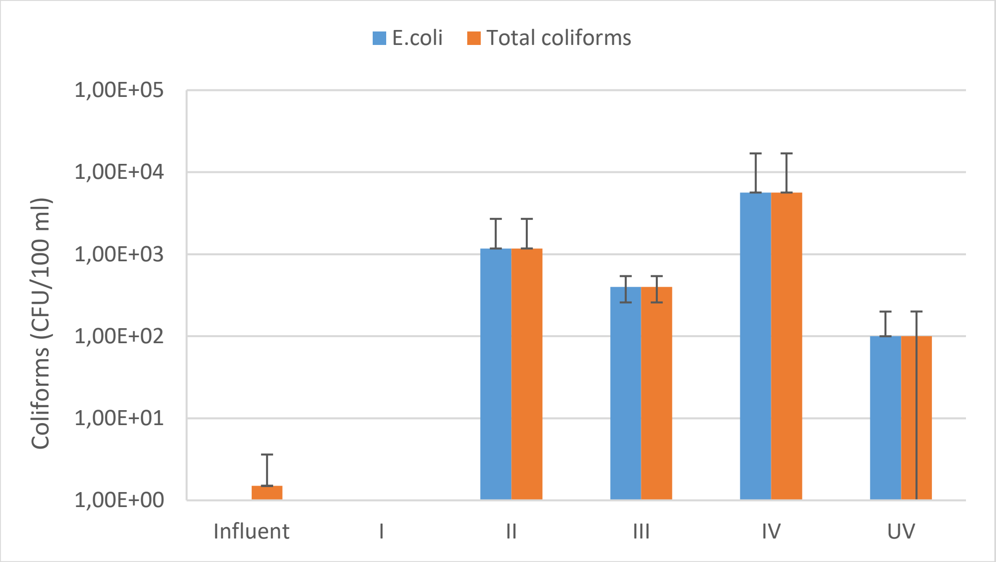


Figure 4-22: Concentrations of total coliforms and E. coli after 10 000 BV for each stream (I: continuous sand filter, II: continuous GAC filter, III: discontinuous sand filter, IV: discontinuous GAC filter, UV: UV disinfected sample)

Figure 4-22 shows the concentrations of total coliforms and *E. coli* for each stream.

Table 4-6 gives the concentrations and the log removal obtained for each step of the filtration process. The influent and the sand filter effluent (I) were free of *E. coli* while the GAC effluents (II & IV) showed an increased presence of almost exclusively *E. coli*. Reports in the literature confirm this trend and accredit the increase in bacteria in GAC filters to the fact that the porous carbon material is an excellent shelter for these micro-organisms. Bacteria can easily adhere to the porous material which shields it from fluid forces while at the same time neutralizes chlorine residuals and provides increased nutrient concentrations. Care should thus be taken when replacing activated carbon filter cartridges to avoid microbial contamination by wearing gloves (Trogolo, 2011). The sand filter of the discontinuous set-up (III) also increased the *E. coli* concentration with 400 CFU/100 ml. A large spike in coliform concentration was detected in the effluent of column IV. The large standard deviations on the concentrations are the result of differences in growth between the two applied techniques. The count of *E. coli* for column II using the filtration technique showed an average of only 1 colony. Using the plating technique, an average of 24 colonies were counted. The same thing occurred with column IV, an average of 1 CFU/100 ml was counted for the filtration, but for plating 1 set op plates showed no presence of *E. coli* while on a second set 226 colonies were counted.

Table 4-6: Concentrations of total coliforms and E. coli for each stream and the log reduction in total coliforms (I: continuous sand filter, II: continuous GAC filter, III: discontinuous sand filter, IV: discontinuous GAC filter, UV: UV disinfected sample)

	Total coliforms (CFU/100 ml)	***E. coli* (CFU/100 ml)**	**Absolute logarithmic change total coliforms**
Influent	(2 ± 2)	(< 1 ± 0)	-
I	(< 1 ± 0)	(< 1 ± 0)	-
II	(1 ± 2) x 10^3	(1 ± 2) x 10^3	-
III	(4 ± 1) x 10^2	(4 ± 1) x 10^2	2.30

IV	(1 ± 1) x 10^4	(1 ± 1) x 10^4	1.15
UV	(1 ± 1) x 10^2	(1 ± 1) x 10^2	1.07

The UV-lamp was positioned after the GAC column (II) of the continuous set-up for disinfection of the effluent. A log reduction of 1.07 was obtained as shown in

Table 4-6. A study by Hamilton *et al.* (2019) compared different treatment strategies for the removal of micro-organisms in rainwater. The closest resembling case to this set-up was the filtration of harvested rainwater (from roofs, courtyards and roads) by a grit filter (0.7 m of expanded clay particles on top of 0.7 m gravel) followed by a UV disinfection step. A log reduction of 2.16 was obtained (0.19 log units by filtration only) for *E. coli*. Another study by Jordan *et al.* (2008) used an off-the-shelf point-of-use rainwater treatment train consisting of 20 µm polypropylene filter, a 5 µm polypropylene filter, an activated carbon impregnated paper cartridge filter and a UV-disinfection. A removal of 6 log units was obtained for *E. coli* (0.22 by filtration only) and 2 log units for total coliforms (0.21 by filtration only). When comparing the results of Table 4-6 with these studies, no removal was obtained by filtration and only 1.07 log units by using UV-irradiation. This leaves room for improvement which should enhance microbiological control in the filtration set-up.

When comparing the effluent concentrations with the maximum allowed concentrations of total coliforms (0 CFU/100 ml) in Flemish drinking water (see Table 2-5) it can be concluded that all the effluents are not conform with the legal limit. However, quality requirements concerning *E. coli* are also given for the reuse of reclaimed water in Spain in Table 2-7. By comparing the concentration of *E. coli* found in the UV-lamp effluent with these values, it can be concluded that this water can be used for every appliance with the exception of residential use (irrigation of gardens and supply to sanitary installations), use in cooling towers and condensers and aquifer recharge by direct injection. However more information on the microbiological quality is required e.g. the concentrations or presence of *Legionella spp*., *Taenia saginata* and *Taenia solium* as these are sometimes additional requirements.

4.3.4.3 Effects of shock chlorination on the microbiological quality and biofilm

After performing a chlorination (5 mg/l) after 13 000 BV for the continuous set-up, samples were taken again for the determination of the microbiological quality. As the continuous sand filter posed the largest problem with regards to biofilm, samples were only taken from this system. Not a single colony was observed in the undiluted samples after an incubation period of 72 hours, as shown in Table 4-7. The chlorination can therefore be considered successful in temporary removing the biofilm. The quality of the effluents conformed with the microbiological Flemish drinking water quality norms (see Table 2-5), the biological norms for the use of water in the Belgian food sector (Table 2-6) and could be used for every application mentioned in Table 2-7 for the reuse of reclaimed water in Spain. A test to determine the residual chlorine concentration in the water was not carried out.

Table 4-7: Microbiological quality of the continuous filtration after chlorination (I: continuous sand filter, II: continuous GAC filter)

	Total viable count (CFU/ml)	**Total coliforms (CFU/100 ml)**	***E. coli* (CFU/100 ml)**
Influent	< 1	< 1	< 1
I	< 1	< 1	< 1
II	< 1	< 1	< 1

4.4 Long-term discontinuous treatment

This set-up also consisted of a sand filter (III) followed by a GAC column (IV). This filtration set-up experienced no major problems caused by biofilm. Columns were never paused outside of their schedule and no overflowing of the columns happened. Regular backflushing kept the biofilm in control for the period of the experiment but a chlorination was nevertheless performed after 6000 BV.

4.4.1 Standard quality parameters of the discontinuous filtration

Figure 7-4 (supplementary materials) shows the changes in pH over the course of the experiment. The pH remained pretty stable throughout the experiment. After 100 BV, a small increase is possibly caused by the presence of alkaline groups in fresh activated carbon. After 7300 BV, the pH of the sand filter effluent (III) is slightly higher. This can be caused by residual chlorine (weak base) or biofilm particles. The average effluent pH of the continuous and discontinuous set-up was both 7.0 so no major difference between the effluents of the two systems was observed.

Figure 4-23 represents the evolution of the turbidity over the course of the experiment. A chlorination was performed after 6000 BV leading to a spike in turbidity. Especially the sand filter effluent after 6200 BV showed a spike with a turbidity of 5.57 NTU indicating a consistent microbiological presence in the sand filter. The GAC effluent at that bed volume also showed presence of biofilm as a value of 3.00 NTU was measured. In general no major differences can be determined between the continuous and discontinuous set-up.

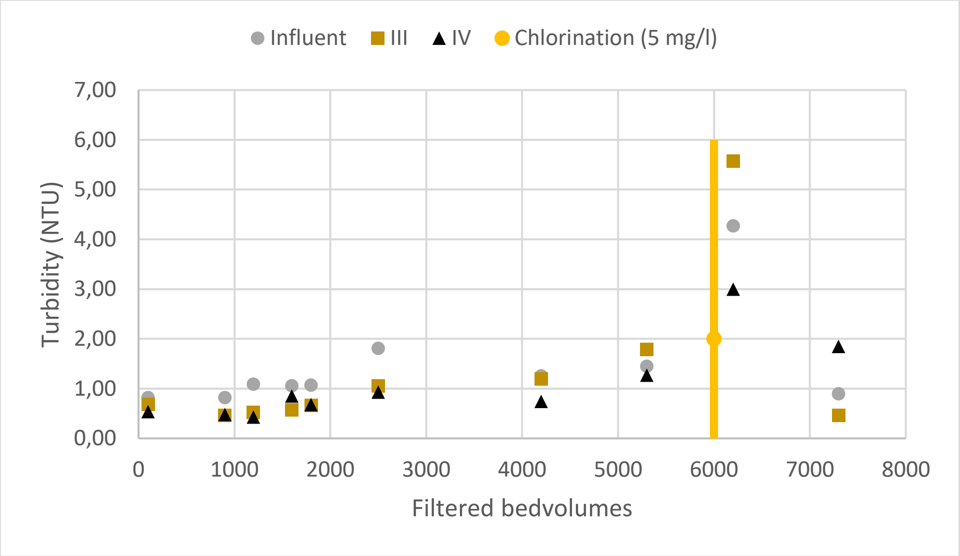


Figure 4-23: Evolution of turbidity over time during discontinuous filtration (yellow line = performed chlorination, III = sand filter, IV = GAC)

The evolution of electrical conductivity over time is presented in Figure 7-5 (supplementary materials). Almost no changes were observed during normal filtration. The effluent of the sand filter after 7300 BV showed an increased conductivity. This is caused by the presence of tap water which was used for backflushing right before sampling. The average electrical conductivity of tap water in 2019 in Flanders was between 300 and 800 µS/cm (VMM, 2019). All effluents conformed to Flemish drinking water quality norms concerning EC (presented in Table 2-5).

The removal of COD by filtration and adsorption in the discontinuous set-up is presented in Figure 4-24. When comparing the evolution of the COD between the continuous and discontinuous set-up, no big differences can be observed. Both set-ups showed elevated concentrations in influent, sand filter and GAC effluent immediately after chlorination. However, after 7300 BV, the concentration of the IV column is still higher than the influent. Accumulation of removed biofilm from the tubing and sand filter in the GAC column due to the lower number of bed volumes could be the reason.

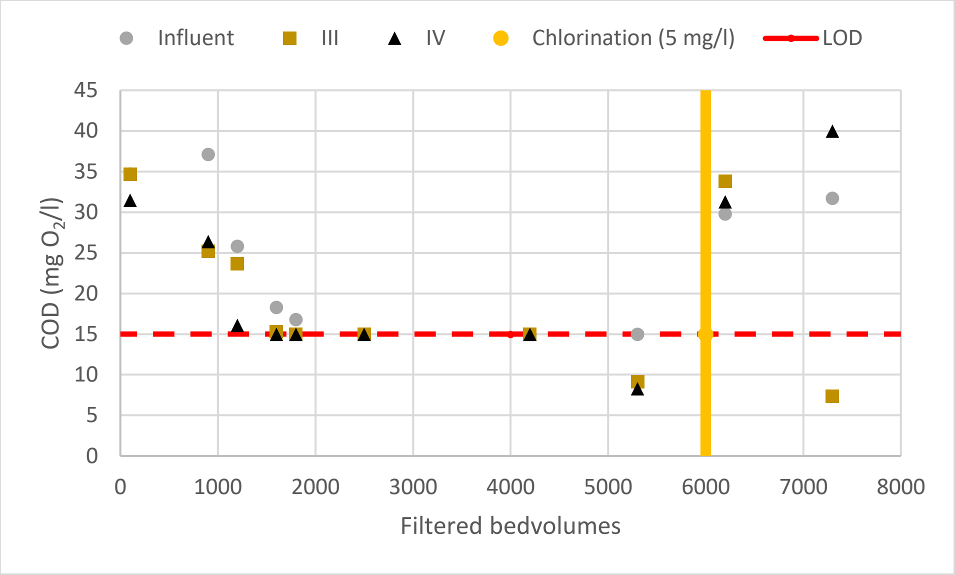


Figure 4-24: Changes in COD over time during discontinuous filtration (LOD = 15 and 1 mg O_2/l, yellow line = performed chlorination, III = sand filter, IV = GAC)

Figure 4-25 shows the evolution of absorbance at 254 nm during the operation. The decrease in organic matter after the first 2000 BV that can be observed in Figure 4-24 is not as visible in the UV-VIS-data. There, the organic content remains pretty stable so the uptake of methanol, which does not absorb at 254 nm, by the biofilm could have played a role in the COD decrease. The sand filter has the highest organic concentration after chlorination. This is probably caused by the presence of dead biofilm. After 7300 BV, the GAC has the highest organic content. The biofilm was probably flushed out of the sand filter and had accumulated in the activated carbon. The UV absorbance of the continuous (see Figure 4-9) and discontinuous filtration is nearly identical. UV_{254} can be considered the better option to quantify organic material as methanol is not included and only “natural” COD is measured. The degradation of methanol for biofilm growth also results in quickly meeting the LOD of the COD test kit.

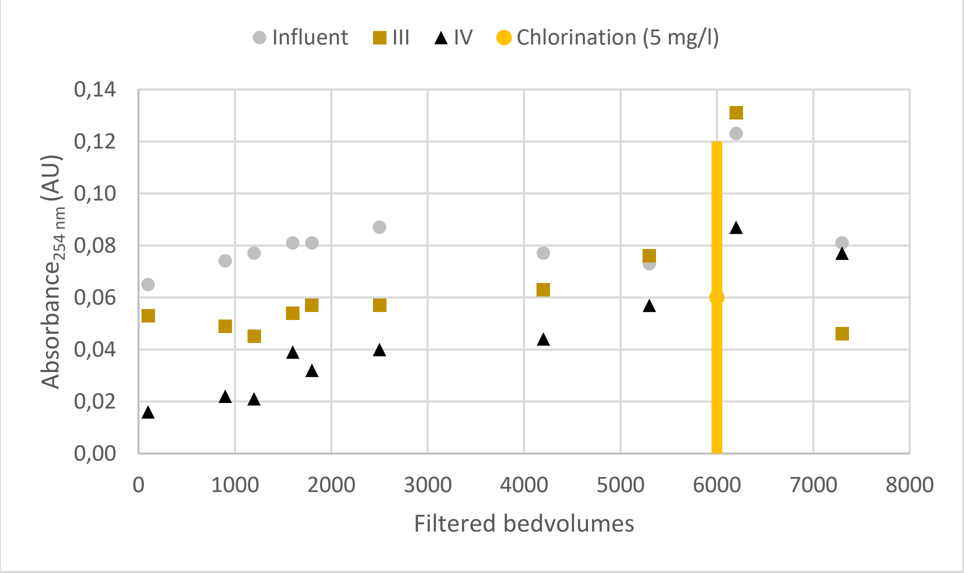


Figure 4-25: Changes in absorbance over time of UV-irradiation at 254 nm during discontinuous filtration (yellow line = performed chlorination, III = sand filter, IV = GAC)

Just as for the continuous filtration, a correlation between the UV-VIS and COD-removal was examined. The difference in operation did not lead to the determination of a significant correlation between the two variables. Insufficient data points (higher than the LOD of the COD test kit) and the presence of methanol were the two primary reasons for the difference between COD and UV_{254} removal.

The evolution of total nitrogen for the discontinuous filtration is presented in Figure 7-6 (supplementary materials). No major changes occurred during the filtration experiment. The chlorination after 6000 BV resulted in a release of nitrogen containing groups. The spike after 7300 BV for the GAC effluent (IV) was due to the presence of tap water. Both the continuous and discontinuous filtration effluents were within the same concentration range throughout the experiment.

4.4.2 Soluble metals of the discontinuous filtration

The removal of soluble metals by sand filtration during discontinuous filtration is shown in Figure 4-26 and Figure 4-27. Lead was not included in the dataset for the same reason as discussed in 4.3.2. One thing stands out when looking at the general evolution of removal of these metals during the experiment and that is the decrease in removal for all metals after 1200 BV. Backwashing data was consulted to try and pinpoint the origin of this change in behaviour. Backwashing happened less regular with the discontinuous sand filter than with the continuous sand filter so the effect of a backwash could have a bigger impact on the removal of metals. It would seem that longer periods with no backwashing increased the removal by sand filtration (see purple dotted lines in Figure 4-26 and Figure 4-27). The chlorination after 6000 BV seemed to have had very little effect on the removal of soluble metals by the sand filter. The sample points of iron were excluded from the data after 4200 BV as the influent concentrations were lower than the LOQ.

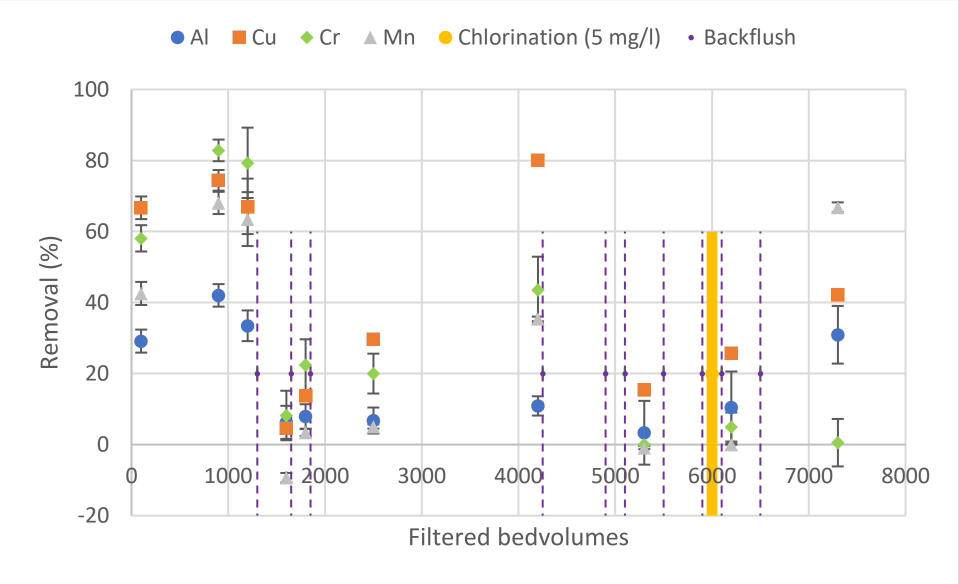


Figure 4-26: Removal of aluminium, copper, chromium and manganese by sand filtration during discontinuous filtration (purple dotted lines = backflushing of sand filter, yellow line = performed chlorination)

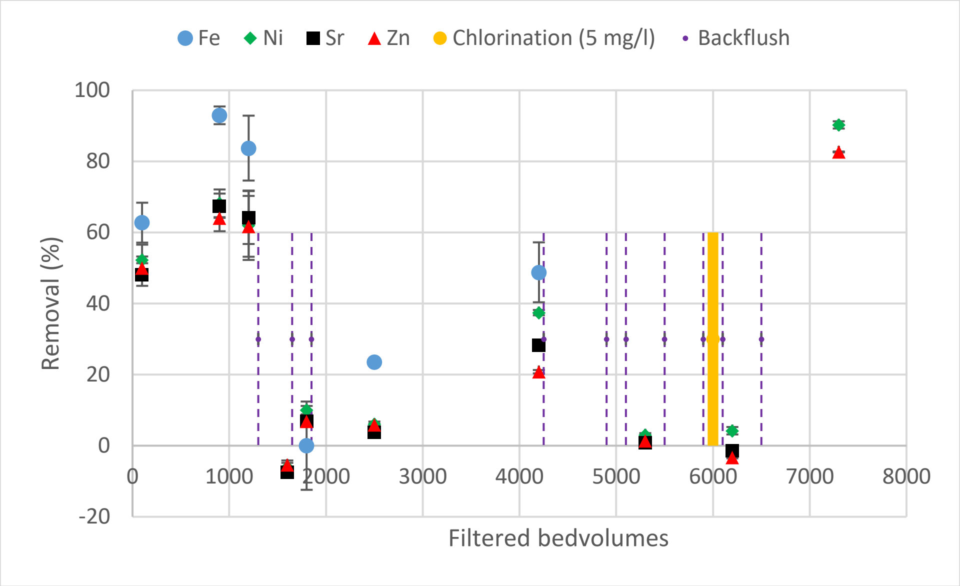


Figure 4-27: Removal of iron, nickel, strontium and zinc by sand filtration during discontinuous filtration (purple dotted lines = backflushing of the sand filter, yellow line = performed chlorination)

The removal of soluble metals from the influent by a combination of sand filtration and GAC during the discontinuous filtration is shown in Figure 4-28 and Figure 4-29. The combination of sand and GAC filtration leads to a more stable removal behaviour than the removal of the sand filter separately. This sequence of treatment can be considered a good option for a general removal. Copper is removed quite well during the experiment, not only by the sand filtration but also partially by the activated carbon. Aluminium showed almost no removal, especially by the GAC. Manganese has a fluctuating removal behaviour with serious leaching after 7300 BV. This is also observed for nickel, strontium and zinc. This behaviour in combination with the improved removal of organic micropollutants after 7300 BV (see 4.4.3) leads to the conclusion that the bonds of the metals with the activated carbon were broken and the adsorption locations on the GAC, previously occupied by metals, were now available to organic compounds which bond preferentially with GAC. This theory is based on the changes in concentrations for the last sampling point. The concentrations of the sand filter effluent after 7300 BV are lower than the influent concentrations but the GAC effluent all have a higher concentrations than the sand filter effluent. So leaching from GAC has occurred due to the performed chlorination. This is an important phenomenon that should be kept in mind when shock chlorinating a rainwater treatment set-up containing GAC. Thoroughly flushing the set-up and discharging this flush water may be the solution. Effluent concentrations can (temporary) be as high as twice the influent concentrations. However a shock chlorination with 5 mg/l NaOCl is quite intensive, disinfection of water with chlorine require concentrations of around 2 mg/l NaOCl (see 2.4.3.1).

Only 3 samples of the discontinuous set-up exceeded the maximum allowed concentration of nickel in drinking water after treatment with sand and GAC filtration (see Table 2-5). The rest of the metal concentrations were all under their respective legal limit for drinking water.

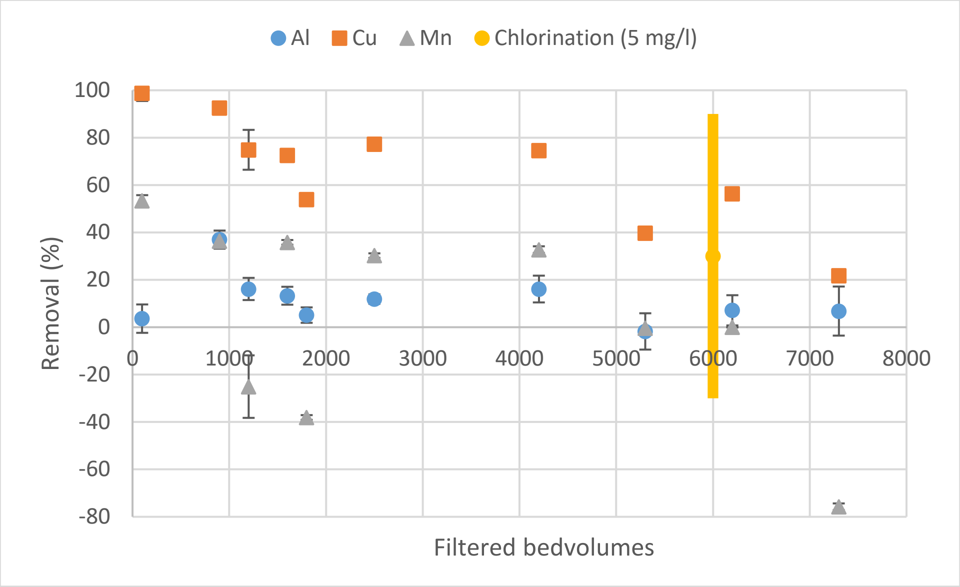


Figure 4-28: Removal of aluminium, copper and manganese over time by a combination of sand and GAC filtration during discontinuous filtration (yellow line = performed chlorination)

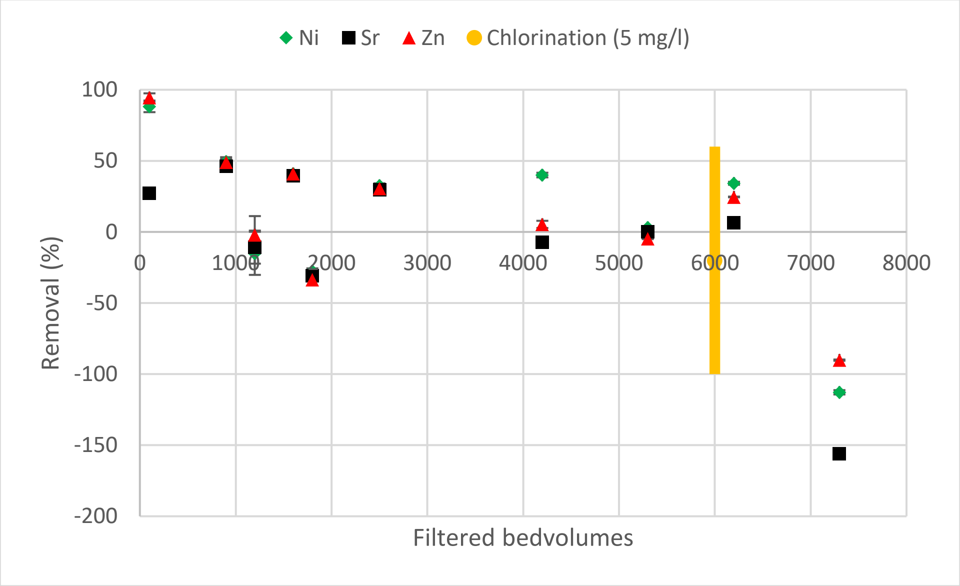


Figure 4-29: Removal of nickel, strontium and zinc over time by a combination of sand and GAC filtration during discontinuous filtration (yellow line = performed chlorination)

4.4.3 PAHs, phthalates and herbicide of the discontinuous filtration

The removal by sand filtration of organic micropollutants by discontinuous filtration is presented in Figure 4-30, Figure 4-31 and Figure 4-32. Just as with the continuous filtration, the PAHs are removed the most. Naphthalene is sometimes removed, the varying behaviour can be accredited to the low molecular weight of the compound. Diuron is almost not removed by sand filtration, DEHP shows a very fluctuating removal, as was the case in the continuous filtration, due to contamination, leaching, water solubility and adsorption phenomena (see 4.3.3). The chlorination seemed to have had an impact on the PAHs (with the exception of naphthalene) and diuron which show a small amount of leaching immediately after the disinfection. The removals of all aforementioned compounds with the exception of diuron exhibited an improved removal by sand filtration.

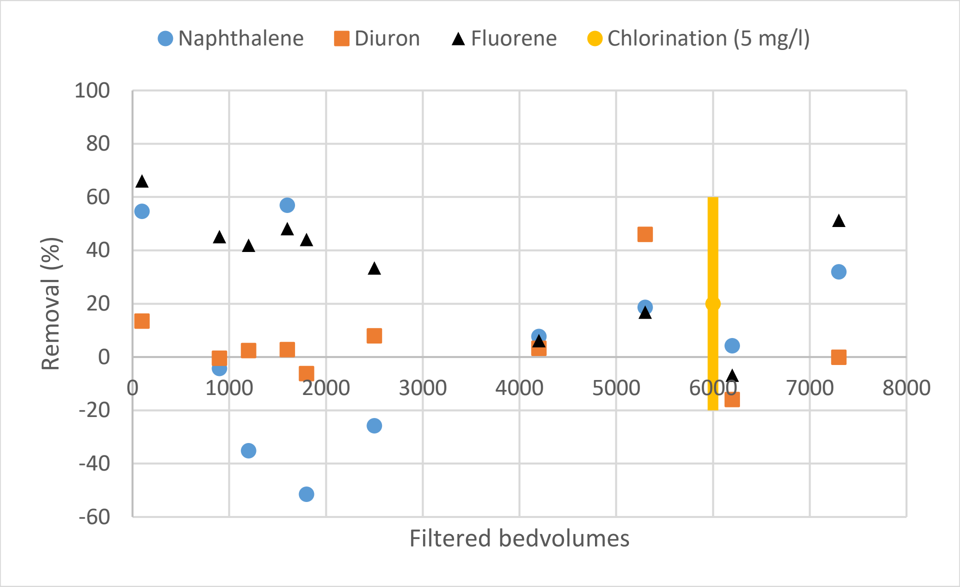


Figure 4-30: Removal of naphthalene, diuron and fluorene by sand filtration during discontinuous filtration (yellow line = performed chlorination)

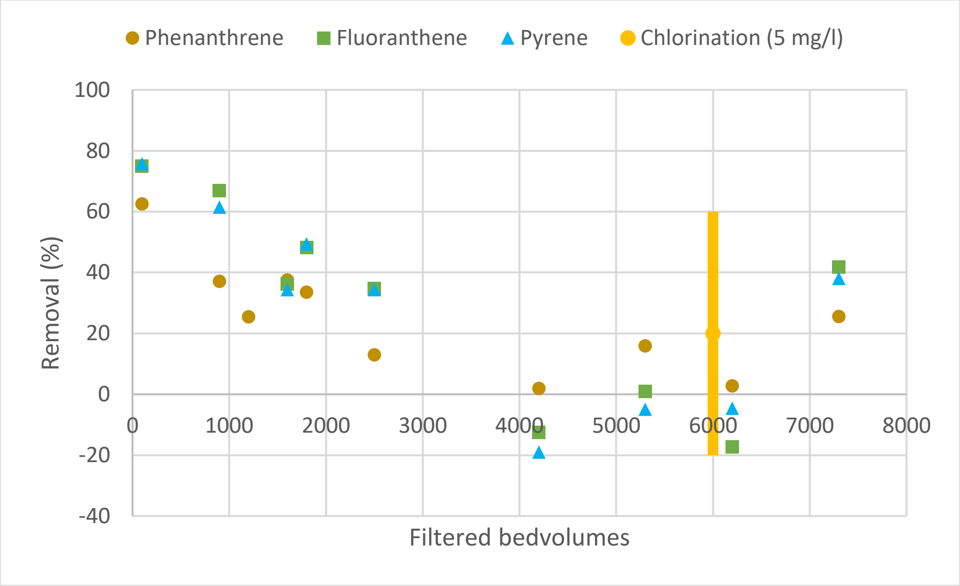


Figure 4-31: Removal of phenanthrene, fluoranthene and pyrene by sand filtration during discontinuous filtration (yellow line = performed chlorination)

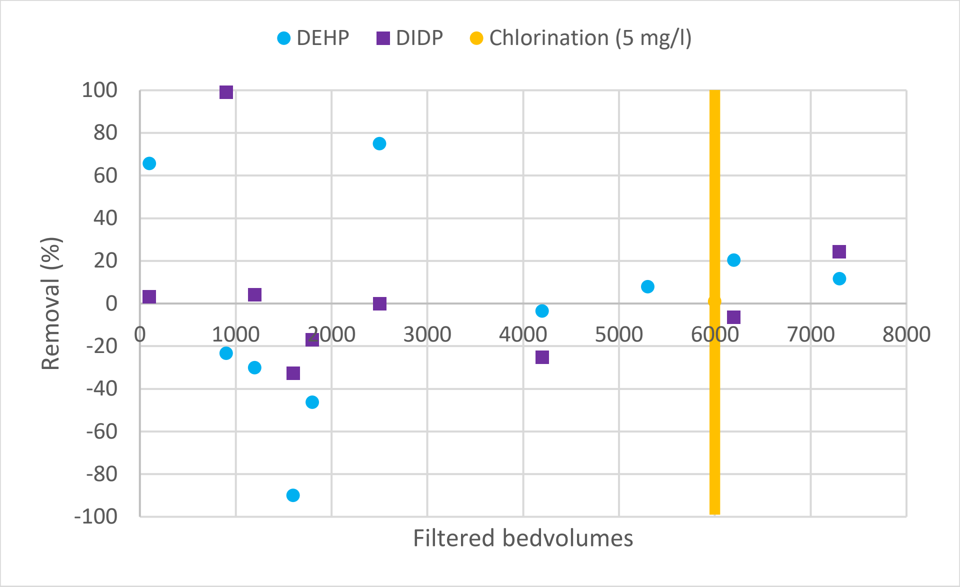


Figure 4-32: Removal of DEHP and DIDP by sand filtration during discontinuous filtration (yellow line = performed chlorination)

When the removal of organic compounds by GAC is taken into account, the evolution over time for each organic pollutant are shown in Figure 4-33, Figure 4-34 and Figure 4-35. Some observations can be made when comparing the removal efficiency of the combination of treatment steps with the efficiency of sand filtration alone. The removal of naphthalene is better with GAC present but removal is still very fluctuating. Diuron is almost exclusively adsorbed by GAC and the removal of the rest of the PAHs is improved by 20 to 30%. The removal of phthalates did not improve by adding activated carbon to the treatment.

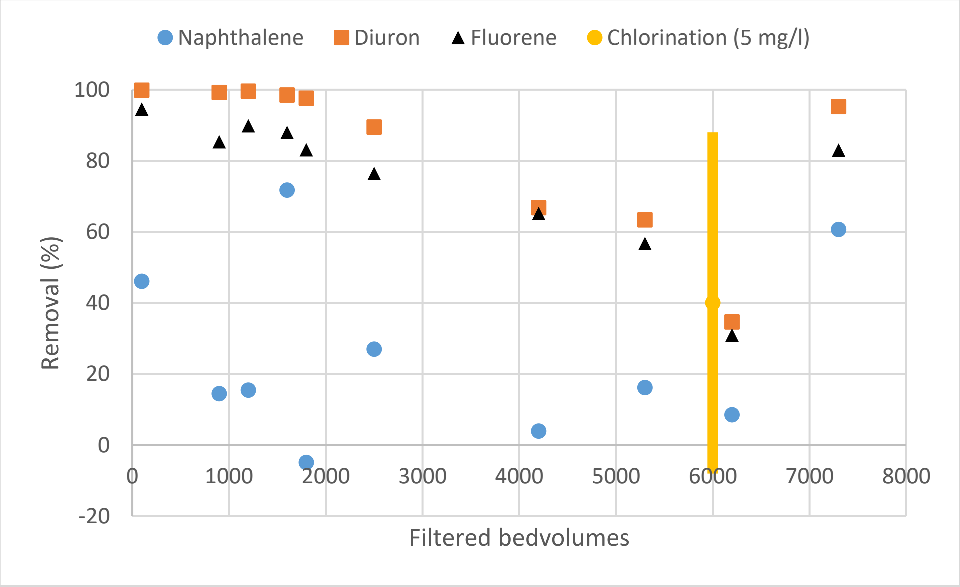


Figure 4-33: Removal of naphthalene, diuron and fluorene over time by sand filtration and GAC during discontinuous filtration (yellow line = performed chlorination)

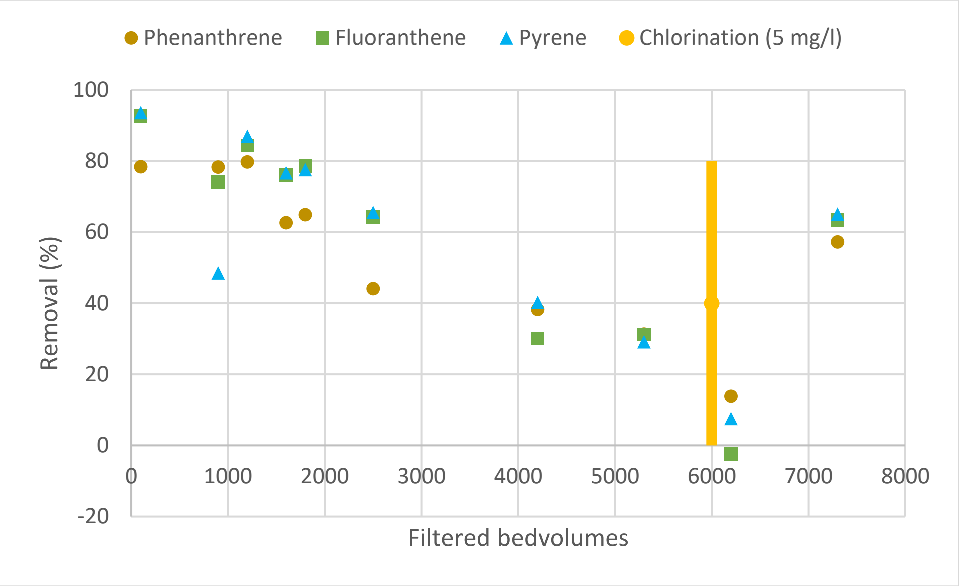


Figure 4-34: Removal of phenanthrene, fluoranthene and pyrene over time by sand filtration and GAC during discontinuous filtration (yellow line = performed chlorination)

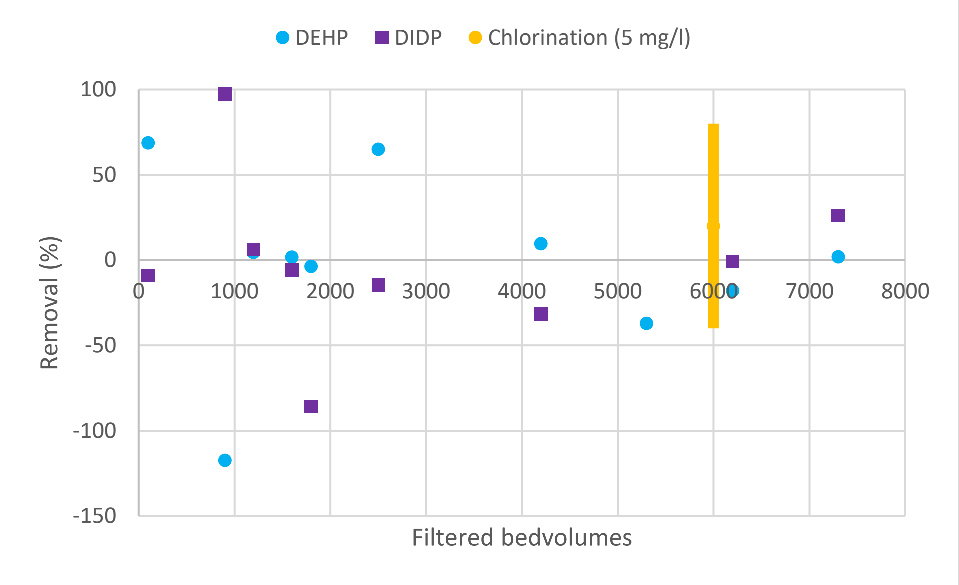


Figure 4-35: Removal of DEHP and DIDP over time by sand filtration and GAC during discontinuous filtration (yellow line = performed chlorination)

A summary overview of the removal by sand filtration and GAC treatment separately along with the effluent concentration ranges for each compounds is given in

Compound	**Maximum removal by sand filter (%)**	**Median sand filter effluent concentration (ng/l)**	**Removal range GAC (%)**	**Median GAC filter effluent concentration (ng/l)**	**Flemish drinking water quality norms for organics (ng/l)**
Naphthalene	57	81	-19 – 43	63	100 (total PAHs)
Diuron	47	9231	33 – 99	830*	500 (total pesticides) 100 (individual)
Fluorene	66	60	36 – 83	9*	100 (total PAHs)
Phenanthrene	67	78	7 – 71	13*	100 (total PAHs)
Fluoranthene	74	55	6 – 83	3*	100 (total PAHs)

Pyrene	94	61	10 – 83	4^*	100 (total PAHs)
DEHP	75	65	-72 – 49	62	8000 (guideline WHO)
DIDP	34	37 389	-44 – 34	19 796	-

. The sum of the concentration of every PAH was calculated for each sample to determine whether the effluents conformed to the Flemish drinking water norm (see Table 2-5). Five samples of the discontinuous filtration had a total PAHs concentration higher than 100 ng/l. The legal limit of diuron in drinking water is 100 ng/l and 4 samples of the discontinuous filtration exceeded this after treatment. Only samples with a concentration higher than the LOD for diuron (830 ng/l) were considered. The WHO guideline for DEHP is 8 µg/l and only one sample contained more than 8 µg/l DEHP in the GAC effluent from the discontinuous filtration.

Table 4-8: Summary of the removal by combination of sand filtration and GAC of the target organic compounds and their median effluent concentration for the discontinuous filtration (= limit of detection)*

Compound	**Maximum removal by sand filter (%)**	**Median sand filter effluent concentration (ng/l)**	**Removal range GAC (%)**	**Median GAC filter effluent concentration (ng/l)**	**Flemish drinking water quality norms for organics (ng/l)**
Naphthalene	57	81	-19 – 43	63	100 (total PAHs)
Diuron	47	9231	33 – 99	830^*	500 (total pesticides) 100 (individual)
Fluorene	66	60	36 – 83	9^*	100 (total PAHs)
Phenanthrene	67	78	7 – 71	13^*	100 (total PAHs)
Fluoranthene	74	55	6 – 83	3^*	100 (total PAHs)
Pyrene	94	61	10 – 83	4^*	100 (total PAHs)
DEHP	75	65	-72 – 49	62	8000 (guideline WHO)
DIDP	34	37 389	-44 – 34	19 796	-

4.5 Determination of the experimental breakthrough of GAC

The expected breakthrough can be calculated by plotting the removal of metals and the number of bed volumes. Breakthrough is expected to occur when the regression line intersects with the abscissa.

4.5.1 Continuous treatment

The experimental breakthrough was determined for both metals and organic compounds. Only the breakthrough of copper could be calculated as the other metals exhibited too much of a fluctuating removal behaviour. The last two sample points were not included because the chlorination changed the removal behaviour of the GAC. The expected breakthrough of copper can be deducted from Figure 4-36 and should occur after 12 422 BV. However, the Pearson coefficient is only 0.26 which indicates a weak correlation. The predicted breakthrough of metals is less relevant than the breakthrough of organic pollutants as ion exchange reactions influence the end results but it can still indicate a general end-of-use time.

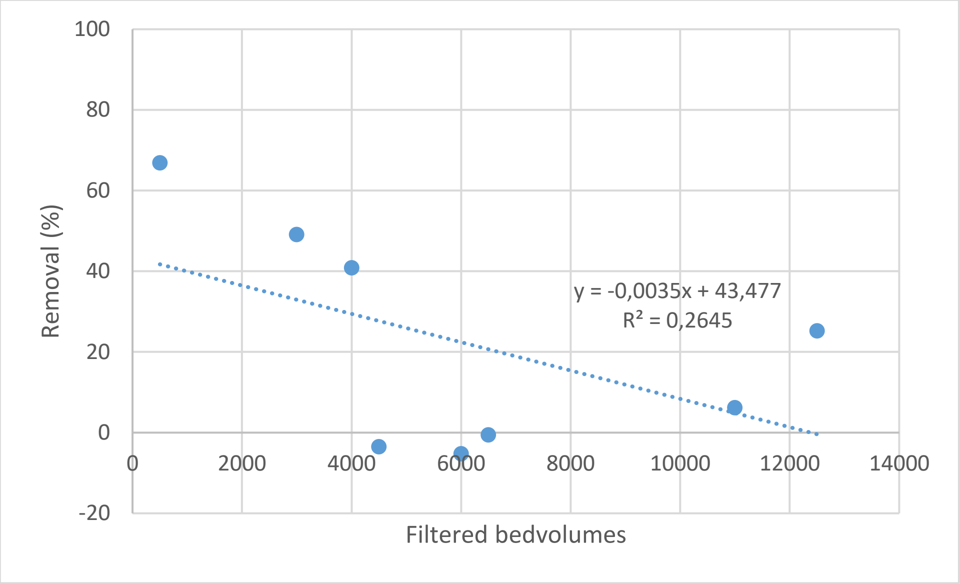


Figure 4-36: Calculation of the expected breakthrough of GAC regarding copper during continuous filtration

The expected breakthrough of organic compounds showed a more linear model than the metals. Only the removal by GAC was considered for this calculation, the data of the sand filter was not incorporated. Also, only the removal data before the chlorination was considered as the chlorination interfered with normal removal operation of the GAC. Table 4-9 summarizes the expected breakthroughs for every compounds and also gives the time in hours until the expected breakthrough is reached if filtration is continuous.

Table 4-9: Experimental breakthrough of GAC for each organic compound used in this experiment during continuous filtration (ND = not determinable)

Compound	**Expected breakthrough (BV)**	**Expected breakthrough (weeks)**	**R²-value**
Naphthalene	16 004	4	0.18
Diuron	29 551	8	0.92
Fluorene	21 375	6	0.91
Phenanthrene	15 708	4	0.55
Fluoranthene	17 540	5	0.80
Pyrene	21 268	6	0.64
DEHP	ND	ND	ND
DIDP	6242	2	0.77

4.5.2 Discontinuous treatment

The breakthrough for each metal was again determined the same way it was calculated for the continuous treatment. The breakthrough could only be predicted for copper: it would happen after 5309 BV ($R^2 = 0.48$). This is equal to 5 weeks of a discontinuous filtration (6 hours/day during the week and 9 hours/day during the weekend).

The breakthrough bed volume of the organic micropollutants can be translated to a breakthrough time, which is more practical, if the exact bed volume and the flow rate of the pump is known. One week equals 48 hours of discontinuous filtration. The results are presented in Table 4-10.

Table 4-10: Experimental breakthrough of GAC for each organic compound used in the experiment during discontinuous filtration

Compound	**Expected breakthrough (BV)**	**Expected breakthrough (weeks)**	**R²**
Naphthalene	6628	7	0.53
Diuron	9133	9	0.85
Fluorene	13 696	14	0.82
Phenanthrene	9361	10	0.25
Fluoranthene	9138	9	0.75
Pyrene	12 041	12	0.41
DEHP	1224	1	0.02
DIDP	1861	2	0.36

Many manufactures of commercial GAC cartridges used in household RWH systems recommend changing the cartridge every 6 months (Honeywell, 2021; Van Marcke, 2021). However, the goal in commercial systems is mainly to remove odour and taste components in water and secondly to withhold and protect the user from a coincidental pollutant load (Carthwright, 2007). When treating rainwater for high grade application and when an enhanced removal of metal of organic micropollutants is desired, a more regularly replacement of the GAC is advised as metals can occur in relatively high concentrations in roof runoff due to leaching and dry deposition (see 2.1.2 and 2.1.4).

The amount of GAC used in a commercial cartridge can be estimated by measuring the dimensions of a cartridge. A GAC cartridge of Honeywell® will be used in this example to estimate the breakthrough when using a commercial system to treat rainwater to fit-for-use applications. The cylindrical cartridge has a height of 255 mm and a layer of 22 mm of compressed activated carbon (pore size 10 µm), it can therefore be calculated that the total volume of GAC is 0.881 ml (Honeywell, 2021). This is around eight times the volume of the GAC bed volume of column II and IV used in the long term filtration tests (see 3.3.2).

The expected breakthrough of a commercial GAC cartridge for each micropollutant can be determined by extrapolating the available data from the long term filtration tests. If the breakthrough of copper during continuous filtration occurred after 12 422 bed volumes (see above) with a volume of 0.108 ml GAC in the column, then the breakthrough using 0.881 l of GAC in a commercial cartridge can be expected to happen around bed volume 101 331 or after 5 years of continuous operation if the same operational conditions apply. The breakthrough of each micropollutant, both during continuous and discontinuous filtration, can be determined this way. The results for the continuous filtration are presented in Table 4-11 and the results for the discontinuous filtration are included in Table 4-12.

Table 4-11: Calculated breakthrough for each micropollutant using a Honeywell® commercial GAC cartridge for continuous, long term filtration

Continuous filtration	Compound	Breakthrough BV	Breakthrough time (years)
	Copper	101 331	5
	Naphthalene	130 551	6
	Diuron	241 060	11
	Fluorene	174 365	8
	Phenanthrene	128 137	6
	Fluoranthene	143 081	7
	Pyrene	173 492	8
	DIDP	50 919	2

Furthermore, the average potential rainwater use (after treatment) per day per person is 66 l according to one of the largest water suppliers in Belgium, *De Watergroep*. This includes rainwater for showers (24 l), cleaning (6 l) and other applications (36 l) (De Watergroep, 2018). A family of three people, which was also used to determine the filtration schedule for the discontinuous set-up (see 3.3.1), would thus consume 198 l of rainwater per day. This is equal to 225 bed volumes that would pass through the GAC cartridge every day. It is possible to determine the breakthrough of a commercial GAC cartridge for each micropollutant based on the consumption pattern of a family discussed above. Only discontinuous filtration is regarded as this is the most realistic treatment model for a household (48 hours a week of filtration). For example, the breakthrough of copper using a commercial cartridge in the long term tests is to be expected around bed volume 39 638 but if 225 bed volumes pass through the cartridge every day, then the breakthrough would occur after around 6 months. All these results are included in Table 4-12 as well. So for the phthalates, only 1-2 months of filtration are possible before the cartridge needs to be replaced. However, the data for the phthalates obtained from the filtration tests was very fluctuating due to several phenomena (adsorption, leaching, increased solubility etc.). The obtained data for the PAHs and diuron was more consistent throughout the experiment so the breakthrough times for these components can be considered more accurate. It can be concluded that a GAC cartridge should be replaced after around six months to guarantee a decent water quality. The commercial cartridge should be replaced yearly if only the organic micropollutants are considered.

An important remark here is that this scenario is to be considered the worst case. Spiked influent was used to determine the breakthrough of each component. Not every compound will be present in the runoff in real life scenarios and the concentration will not always be this high and will vary more. The expected breakthrough will therefore most certainly take place later than calculated here. The breakthrough is also directly correlated with the amount of activated carbon present in the cartridge so implementing a bigger one will increase the cycle time of the treatment system.

Table 4-12: Calculated breakthrough of each micropollutant using a Honeywell® commercial GAC cartridge for both discontinuous, long term filtration and treatment of rainwater on a household level

Discontinuous filtration	Compound	Breakthrough BV	Breakthrough time (years)	Breakthrough on household level (months)
	Copper	39 638	6	6
	Naphthalene	49 485	8	8
	Diuron	68 188	11	11
	Fluorene	104 294	17	17
	Phenanthrene	69 890	11	11
	Fluoranthene	68 225	11	11
	Pyrene	89 899	14	14
	DEHP	9139	1	1
	DIDP	13 984	2	2

4.6 Laundry company case

The goal of this case was to provide water with a quality that was in between drinking water quality and fresh roof runoff. As the long term filtration tests show, it is not evident to produce potable water at any time with a combination of sand and GAC filtration. Specific requirements (see Table 4-13) regarding standard water quality parameters and soluble metal concentrations had to be met to ensure smooth operation of the laundry machines. Influent was prepared according to 3.4.2 and a filtration train consisting of a lava rock filter, sand filter and GAC was used to treat the water. Samples were taken in between to determine the efficiency of each treatment step. The samples were afterwards disinfected for microbiological control.

Table 4-13: Quality requirements for process water used in the laundry industry (same as Table 3-4, repeated for convenience)

Parameter	Requirements for laundry industry process water
Conductivity (µS/cm)	< 1800 but preferably as low as possible
pH	Between 6.5 and 8.5
Hardness (mg/l CaO)	0 - 40
Metals (mg/l)	Fe < 0.1 Cu < 0.05 Mn < 0.05
Total viable count	< 100 CFU/ml
Total coliforms	0 CFU/ml
Other requirements	Clear water with no suspended particles No odour

4.6.1 Overview of individual streams before treatment

As discussed in 3.4.1, the goal in the near future was to use the runoff of three roofs to partially substitute tap water which is currently used as washing water. Each stream of water was therefore analysed to be able to explain certain characteristics of the combined influent. The result of this screening is presented in Table 4-14. This includes the runoff of each roof and also a sample of the buffer which is water from the roof of neighbour 2 that was treated by sand filtration and chlorination. This water had already been in the buffer for quite some time (recirculating) while the other samples were quite fresh as the water containers, where samples were taken, were relatively small (high refresh rate).

Table 4-14: Results of the analyses for each separate stream (: < LOD)*

Parameter	**Laundry company**	**Neighbour 1 (commercial)**	**Neighbour 2 (industrial)**	**Buffer**
pH	10.1 ± 0.1	7.5 ± 0.1	7.4 ± 0.1	6.4 ± 0.1
EC (µS/cm)	84.7 ± 0.1	124.5 ± 0.1	854.0 ± 0.4	250 ± 0.6
Turbidity (NTU)	0.95	0.85	65.40	2.84
COD (mg O_2/l)	8.2	31.1	14.2	15.6
TN (mg T_b/l)	1.72	4.99	2.99	<1.00*
TP (mg PO_4/l)	0.187	0.135	0.464	<0.050*
Ca (µg/l)	9514 ± 3	14 304 ± 3	38 153 ± 6	44 138 ± 5
Cu (µg/l)	3.5 ± 0.1	5.7 ± 0.1	<0.8 ± 0.2*	442.9 ± 0.5
Fe (µg/l)	<1.1 ± 0.1*	76.9 ± 0.2	<1.1 ± 0.2*	32.8 ± 0.2
Mn (µg/l)	8.7 ± 0.4	8 ± 0	1075.7 ± 0.7	214.7 ± 0.3
Total viable count (22°C) (CFU/ml)	(2.1 ± 0.6) x 10^3	(3.8 ± 0.5) x 10^5	(4.1 ± 0.8) x 10^3	(2.4 ± 0.9) x 10^3
Total coliforms (37°C) (CFU/100 ml)	(2 ± 1) x 10^3	(2 ± 1) x 10^4	(2 ± 1) x 10^4	(<1 ± 0)
E. coli (CFU/100 ml)	(5 ± 3) x 10	(4.9 ± 0.2) x 10^3	(9.5 ± 0.7) x 10^2	(<1 ± 0)

The pH of the laundry company is quite high in comparison with the other streams. This can be caused by leaching of carbonate from the concrete tank in which the water is stored. The pH-value of this water is too high to be used directly so a pH-correction by treatment is necessary (see Table 4-14). The EC is significantly higher for the runoff of the industrial building. This could indicate a high concentration of ions in the sample. Turbidity is high for runoff from the industrial building and very low for the samples of the laundry company and neighbour 1. The sample location of the industrial building was not very clean and contamination could have happened when taking the sample. The container was also quite small so agitation of the sediment could have happened easily. The samples of the company and commercial building were taken from closed-off underground storage tanks. These act as settlers which explains the low turbidity values. The COD and TN concentrations are the highest for the water of the commercial building. The storage tank that collected this water was located under a hen house so nutrients and organic material could have ended up in the water. Total phosphorus levels were within the expected range of roof runoff, the concentration in the buffer was lower than the LOD so the sand filtration may have removed TP from the industrial roof runoff.

The hardness of the water in the buffer is too high to be used immediately for laundry applications (calcium > 40 mg/l, 10 FD, see Table 4-13). The concentration of copper and manganese in the buffer and the concentration of manganese in the runoff of neighbour 2 are the only ones which are not within the limits and are higher than the other samples. This may indicate a leaching problem or the occurrence of a contamination in the buffer circuit and from the roofing material of neighbour 2. The treatment should reduce the manganese concentrations in the industrial runoff and buffer 4 or 20 times respectively to be able to be used in the washing process. Manganese usually ends up in rainwater through deposition related to traffic, soil related sources and raw materials of the steel industry. Historical pollution of older industries in the neighbourhood could be the source. Contamination of the runoff with water from the nearby river the *Dender* could have played a part as the sampling point of the industrial building was located very close to the river and was on the same height as the level of the river. However, the average concentration of soluble manganese measured at the closest analysing location of the river was 80 ± 60 µg/l (VMM, 2021a). All samples met the requirements for the iron concentrations, two samples had concentrations lower than the LOD. For the other requirements, namely the clearness and odour of the water, only the water of the industrial roof did not conform and clearly had odour problems.

Total viable count (22°C) was determined for samples of each stream which made up the influent. Only one sample (that of the commercial building) had a clear pollution of micro-organisms. The reason for this is the location of the storage tank where the sample was taken. It was located under a hen house. Leaching of contaminated faeces into the tank caused the high concentration of TN and COD in the sample. The other samples were all in the 10^3 CFU/ml concentration range. These concentrations are within the limits found in literature for roof runoff (see Table 2-4). The only remark that can be made is that the sample of the buffer still contained (2.4 ± 0.9) x 10^3 CFU/ml. This water is continuously disinfected by a chlorine tablet and is already used in test projects in the company. The requirement however concerning the total viable count are that the count should be lower than 100 CFU/ml (see Table 4-13) which is clearly not met under the present circumstances. Additional or increased disinfection should therefore be considered for future projects.

The same samples that were collected after each filtration test and who were analysed for total viable count were also used to determine the concentration of total coliforms and *E. coli* at 37°C. The maximum allowed concentration was 0 CFU/ml according to Table 4-13. Two techniques were used: water filtration and plating (see 3.6.4). The highest concentration was measured in the samples of neighbour 1. This is caused by the presence of a hen house. Samples from the neighbour 2 also contained a high concentration of total coliforms. When comparing these concentrations with values found in the literature summarized in Table 2-4, it can be concluded that the concentrations of total coliforms for both these samples are higher but the concentration of *E. coli* is within the limits. Runoff from the laundry company itself had the lowest concentration and the water of the buffer was completely free of coliforms due to the performed chlorination. Adequate disinfection will be necessary as the influent is made up of the runoff of the laundry company and its two neighbours.

4.6.2 Standard water quality parameters after treatment

The changes in pH are shown in Figure 4-37. The lava rock and sand filter did not have a major impact on the pH level of the effluents, the GAC did increase the pH but this is most likely a temporary phenomenon as fresh GAC leaches alkaline groups from the carbon during the first 500 BV (Desotec, 2021). The complete treatment train showed an even more moderate increase in the effluent. The pH should maximally be 8.5 (see Table 4-13), only the effluent of the sand filter + GAC had a higher pH-value (pH = 8.81). The pH of all samples was increased by their respective treatment and along with the high pH value of the runoff of the laundry company, this has a negative impact on the total pH of the influent. Further research should be conducted to determine whether the pH increase was a temporary or a recurring phenomenon. Using a different mixing ratio, where less runoff from the laundry company is added, could lower the total pH.

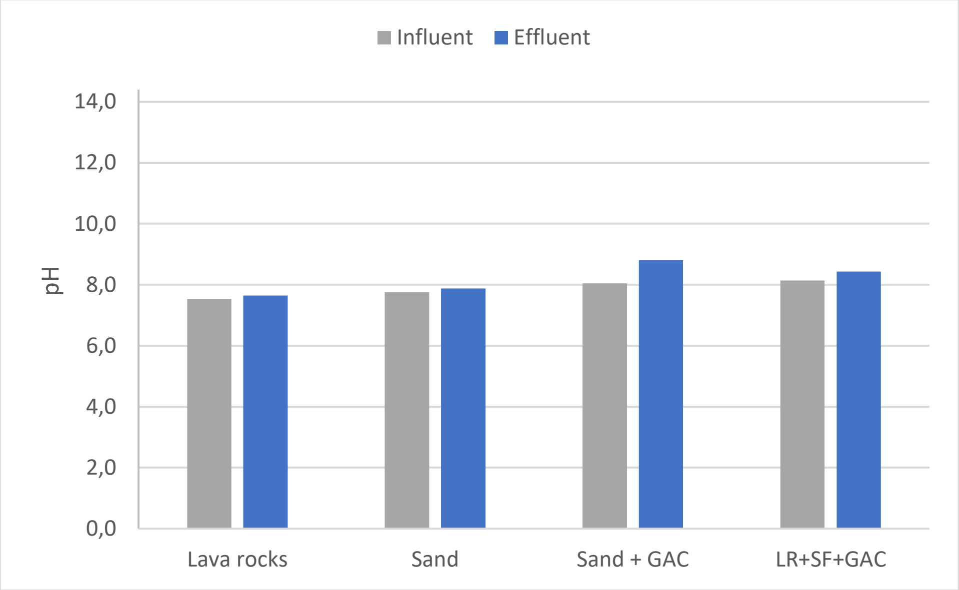


Figure 4-37: Changes in pH for each treatment of the laundry case

The changes in electrical conductivity are presented in Figure 4-38. All the influents and effluents are much lower than the maximum EC-value of 1800 µS/cm. The lava rocks and sand filter showed minimal removal of ions from the influents, the GAC was able to lower the EC by 10 µS/cm. The complete treatment train was able to decrease the conductivity by 30 µS/cm. The latter was thus the best option as the lowest possible conductivity is desired.

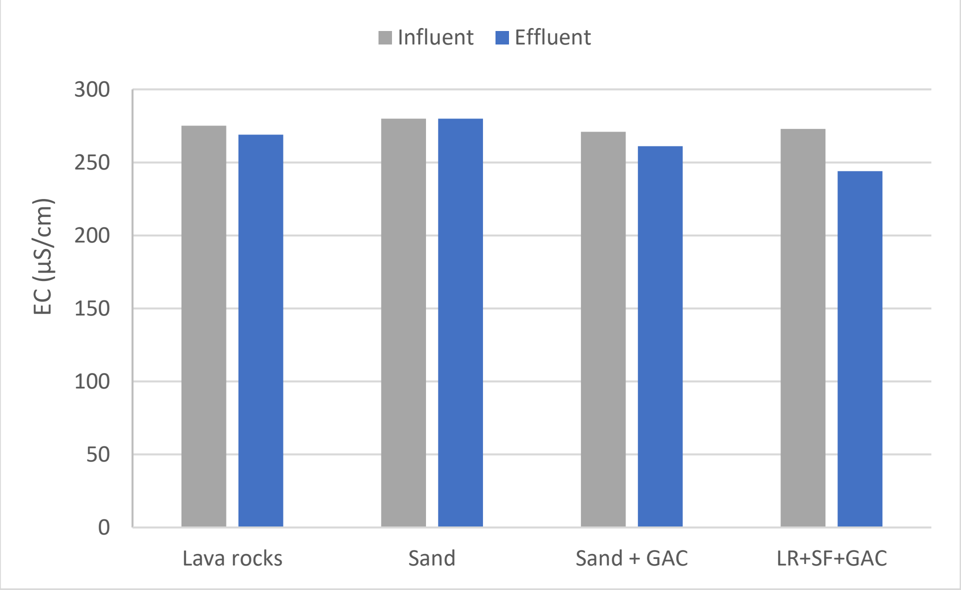


Figure 4-38: Changes in electrical conductivity for each treatment of the laundry case

The impact of each treatment step on the turbidity of the influent is shown in Figure 4-39. Because the tests lasted over a period of two weeks and only 1 volume of influent was prepared, the turbidity of the influents shows a decrease as the influent remained in the container for the duration of the experiment. During that time, the container acted as a settler. All treatment steps were able to lower the turbidity, however the lava rock and sand filter only to a certain amount. The combination of sand filtration with GAC or the complete treatment train were able to halve the turbidity of the influent. It can be concluded that the last two options produce the best results for this parameter and that the introduction of a settler, which is present in the current filtration system of the laundry company (see Figure 3-3), has a positive impact on the water quality to remove suspended solids.

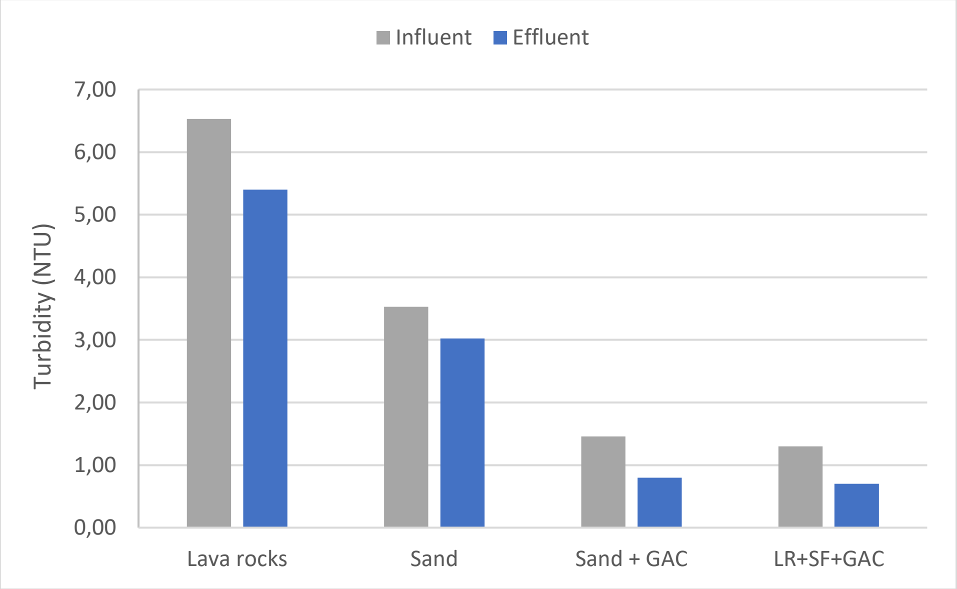


Figure 4-39: Changes in turbidity for each treatment of the laundry case

4.6.3 Removal of soluble metals after treatment

The results of the filtration test regarding the copper concentration are presented in Figure 4-40. All concentrations, of both the influents and effluents, were below the imposed limit of 50 µg/l. Filtration by a lava rock filter or by a combination of sand filtration and GAC gave the best results. The sand filter separately seemed to have increased the copper concentration. The same can be said about the complete treatment train: filtration with sand and GAC removed copper efficiently (to even below the LOQ) but when a lava rock filter was introduced, which also removed around half of the copper from its influent, the concentration in the effluent of the total train increased.

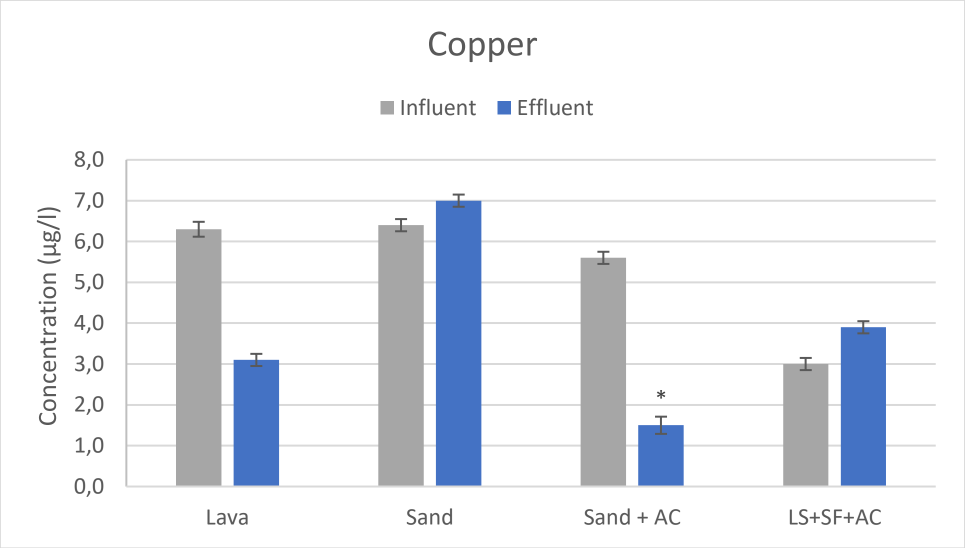


Figure 4-40: Effect of each treatment step on the copper concentration of the influent for the laundry case (= concentration copper < LOQ, quality requirement: < 50 µg/l)*

The effect of each treatment step on the concentration of iron in the influents is given in Figure 4-41. All samples were lower than the requested concentration of 100 µg/l. All the effluents were below the LOQ of iron. The high concentration of the influent of the sand + GAC filtration is unusual. No extra iron was spiked and leaching of iron from the plastic storage container is highly unlikely, so only a measuring error could have happened. The influent concentration of the total treatment train was below the LOQ because sedimentation of iron took place in the container over time as discussed in 4.1.1. The biggest contribution of iron comes from the runoff of the industrial building (see Table 4-14).

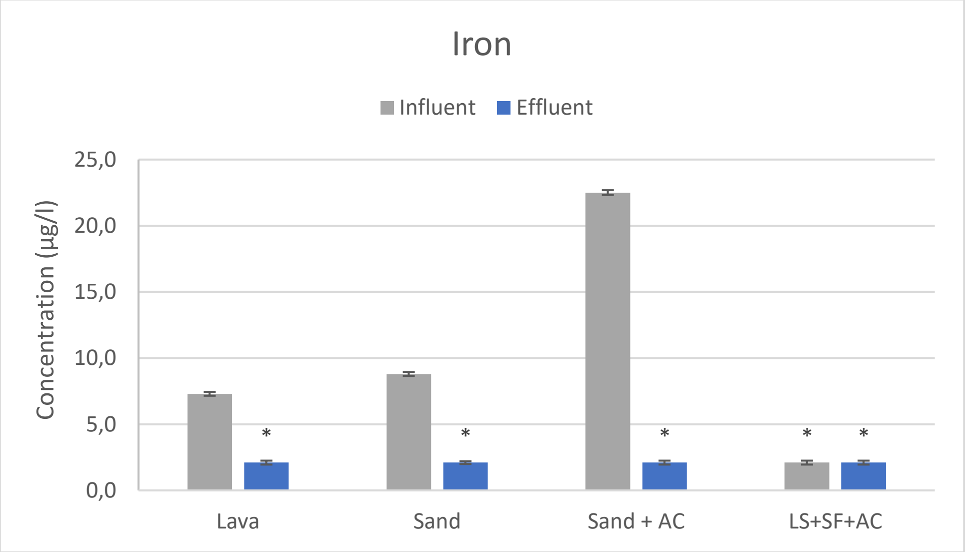


Figure 4-41: Effect of each treatment step on the iron concentration of the influent for the laundry case (= concentration iron < LOQ, quality requirement: < 100 µg/l)*

The concentrations of manganese in the effluents were not low enough to be used as washing water as 50 µg/l was the proposed limit. As shown in Figure 4-42, the lowest obtained concentration was 227.2 µg/l for the total treatment train. Both the lava rock and sand filter were able to more than halve the manganese concentration in the influent. The introduction of activated carbon in the treatment train showed an even better removal. The total treatment train can therefore be considered the best option. However to reach the limit of 50 µg/l, other techniques such as ion exchange might be necessary as the influent concentration is just too high. The concentration of manganese in the influents also decrease as the prepared influent remained in the container for some time. Sedimentation of manganese to the bottom of the container is the reason for the decrease in influent concentration (Niemiec, Arasimowicz and Wiśniowska-Kielian, 2012). The highest contribution of manganese to the influent comes from the industrial runoff. Comparing this concentration (1075.7 µg/l, see Table 4-14) with values found in the literature (c_{max} = 890 µg/l, c_{mean} = 24 µg/l, see Table 2-2) it becomes clear that this is an exceptionally high concentration. Manganese usually ends up in rainwater through deposition related to traffic, soil related sources and raw materials of the steel industry. Historical pollution of older industries in the neighbourhood could be the source. Contamination of the runoff with water from the nearby river the *Dender* could have played a part as the sampling point of the industrial building was located very close to the river and was on the same height as the level of the river. However, the average concentration of soluble manganese measured at the closest analysing location of the river was 80 ± 60 µg/l (VMM, 2021a).

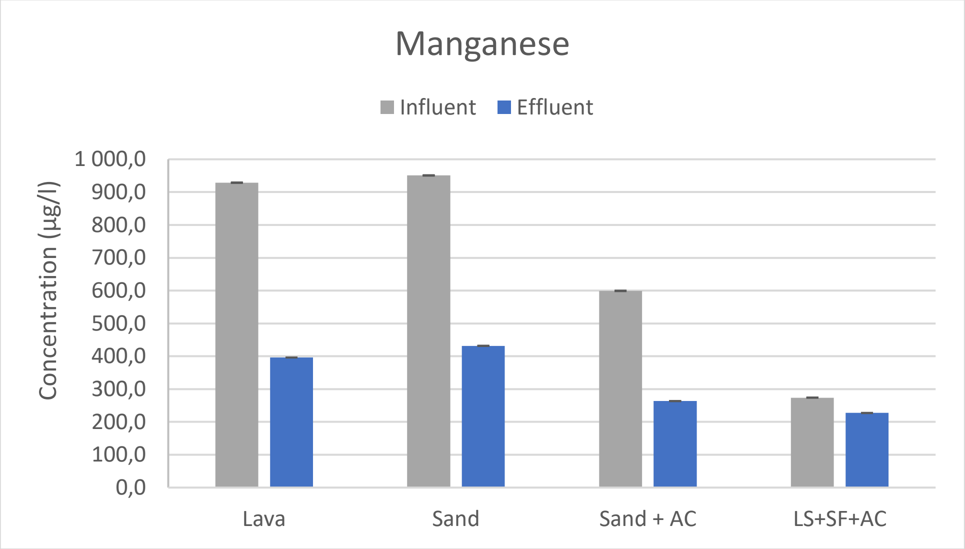


Figure 4-42: Effect of each treatment step on the manganese concentration of the influent for the laundry case (quality requirement: < 50 µg/l)

The variation in calcium concentrations for the laundry case are presented in Figure 4-43. The influents had the same concentrations over time with the exception of the influent of the total treatment train. One possible complication is the use of a Certipur® ICP multi-element standard solution IV to prepare calibration curves with a range from 1 to 1000 µg/l. The concentration of calcium in the influents however is between 30 000 and 50 000 µg/l which is way out of bounds of the calibration curves. The accuracy of the results is thus not very high and hardness titrations or calcium test kits would have yielded more accurate results.

Every treatment step was able to remove a fraction of the calcium concentration from its influent. The lava rock, sand and sand + GAC filter all were able to remove more or less the same amount of calcium. Only the complete treatment train exhibited unusual behaviour: the influent concentration is lower than the other influents (probably due to sampling and dilution errors) and the effluent concentration is higher than the influent. No backwashing operation was carried out during the experiments so the presence of tap water residues did not play a role. If sedimentation happened, an evolution as with copper (see Figure 4-40) and manganese (see Figure 4-42) should be expected. All effluents with the exception of lava rock + sand filter + GAC conformed to the requirement of a maximum concentration of calcium in the effluent of 40 mg/l (see Table 4-13). An extra test should be able to determine whether a measuring error occurred for the last test or a substantial increase of calcium can be detected. The lava rock filter and sand filter + GAC both showed promising results and it is to be expected that a combination of these techniques would work even better, so the total treatment train should be considered the best option.

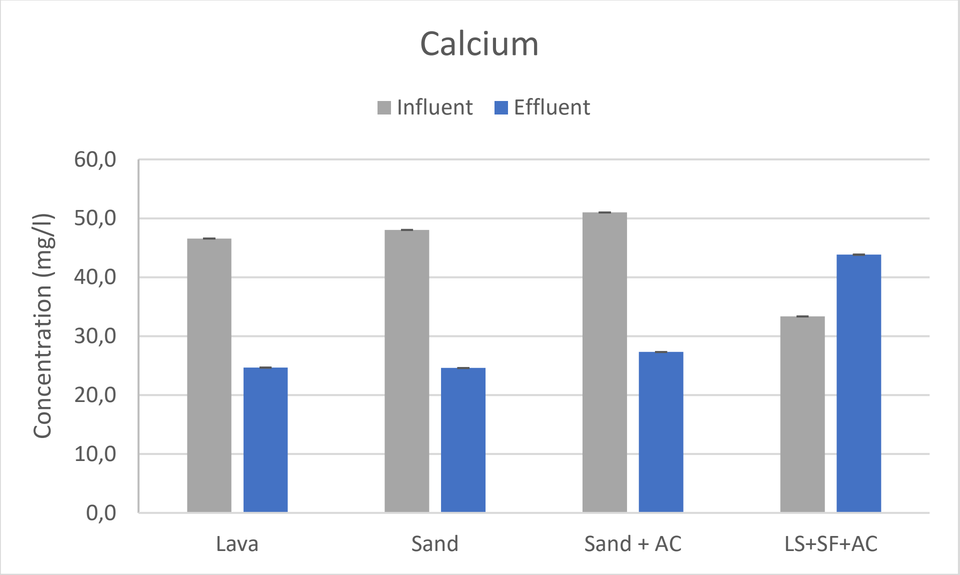


Figure 4-43: Effect of each treatment step on the calcium concentration of the influent for the laundry case (quality requirement: < 40 mg/l)

4.6.4 Microbiological quality after treatment

Each stream was also analysed for microbiological activity. Next, a variety of disinfection techniques were tested to see whether these were successful in disinfecting the prepared influent. Hydrogen peroxide and chlorine disinfection were applied in different concentrations to determine the optimal dosage. Lastly, the effluents of each treatment operation were disinfected by four different techniques: UV-irradiation, H_2O_2, chlorine and ozone. These treated samples were analysed for total coliforms (37°C) and total viable count (22°C) as discussed in 3.6.4.

Different disinfection techniques with various applied concentrations were compared to see if the disinfection would be sufficient using that concentration. The results of the screening test are presented in Figure 4-44 and Figure 4-45. The influent consisted of the prepared mix of streams used for the analyses of standard parameters and soluble metals for the laundry case.

Total viable count (see Figure 4-44) is discussed first. The influent had a concentration of (1.8 ± 0.5) x 10^3 CFU/ml and the samples disinfected with chlorine contained the lowest concentration with ((5 ± 2) x 10 CFU/ml for 1 mg/l Cl_2 and (2.2 ± 0.7) x 10 CFU/ml for 2 mg/l Cl_2). The sample with the highest concentration of H_2O_2 (90 mg/l) had the lowest concentration ((2.9 ± 0.9) x 10^2 CFU/ml) as can be expected. The disinfection with ozone was less successful due to the low solubility of ozone caused by insufficient cooling of the stock solution during the bubbling of ozone through the double distilled water (see 3.5.1). The concentration of 2 mg/l is most likely not achieved (indicated by * in Figure 4-44). When comparing all the techniques, chlorine stands out due to the low cost, easy of appliance and high effectiveness of disinfection. The residual chlorine concentrations were also determined by using chlorine test kits (see Table 3-6). A comparison of the effectiveness of the disinfection by chlorine for each treatment step could be made. A high residual chlorine concentration is an indication that less chlorine was consumed to disinfect the water sample and the physicochemical treatment was more effective in the removal of micro-organisms from the influent.

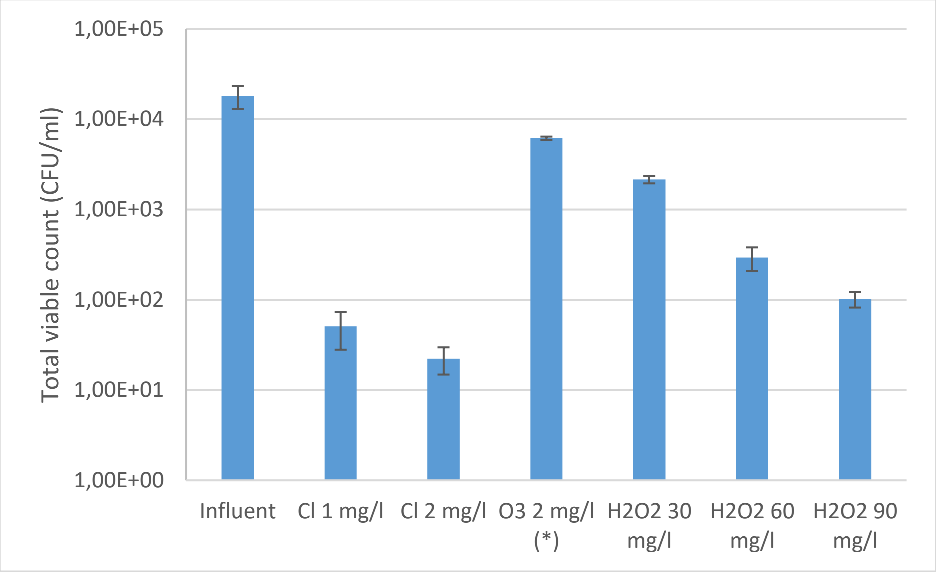


*Figure 4-44: Comparison of the effectiveness of each disinfection technique with their respective concentration analysed by total viable count (22°C) (requirement: < 10^2 CFU/ml, * = target concentration)*

The results of the screening test for total coliforms and *E. coli* to determine the appropriate disinfection techniques and concentrations are presented in Figure 4-45. The only technique that failed to disinfect the influent was ozone due to insufficient cooling of the stock solution, as discussed above. The rest of the applied techniques were more or less effective. Chlorination (1 and 2 mg/l dosage) was able to reach the requirement of complete disinfection, hydrogen peroxide with a dose of 90 mg/l came close with a concentration of 1 CFU/100 ml total coliforms and < 1 CFU/100 ml *E. coli*.

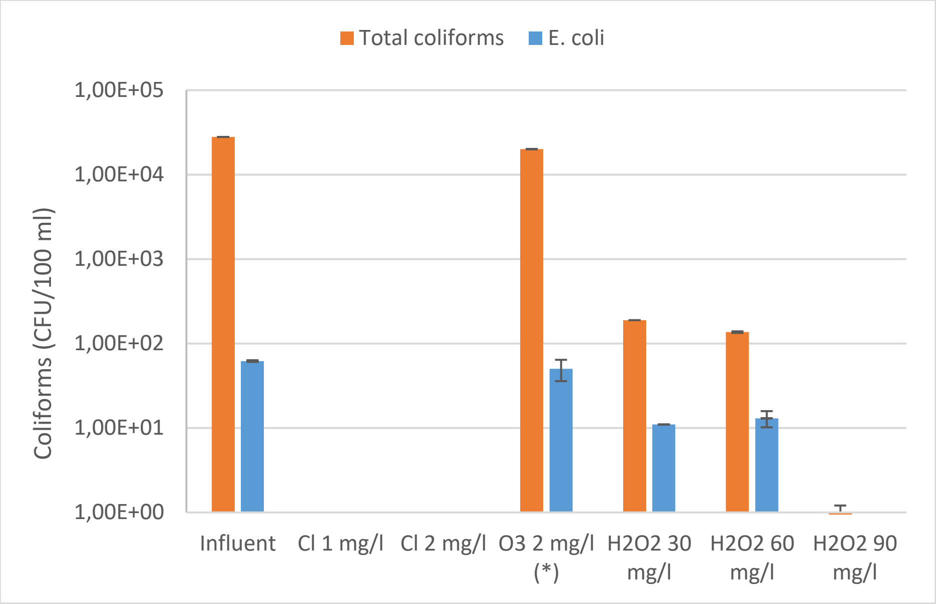


*Figure 4-45: Comparison of the effectiveness of each disinfection technique with their respective concentration analysed by total coliforms (37°C) (requirement: < 1 CFU/100 ml, * = target concentration)*

The following disinfection techniques were applied for the effluents of the filtration tests after analysing the disinfection screening tests: UV-irradiation, ozone (5 mg/l), chlorination (2 mg/l) and hydrogen peroxide (90 mg/l). Samples were taken after each filtration step to determine the efficiency in removing micro-organisms from the influent by physicochemical treatment. Then samples were also taken to disinfect the effluent of the filtration. All samples were taken and analysed according to 3.6.4. The results of the lava rock filtration regarding the total viable count are shown in Figure 4-46.

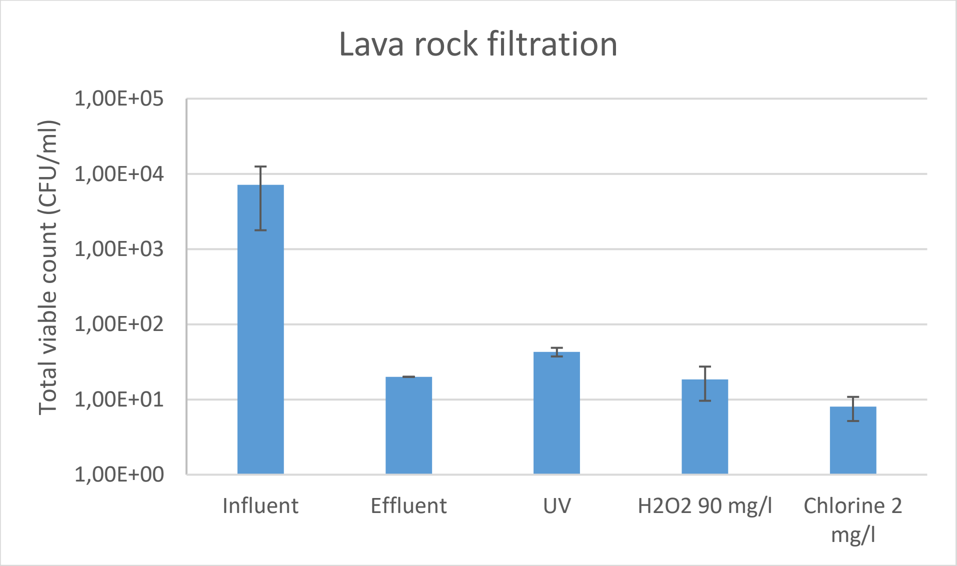


Figure 4-46: Total viable count (22°C) results of the lava rock filter effluent and its disinfected samples (requirement: < 10^2 CFU/ml)

The lava rock filter was able to withhold a large fraction of micro-organisms from the influent (concentration effluent = 20 ± 0 CFU/ml). The removal was improved by adding disinfectants with the exception of ozone (data excluded). The cooling was insufficient so the stock solution was not saturated and not effective. This was also the only sample which did not meet the requirements regarding the total viable count given in Table 4-13. The most effective disinfectant was chlorine (2 mg/l) which yielded a concentration of 8 ± 3 CFU/ml.

The results of the lava rock filtration for total coliforms and *E. coli* are shown in Figure 4-47. The concentration of total coliforms increased after the filtration but the concentration of *E. coli* remained the same. Microbiological contamination thus occurred in the lava rock filter. Ozone and UV-samples did not conform to the requirement although the *E. coli* concentration in the UV samples were < 1 CFU/100 ml. The data for ozonation is for the same reason as for the total viable count excluded. Only the sample disinfected with chlorine met the requirement for total coliforms.

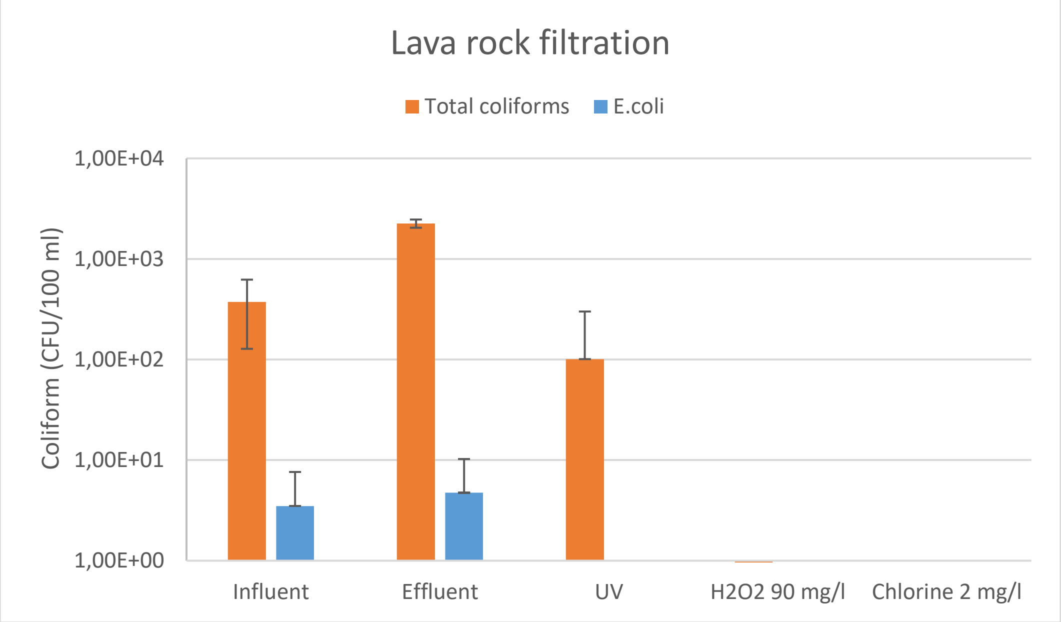


Figure 4-47: Total coliform (37°C) results of the lava rock filter effluent and its disinfected samples (requirement: < 1 CFU/100 ml)

The results of the sand filtration for total viable count are shown in Figure 4-48. The concentration of the influent has decreased in comparison with the lava rock filtration (see Figure 4-46). The effluent now had a higher concentration than the influent. The UV disinfected sample also showed no effective disinfection in comparison with the other techniques. Contamination of the lamp interior and outflow tubing can be the reason. The cooling of the ozone stock solution was now successful and it was effective in removing micro-organisms. Ozone, H_2O_2 and chlorine were the only samples which met the requirement. Chlorine was the most effective with a concentration of 5 ± 4 CFU/ml.

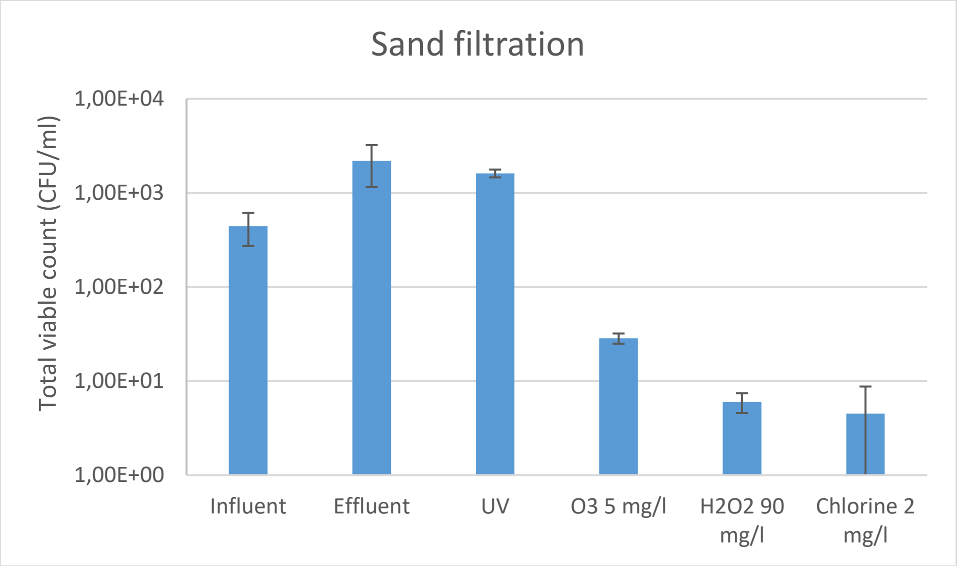


Figure 4-48: Total viable count (22°C) results of the sand filter effluent and its disinfected samples (requirement: < 10^2 CFU/ml)

Figure 4-49 presents the results of the sand filtration regarding the coliforms. The effluent concentration is equal to the influent concentration and a small increase in *E. coli* concentration can also be observed. All disinfection techniques were able to meet the requirement requested by the industry. UV-irradiation was in contrary to the total viable count (see Figure 4-48) able to successfully remove coliforms from the effluent.

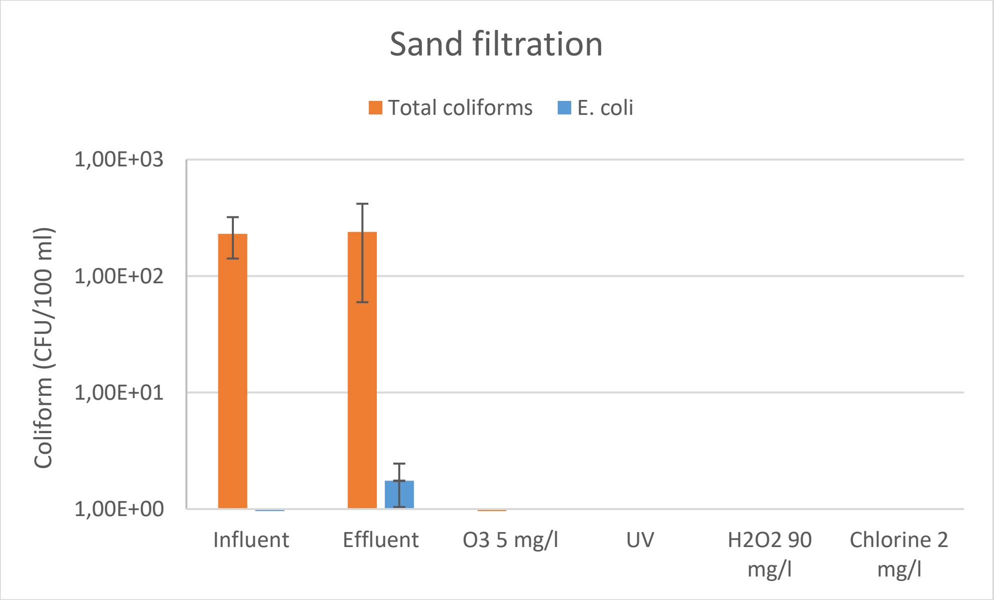


Figure 4-49: Total coliform (37°C) results of the sand filter effluent and its disinfected samples (requirement: < 1 CFU/100 ml)

Figure 4-50 shows the effectiveness of the sand filter combined with activated carbon for the total viable count. The effluent contained twice as much micro-organisms as the influent which is an improvement when comparing this with the results of the sand filtration (see Figure 4-48). Microbiological contamination of the GAC (caused by biofilm growth) and sand filter could have caused the increase as it was the case for the microbial quality of the long term filtration tests (see 4.3.4). The contamination of the UV-lamp was still present even after a bleach solution of 5 mg/l was pumped through the lamp. Insufficient contact time with the outflow tubing might be the reason. The influent, effluent and UV-sample were higher than 100 CFU/ml, chlorine had the lowest concentration with < 1 CFU/ml.

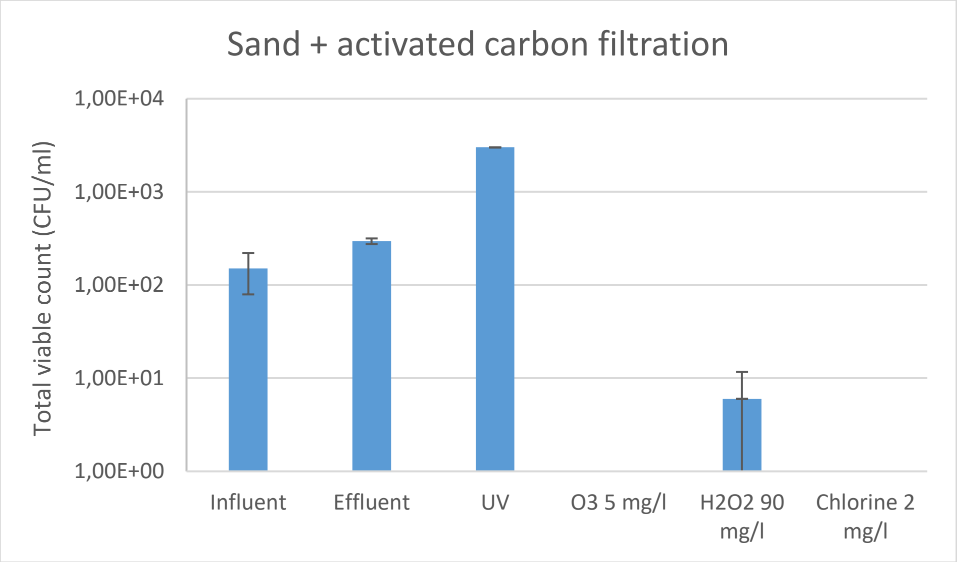


Figure 4-50: Total viable count (22°C) results of the sand filter + GAC effluent and its disinfected samples (requirement: < 10^2 CFU/ml)

The results of the sand filter + GAC set-up for total coliforms are presented in Figure 4-51. No presence of *E. coli* was detected in any sample and all techniques were effective to disinfect the effluent and to achieve the requirement.

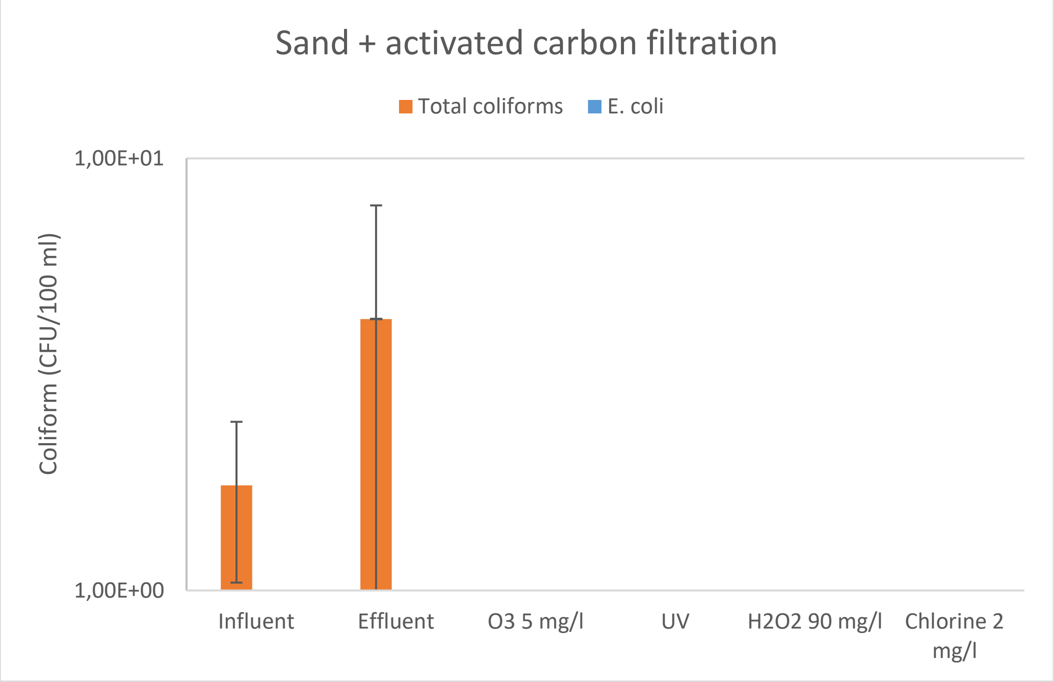


Figure 4-51: Total coliform (37°C) results of the sand filter + GAC effluent and its disinfected samples (requirement: < 1 CFU/100 ml)

Finally, the complete treatment train was also tested and samples were taken for microbiological analyses. The influent concentration was once again lower than the previous test. The effluent concentration was much higher than that of the influent (see Figure 4-52).

Contamination in one of the filters, probably the sand filter when looking at the results of the sand filtration, has occurred. The concentration of the UV sample was lower than the those of the previous tests but is still not comparable in effectiveness with the other disinfection techniques. Only the effluent and UV-samples had a concentration higher than 100 CFU/ml. The most effective disinfectants for this test were ozone and chlorine (both had a concentration < 1 CFU/ml).

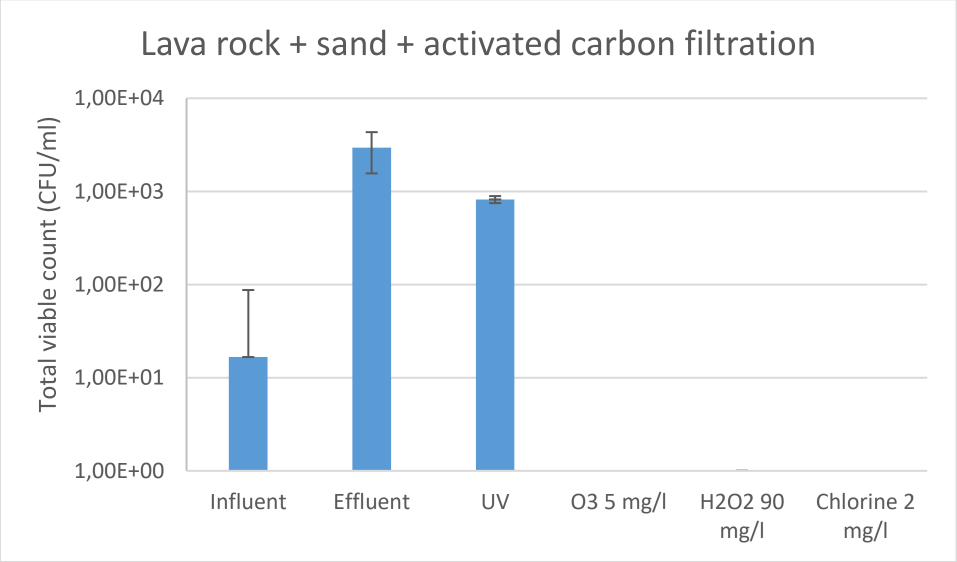


Figure 4-52: Total viable count (22°C) results for the effluent of the complete filtration train and its disinfected by-products (requirement: < 10^2 CFU/ml)

The effluent concentration of this set-up (see Figure 4-53) had a higher concentration of total coliforms than in the previous test. However, as with the sand filter + GAC samples, all disinfection agents were able to meet the requirement.

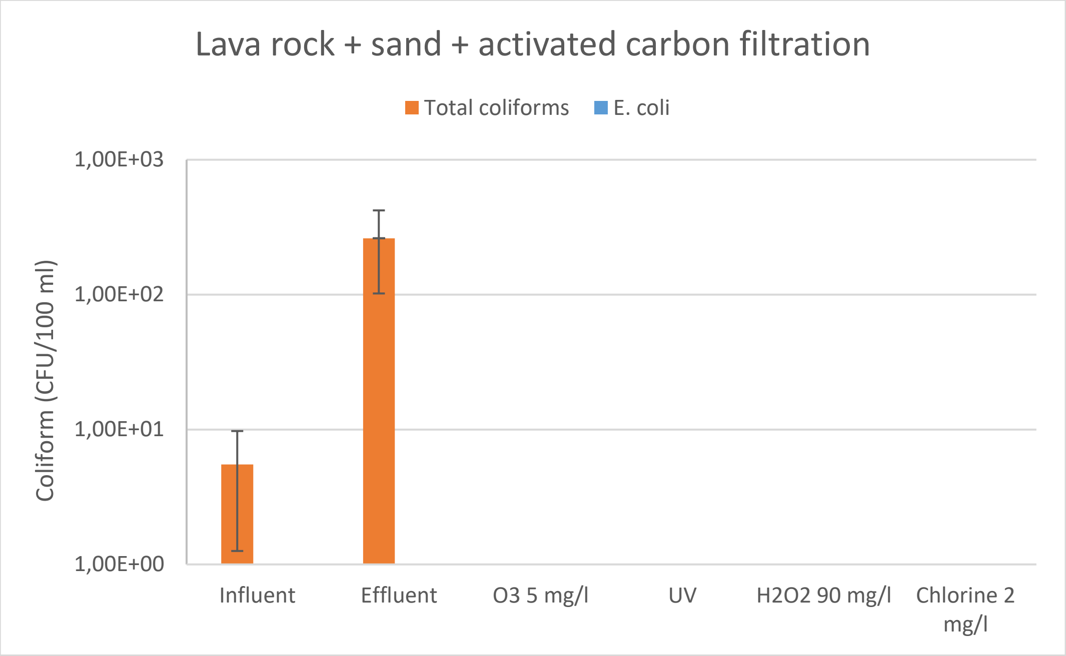


Figure 4-53: Total coliform (37°C) results of the total treatment train effluent and its disinfected samples (requirement: < 1 CFU/100 ml)

It can be concluded when comparing all these tests that the complete treatment train or the sand + GAC filter are a good option to obtain microbiological safe water that conforms to the requirements. Disinfection techniques such as hydrogen peroxide, chlorine and ozone were effective in removing pathogens from the effluent. Some techniques however require more know-how while some are more robust. Chlorine stands out in each test because of its effectiveness. The low cost and ease of use are also advantages worth noting. UV-irradiation was the least promising technique regarding the total viable count in these tests. Contamination was detected and was hard to get rid of. The lifespan of the lamp, particle accumulation on the quartz sleeve etc. all could have played a negative role in the effectiveness. However, this technique was successful in removing coliforms from the effluent (with the exception of the lava rock effluent).

The residual chlorine concentration in the samples disinfected with chlorine was measured and is presented in Table 4-15. Residual chlorine is used to keep disinfecting properties in the water when it is stored or transported through pipes. Contamination caused by transportation and storage of the treated water is thus avoided. As discussed in 2.4.3.1, the optimal residual concentration should be between 0.2 and 0.5 mg/l. Only the samples of the sand filter + GAC and the complete treatment train had a residual concentration which was higher, so a lower dosage is advised when applying these techniques. A concentration of 1 mg/l used for the direct disinfection of influent without physicochemical treatment (as performed in the screening tests) is too low to have residual disinfecting properties.

Table 4-15: Residual chlorine concentrations in all of the applied filtration tests

Test (dosage)	**Residual chlorine (mg/l Cl_2)**
Comparison disinfection techniques	1 mg/l: 0.149 2 mg/l: 0.243
Lava rock filtration (2 mg/l)	0.205
Sand filtration (2 mg/l)	0.206
Sand filtration + GAC (2 mg/l)	0.525
Lava rock + sand + GAC (2 mg/l)	0.843

It can be concluded that the physicochemical treatment had, in general, a negative effect on the removal of coliforms from the influent. This type of treatment was needed to improve the standard water quality parameters and metal concentration. Robust and effective disinfection was therefore needed. Chlorination can be considered the best option, as it was for total viable count analyses, as it is reliable, cheap and effective in use.

4.6.5 Conclusion of the laundry case

When reviewing all the obtained results from the laundry case, it can be concluded that the treatment consisting of a sand and activated carbon filter is a good choice to make sure the predetermined physicochemical requirements are met. Only the pH and the concentration of manganese were too high in the effluent. Adjusting the mixing ratio of the runoff of the three roofs could potentially solve this problem. The manganese concentration in the effluent could also be lowered by using IEX techniques. Adding lava rocks to the set-up does not add enough extra value to justify the costs. The closest resembling set-up found in the literature was the one used by Teixeira and Ghisi (2019) who used gravel along with sand and GAC as filtration medium. No metal concentrations however were measured there with the exception of calcium, only the pH and the turbidity can be compared. The pH showed similar behaviour but the set-up that was tested here was more effective in lowering the turbidity. The calcium levels in the research of Teixeira and Ghisi (2019) also increased after filtration due to leaching of carbonate from the sand and activated carbon.

Chlorine can be considered the best option to disinfect the filtration effluent. Robust disinfection is needed as microbial contamination can occur in the filter, especially when using GAC. The great effectiveness of this agent during the tests, the low cost and ease of use make this the preferred choice.

5 Conclusion

Roof runoff is a promising alternative source of water that can potentially be used for household and industrial applications. Several sources of pollution reduce the quality of rainwater before it is labelled runoff. The different sources along with the average values of standard quality parameters are discussed. Emphasis was put on the presence of inorganic and organic micropollutants in runoff as these are less discussed in literature. The current legislative framework concerning the reuse of runoff in Flanders in almost non-existent. Only the drinking water standard and the requirements determined by the laundry sector can be used as comparison. Other countries such as Spain offer a more comprehensive framework which encourages the reuse of reclaimed water for various applications. The financial viability of RWH systems is a decisive factor when opting for the implementation of such a system in a household or industrial setting. Many factors influence the total costs and investment analyses should therefore be performed on a case-by-case basis.

This research consisted of two major parts. The first part focussed on executing long term filtration tests, both in continuous and discontinuous operating mode. Therefore, 9 metals and 8 organic compounds were added to collected roof runoff and this water was treated by sand filtration followed by a GAC column. First off, side-effects influencing the stability of the spiking solutions were examined. For the standard quality parameters, no substantial difference between the two operating modes could be determined. The water was already fairly clean due to the pre-treatment at the sampling point. The quality of all GAC effluent samples conformed with the drinking water standard. A correlation between the COD levels and UV_{254} for online monitoring of the water quality could not be determined. The removal of soluble metals by sand and activated carbon filtration fluctuated. For the discontinuous treatment it could be established that longer periods without backflushing of the sand filter increased the removal of metals. This is probably caused by the presence of biofilm which improved the removal of metals. This phenomenon was not observed for the continuous filtration as this set-up was backflushed more regularly than the discontinuous set-up. Only the concentration of nickel in the effluent of both the discontinuous and continuous filtration was higher than legally allowed, for 3 and 2 samples respectively. The other metal concentrations were lower than the legal limit. The performed shock chlorination caused leaching of metals that were adsorbed onto the GAC. Temporary elevated concentrations of metals in the effluent (all lower than the legal concentration limits) were detected. The removal of organic compounds (PAHs, diuron and phthalates) was more consistent because of the specific type of activated carbon that was used. The effectiveness of the sand filtration in removing the spiked organic compounds was dependant on the molecular weight and solubility of the compounds. This treatment resulted in a partial and varying removal. By introducing activated carbon, removal of organic was improved and a clear decreasing evolution, indicating a breakthrough of the GAC, could be observed for the PAHs and diuron. The phthalates continued to show a lot of fluctuation. An increased removal of organics could be observed after the chlorination caused by the desorption of metals from the GAC as mentioned before or by the detachment of biofilm from the GAC surface. Five samples of both the continuous and discontinuous set-up exceeded the drinking water limit for total

PAHs after treatment. Eight samples of the continuous and 4 samples of the discontinuous set-up exceeded the concentration limit for diuron in drinking water. Finally, two samples of the continuous and one sample of the discontinuous set-up exceeded the WHO guideline for DEHP in drinking water. No major discrepancies were observed between the continuous and discontinuous set-up for the removal of the spiked organic or inorganic micropollutants. The microbial quality was also assessed for the continuous filtration. Influent concentrations concerning total viable count and coliforms were similar as concentrations mentioned in the literature. UV-irradiation was chosen as disinfection agent for its ease of use, effectiveness and low cost. The effluent of the UV-lamp still contained 170 CFU/ml which is higher than the legal limit for drinking water of 100 CFU/ml. It can be concluded when regarding the concentration of coliforms that microbiological contamination occurred in the sand filter of the discontinuous and GAC columns of both set-ups. The UV-lamp was only able to achieve a 1.07 log unit reduction. With an UV-effluent concentration of (1 ± 1) x 10^2 CFU/100 ml of total coliforms, this stream did not conform with the Flemish drinking water norm which dictates a concentration of 0 CFU/100 ml. After performing the shock chlorination (5 mg/l) to control the biofilm growth in the whole filtration set-up, no microbial activity was measured in any of the influent and effluent samples. The last part of the long-term filtration tests consisted of predicting the breakthrough of each spiked component based on the removal behaviour over time. The breakthrough bed volume of copper for both the continuous and discontinuous filtration was less relevant as the removal of this metal was variable. The breakthrough of PAHs and diuron however showed a better correlation and is thus more relevant. The breakthrough of a commercial GAC cartridge when implemented into the long-term filtration set-up as well as in a household consumption scenario was predicted. The proposed cartridge had a larger volume so the breakthrough for each component would take longer. For the household scenario, it was assumed that 66 l of runoff per person per day would need to be filtered. This is equal to 225 bed volumes if three people are considered. A replacement of the cartridge would then need to take place after 6 months if the metals are considered or after 1 year when only the organic compounds are considered and the phthalates are disregarded.

The second part consisted of performing filtration tests to obtain water which is fit-for-use in a laundry company. Runoff from three different roofs was collected, mixed and treated to conform with the predetermined quality requirements for washing water. The treatment train was made up of a lava rock filter followed by a sand filter and ultimately a GAC column. As microbial quality was paramount, different disinfection agents were applied and the results compared. Chlorination, hydrogen peroxide, ozonation and UV-irradiation were used for these tests. The pH of the total treatment train effluent was high but could be controlled by modifying the mixing ratio of the runoff volumes from the different roofs as one roof delivered runoff with a high pH. The conductivity and turbidity of the effluent were low enough to be used as process water in the laundry industry. Regarding the concentrations of soluble metals, copper was far below the required concentration limit of 100 µg/l, the concentration of iron in the effluents was always lower than the LOQ. The only concentration limit that could not be reached was the one for manganese, the effluent of the total treatment train still contained 227 µg/l while a maximum of 50 µg/l was allowed. The presence of activated carbon certainly helped to reduce the manganese concentration but in one runoff stream, from the industrial building, that made up the influent, an exceptionally high concentration of 1076 µg/l was detected. Possible solutions are to apply IEX or to modify the mixing ratio of the three streams so less water from the industrial building is used. Lastly, the calcium concentration in the effluent of the total treatment train was higher than the imposed limit of 40 mg/l but it cannot be concluded that this choice of treatment had a negative impact on the calcium concentration. The effluents of the separate treatment steps (lava rock filter, sand filter and sand + GAC filter) all had a lower calcium concentration than their respective influent and well under the 40 mg/l limit. A measuring or sampling error may be the cause for the increase in concentration for the total treatment train. When comparing all the results, it can be concluded that a system consisting of a sand and GAC filter is sufficient to meet the requirements. The extra quality improvement by lava rock filtration does not justify the costs. Last but not least, the microbiological quality was also an important parameter. When comparing the results of the performed tests, it can be concluded that only chlorination (2 mg/l) was able to continuously meet the quality requirement, both for the total viable count (22°C) and total coliforms (37°C) concentration. The total treatment train also contained the lowest residual chlorine concentration, indicating that this sequence of treatment steps yielded the lowest concentration of micro-organisms and NOM after the physicochemical treatment and less chlorine is used for disinfection or the production of disinfection by-products. This is also one of the main drawbacks when using chlorine besides presence of the typical odour. Chlorination should thus be preferred over the other disinfection alternatives because of its effectiveness, low cost and ease of use. The other alternatives (i.e. ozonation, UV-irradiation and hydrogen peroxide) were less effective in these tests.

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