Costs of Removal of Micropollutants from Effluents of Municipal Wastewater Treatment Plants

General Cost Estimates for the Netherlands based on Implemented Full Scale Post Treatments of Effluents of Wastewater Treatment Plants in Germany and Switzerland

On behalf of

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Summary

Municipal wastewater treatment plants play a key role in micropollutant entry to surface water bodies, as they are the collection point of urban wastewater. Conventional wastewater treatment processes are basically designed to remove macropollutants and remove micropollutants only partially. The non-removable part of micropollutants enters water bodies through the discharge of effluents of wastewater treatment plants.

STOWA and the Interreg IV-B project TAPES have taken the initiative to do a first general study on the current knowledge on removal of micropollutants from effluents of municipal wastewater treatment plants in Germany and Switzerland. The objective of this study is to translate the current experience in full scale applications from Germany and Switzerland to Dutch conditions, especially on costs involved. The techniques which are extensively researched on a large scale on effluents of wastewater treatment plants are:

- Ozonation
- Powdered Activated Carbon (PAC) dosage and
- Granular Activated Carbon (GAC) filtration

For these three techniques costs are estimated in this report for three scales of wastewater treatment plants, which are common in the Netherlands (see table A).

Table A – Costs / treated m³ of wwtp effluent for micropollutant removal in the Netherlands; assumed DOC concentration 7-15 mg/l

<table>
<thead>
<tr>
<th>Capacity wastewater treatment plant</th>
<th>20.000 p.e.</th>
<th>100.000 p.e</th>
<th>300.000 p.e</th>
</tr>
</thead>
<tbody>
<tr>
<td>150 g TOD</td>
<td>150 g TOD</td>
<td>150 g TOD</td>
<td></td>
</tr>
<tr>
<td>Ozonation + sand filtration</td>
<td>€0.22 ± € 0.04</td>
<td>€0.18 ± € 0.03</td>
<td>€0.16 ± € 0.03</td>
</tr>
<tr>
<td>PAC + sand filtration</td>
<td>€0.26 ± € 0.04</td>
<td>€0.20 ± € 0.03</td>
<td>€0.18 ± € 0.03</td>
</tr>
<tr>
<td>GAC</td>
<td>€0.29 ± € 0.04</td>
<td>€0.27 ± € 0.04</td>
<td>€0.26 ± € 0.04</td>
</tr>
</tbody>
</table>

Removal efficiencies of micropollutants differ per post treatment. Depending on the substance, the technique and the way in which the technique is implemented, different substances will have different removal rates. In general persistent micropollutants like x-ray contrast media will not be removed by post treatment of wwtp effluent. Other micropollutants will generally be removed in the range of 30-50% to more than 80%.

For ozonation, the formation of toxic transformation products is a topic of discussion. In Germany and Switzerland it is advised to implement a biological sand filtration step after ozonation, to remove any biodegradable transformation products formed during ozonation. Whether this sand filtration after ozonation is adequate enough is not known. For PAC treatment, sand filtration is necessary to remove small PAC particles, not because of the formation of metabolites. To reduce the risk of discharging toxic transformation products and metabolites into the environment, the Dutch 1-STEP© concept can be implemented [54]. In this case the sand filter after ozonation will be filled with activated carbon, through which more metabolites and transformation products presumably will be removed, but this will increase costs per treated m³ of effluent by 35%.
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1 Introduction

Pollution of water resources by chemical compounds draws the attention of environmental scientists for many decades. Urban development resulted in the necessity for urban water infrastructures, basically to collect and treat the polluted wastewater. In developed regions of the world, these infrastructures are designed at first to remove carbonaceous compounds, and later also the nutrients, nitrogen and phosphorus. These substances are known as common wastewater constituents found in domestic wastewater in milligram per litre range and referred to as “macropollutants”. In the last decade, another term “micropollutants” has become an increasingly pronounced expression. The term basically describes the chemicals released to the water-soil-air matrix by anthropological activities, yet these substances differ from macropollutants, basically by their very low concentration in range of microgram to picogram per litre. An extremely large number of substances with different origins and chemical properties are considered in this group, among which pharmaceuticals, personal care products, pesticides, biocides and several other industrial chemicals. The variety of substances makes a general statement on their environmental impact rather challenging, yet scientific studies readily indicate that in spite of their low concentrations, micropollutants can cause serious consequences in natural water bodies. Some examples are given in chapter 2.

Micropollutants can enter the aquatic environment through diffuse or point sources. In urbanised regions, they are basically transferred to wastewater, for instance, from households and transported by sewer systems to wastewater treatment plants. Conventional wastewater treatment processes are basically designed for the removal of macropollutants and can offer only a partial removal of micropollutants. The non-removable part of them enters through the treatment plant effluent into water bodies. Some good soluble substances go through the conventional treatment process almost unchanged.

The issue of micropollutants in water bodies is particularly relevant for regions with a large population density and for those which withdraw potable water from surface water resources. Solution should be sought basically at the source; critical substances should be substituted as far as possible. However, it is not likely that all of the substances, particularly pharmaceuticals, can be replaced with harmless alternatives, thus the end-of-pipe technologies seem to be an indispensible part of the solution.

Recent laboratory and later pilot scale investigations proved that a wide range of micropollutants can be removed by adsorptive and oxidative techniques within cost-effective applications implemented at the effluent of the conventional treatment plants. This additional micropollutant removal step is often referred to as the “fourth step”. In Germany and Switzerland additional micropollutant removal is already implemented at different wastewater treatment plants: 15 fourth step plants have been implemented in Germany and one in Switzerland. These plants treat 80-100 % of the annual wastewater amount coming to the treatment plants where they are erected and thus they are considered here as full scale plants. In all of these plants the implemented processes basically consist of activated carbon adsorption or ozonation.

The awareness regarding the impact of micropollutants on water quality also increased in the last decade in Netherlands. Up to date, some pilot scale research has been carried out on the removal of micropollutants from wwtp effluents, yet only one full scale installation has been implemented. The latter installation is the treatment of effluent of the wwtp Horstermeer using the 1-STEP®-process, which is a fixed bed filter filled with granular activated carbon. Research has been carried out on the removal of macropollutants like nitrogen and
phosphorus, but also on micropollutants. However the main objective and the operation of the 1-STEP-filter is aimed at the removal of nitrogen and phosphorus, not the removal of micropollutants.

Currently there are no clear legal tools to oblige the extension of treatment plants with an additional step for micropollution removal. Thus at the current stage, it is mainly a question of costs, under which conditions actions can be taken to control micropollutant entry from wastewater treatment plants. To assess the situation for the Netherlands, the Dutch water authorities can make use of the past experiences from Germany and Switzerland.

1.1 Objectives of the study

STOWA and the Interreg IV-B project TAPES have taken the initiative to do a first general study on the current knowledge on removal of micropollutants from effluents of municipal wastewater treatment plants in Germany and Switzerland. This study has been conducted in order to translate the current experience on the pilot and full scale applications from Germany and Switzerland to Dutch conditions.

This study considers mainly the full scale, in some cases also the pilot scale implementations in German and Swiss municipal wastewater treatment plants, where 80-100% annual wastewater arriving in the plant is treated for the micropollutant removal. The implemented processes include the ozonisation process, activated carbon adsorption with powdered activated carbon (PAC) and with granular activated carbon filtration (GAC). These processes are often combined with an additional filtration step; in case of ozonisation generally for biological post-treatment purposes and in case of PAC adsorption for particle detention purposes. These processes are also subject of this study.

Regarding the applications where the number of full scale applications is yet too few, also pilot scale applications are considered. Pilot scale applications are often given under research works in the literature, thus they are presented in this study also as research work. However, a detailed review of the research works on micropollutants is not within the scope of this study. Interesting reviews of research works regarding the design, efficiency and the costs of micropollutant removal are given in several other reports [23][25][32][38][39][53][63][65][66].

This study presents an estimate of the costs of micropollutant removal in the Netherlands, based on the state of the art at full scale implementations in German and Swiss municipal wastewater treatment plants. As an example costs are calculated for three scales of wastewater treatment plants which are common in the Netherlands. Also first insights on the operation and maintenance of post treatment of effluents of wastewater treatment plants are given.

This is a first general study and is not meant to fully elaborate on all the differences between the Netherlands, Germany and Switzerland considering municipal wastewater treatment. Also it is not the objective to give policy scenario’s and full cost calculations to prevent micropollutants from entering water bodies through discharge of effluents of wastewater treatment plant.

The report consists of 6 chapters in total, chapter 1 being the introduction and the current chapter describing the objectives of the study. In chapter 2 some background information is given on micropollutants. In this chapter the origin and impact of micropollutants in the watercycle and aquatic environment are explained. Also the legal situation concerning
micropollutant is shortly discussed for the Netherlands, Germany and Switzerland, as well as the research on post treatment of micropollutants in Germany and Switzerland. Then in chapter 3, the functionality, operation and maintenance aspects of implemented techniques for post treatment of wwtp effluent are explained. The discussed techniques include ozonation, powdered activated carbon (PAC) adsorption and granular activated carbon (GAC) filtration. In chapter 4 the design criteria of these three techniques are discussed. The costs are calculated in chapter 5. Conclusions and recommendations are summarized in chapter 6.


2 Background information on micropollutants and micropollutant removal

2.1 Basic properties of micropollutants

Micropollutants basically describe the residual substances released to the water-soil-air matrix after anthropological activities in very low concentrations such as microgram to picogram per litre. They can be of natural origin or industrially synthesised, but the substances subject to this study are basically the industrially synthesised chemical compounds.

Micropollutants as a group contain extremely large number of chemicals with different origins. The residues of pharmaceuticals, personal care products, pesticides, biocides, plasticiser, flame retardants and several other industrial chemicals are counted in this group. According to the actual EINECS data (European Inventory for Existing Commercial Chemical Substances), there are more than 100.000 registered chemicals in the market with known data. The growth of newly synthesized chemicals shows an exponential trend [18]. Not only the number is large but also the physicochemical properties of micropollutants vary in a wide range. They can be polar or apolar, biodegradable or persistent, hydrophilic or hydrophobic, in fact, they can behave different depending on the physical conditions of the aquatic milieu, such as the pH value. Thus their behaviour in aquatic systems cannot be standardised. The variety of the substances constitutes one of the main challenges to assess and control micropollutants. Also their occurrence in very low concentrations can be an analytical challenge.

Micropollutants are often found in a cocktail, where not all of the substances are known and the determination of individual impact of substances is almost impossible. They can be transformed to other compounds, which can be more mobile and toxic than the original compound. The unknown (metabolic) substances in a chemical cocktail can cause far greater ecotoxicological impact than the known ones [19]. All these factors distinguish the micropollutants from the commonly known macropollutants, namely carbonaceous, nitrogenous and phosphorous compounds and make both the assessment and the control rather challenging.

2.2 Impacts of micropollutants

Even though the variety of substances considered as micropollutants makes a general statement rather challenging, scientific studies readily indicate that micropollutants can cause, in spite of their low concentrations, serious consequences in the life in natural water bodies. They show, for instance, similarities with the compounds regulating metabolic functions of living organisms, such as hormones and enzymes, and can thus manipulate these functions. Especially the effects of endocrine disrupting compounds (EDCs) on both animals and human beings were researched [1]. A well-known example to this in the aquatic environment is the feminization of male fish in rivers [2], [3], [4]. Besides reproduction, effects in polluted systems can also occur on immune function, behaviour and memory of organisms [5]. Various studies documented observable changes in fishes, amphibians, crustaceans and shellfish possibly linked with the occurrence of micropollutants in their environment [5], [6], [8], [9], [10], [11], [12], [13], [14]. Some substances were detected also in human body, for instance in blood, fat and breast milk [19] indicating also the tendency for bioaccumulation in humans. However, toxicological impact of micropollutants on larger organisms is rather difficult to determine, particularly the chronic toxicity of micropollutants in subtoxical range is rather an unknown field hosting numerous questions to be answered [15].
2.3 Micropollutant entry pathways into aquatic environment

Micropollutants enter the aquatic environment through several pathways. A general overview of entry pathways for regions with an accomplished urban water infrastructure and frequent type of substances according to the source is presented in Figure 1.

As mentioned in the above chapter, micropollutants can be of natural and synthetic origin, yet the majority of micropollutants considered in this study are industrially synthesised products, thus the factories producing them can be classified as “direct” sources at the production level. Any industrially synthesised product can be expected in the production level. The products are utilized in industries, households and public units such as hospitals. Resulting chemicals such as personal care products, pharmaceuticals, cleaning agents, biocides are released to the wastewater, which is then collected by sewer networks and transported mainly to wastewater treatment plants (wwtp). In case of a heavy rainfall, the diluted wastewater in mixed sewer systems can be collected in storm water detention vaults (SWDV) before being released to the wwtp or discharged into receiving water body directly. They can also be transported to aquatic environment by groundwater infiltration through unsealed soil as in agricultural activities. Nutrient recycling by biosolid amendment of soils, for instance land application of sewage sludge, can contribute to the release into groundwater additionally. Another pathway into the aquatic environment is by the surface runoff from streets and other polluted areas, which contain the micropollutants originating from traffic and industrial emissions washed by rainfall from the atmosphere.

Municipal wastewater treatment plants play one of the key roles in micropollutant entry to surface water bodies, as they are the collection point of urban wastewater. Conventional wastewater treatment processes are designed basically for the removal of macropollutants and can offer only a partial removal of micropollutants. The non-removable part of them enters through the treatment plant effluent into water bodies. Figure 2 presents the removal rates of micropollutants in conventional wastewater treatment plants in the Netherlands.

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Figure 1 - Entry pathways of micropollutants into natural water bodies in countries with a developed urban water infrastructure, modified from [17].
Figure 2 - Removal efficiencies of micropollutants in conventional WWTPs in the Netherlands, n represents the amount of samples of influent and effluent on which the removal efficiency is based [28]
The degree to which these are removed varies per substance and is influenced by the physical-chemical properties of the substances. However, it can be seen that for quite a few substances the removal rate remains below 40% in conventional plants. Commonly utilised pharmaceuticals Carbamazepine and Diclofenac are among the substances with relatively low removal rates. This fact makes the conventional wastewater treatment plants one of the key points sources for micropollutants into natural water bodies.

Micropollutants currently released by municipal wastewater treatment plants can be technically controlled by additional units to be constructed in the existing plants. Yet the current legal implications in the European Union do not clearly enforce to control the micropollutant entry into aquatic environment.

### 2.4 Legal situation on the removal of micropollutants

In the Netherlands the research works to remove the micropollutants from wastewater treatment plant effluents are so far limited to pilot scale and 1 full scale installations (1-STEP® filter). The main reasons for this are the absence of legislation and the high costs which are involved in realizing a post treatment at wastewater treatment plants for the removal of micropollutants. The last years, studies on possibly harmful concentrations of micropollutants for aquatic ecology and major rivers like the Rhine and Meuse have drawn the attention of policy makers and politicians [62]. Considering the fact that 35% of drinking water in Netherlands is supplied from surface water resources, this may indicate also a risk for the human health. The adjoining studies indicating the unwanted effects in the aquatic environment and threat of clean drinking water sources lead to a statement by the secretary of the Ministry of Infrastructure and Environment in 2014, that water boards are asked to do further research into the necessity and costs of micropollutant removal from the effluents of wastewater treatment plants.

Switzerland is the first country in Europe, which changed its water law in 2014 with a target of reducing the micropollutant entry to natural water bodies from wastewater treatment plants. The new law requires primarily 100 of the total 700 treatment plants in the country to implement the fourth step. The selected plants are larger ones to treat 50% of the total wastewater. The target hereby is 80% removal of micropollutants in the upgraded plants, resulting in the 50% elimination of micropollutants in Switzerland overall.

Although it is not yet a legal requirement, some treatment plants in Germany, particularly in North Rhine-Westphalia and Baden-Württemberg, have already constructed additional steps for micropollutant removal. Moreover in numerous plants feasibility studies are conducted currently, indicating a certain tendency for more full scale applications (see appendix 1).

### 2.5 Pilot scale research plants and full scale implementations in Germany and Switzerland

In Switzerland and Germany prior to full scale implementations extensive research has been carried out on the removal of micropollutants from wastewater. Laboratory scale experiments have led to pilot scale research on the techniques ozonation and adsorption through powdered activated carbon (PAC) and granular activated carbon (GAC). Other techniques were not competitive on small scale [32]. This decision was made based on removal rates, costs and ease of operation. The researched techniques included membrane filtration like ultra and nanofiltration and reverse osmosis; advanced oxidation processes with UV-light in combination with catalysts like peroxide, ozone and titanium; photolysis and ultrasonic and physical-chemical treatment methods [23][29][30][31] [32].
In Switzerland large scale\(^1\) pilot experiments were carried out at 3 wwtps (ozonation and PAC-treatment) and 1 wwtp has been full scale adapted and in operation since April 2014 (Neugut, ozonation). In Germany pilot scale and large scale experiments were carried out at many wwtp’s for the removal of micropollutants (see appendix 1). At the present situation 16 of these large scale\(^1\) installations are still in operation, 6 installations are currently under construction and for another 11 wwtp’s a post treatment is planned. In general 80 – 100% of the total effluent flow of the wwtp is treated, which is considered a full scale treatment. The techniques which are used in Germany are ozonation and powder activated carbon (PAC) and granular activated carbon filtration (GAC) of wwtp effluent (see table 1) [63].

\(^1\) Large scale is defined by a treated wastewater amount > 200 m\(^3\)/h, aimed at the removal of micropollutants

Table 1- Large scale\(^1\) operations and full scale implementations for removal of micropollutants from municipal wastewater (As of January 2015 [63]; see also appendix 1)

<table>
<thead>
<tr>
<th>Plant size / Design capacity [p.e.]</th>
<th>Technology</th>
<th>Treated amount of wwtp effluent</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>WWTP in North Rhine-Westphalia</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Aachen-Soers x 480.000 Ozone</td>
<td></td>
<td>Q(_{\text{max}}) = 300 m(^3)/h</td>
</tr>
<tr>
<td>Bad Sassendorf x 13.000 Ozone</td>
<td></td>
<td>Q(_{\text{max}}) = 370 m(^3)/h</td>
</tr>
<tr>
<td>Bad Oeynhausen x 78.500 GAC</td>
<td></td>
<td>Q(_{\text{max}}) = 300 m(^3)/h</td>
</tr>
<tr>
<td>Barntrup x 12.000 PAC</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Detmold x 135.000 Ozone</td>
<td></td>
<td>Q(_{\text{max}}) = 300 m(^3)/h</td>
</tr>
<tr>
<td>Duisburg Verlinden x 30.000 Ozone</td>
<td></td>
<td>Q(_{\text{max}}) = 400 m(^3)/h</td>
</tr>
<tr>
<td>Dülmen x 55.000 PAC</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Espelkamp x 33.000 Ozone</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gütersloh x 150.600 GAC</td>
<td></td>
<td>Q(_{\text{max}}) = 840 m(^3)/h</td>
</tr>
<tr>
<td>Harsewinkel x 570.000 -</td>
<td></td>
<td>Q(_{\text{max}}) = 300 m(^3)/h</td>
</tr>
<tr>
<td>Neuss Ost x 280.000 -</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Obere Lutter x 380.000 GAC</td>
<td></td>
<td>Q(_{\text{max}}) = 960 m(^3)/h</td>
</tr>
<tr>
<td>Rietberg x 46.500 GAC</td>
<td></td>
<td>Q(_{\text{max}}) = 360 m(^3)/h</td>
</tr>
<tr>
<td>Rheda x 94.000 Ozone</td>
<td></td>
<td>Q(_{\text{max}}) = 1000 m(^3)/h</td>
</tr>
<tr>
<td>Schwerte x 50.000 Ozone</td>
<td></td>
<td>Q(_{\text{max}}) = 1100 m(^3)/h</td>
</tr>
<tr>
<td>Warburg x 70.000 Ozone</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>WWTP in Baden-Württemberg</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Albstadt x 125.000 PAC</td>
<td></td>
<td>Q(_{\text{max}}) = 3500 m(^3)/h</td>
</tr>
<tr>
<td>Büsnau x 9.680 GAC</td>
<td></td>
<td>Q(_{\text{max}}) = 70 m(^3)/h</td>
</tr>
<tr>
<td>Emmingen-Liptingen x 7.500 GAC</td>
<td></td>
<td>Q(_{\text{max}}) = 70 m(^3)/h</td>
</tr>
<tr>
<td>Freiburg x 600.000 -</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hechingen x 57.200 PAC</td>
<td></td>
<td>Q(_{\text{max}}) = 1440 m(^3)/h</td>
</tr>
<tr>
<td>Karlsruhe x 700.000 PAC</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kressbron x 24.000 PAC</td>
<td></td>
<td>Q(_{\text{max}}) = 900 m(^3)/h</td>
</tr>
<tr>
<td>Lahr x 100.000 PAC</td>
<td></td>
<td>Q(_{\text{max}}) = 1260 m(^3)/h</td>
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<tr>
<td>Laichingen x 35.000 PAC</td>
<td></td>
<td>Q(_{\text{max}}) = 540 m(^3)/h</td>
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<tr>
<td>Lautingen x 36.000 PAC</td>
<td></td>
<td>Q(_{\text{max}}) = 800 m(^3)/h</td>
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<tr>
<td>Mannheim x 725.000 PAC</td>
<td></td>
<td>Q(_{\text{max}}) = 1100 m(^3)/h</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Future Q(_{\text{max}}) = 5400 m(^3)/h</td>
</tr>
</tbody>
</table>

\[1\] Large scale is defined by a treated wastewater amount > 200 m\(^3\)/h, aimed at the removal of micropollutants
<table>
<thead>
<tr>
<th>Location</th>
<th>Implemented</th>
<th>Under construction</th>
<th>Plant size / Design capacity [p.e.]</th>
<th>Technology</th>
<th>Treated amount of wwtp effluent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Öhringen</td>
<td>x</td>
<td></td>
<td>46.000</td>
<td>PAC</td>
<td>-</td>
</tr>
<tr>
<td>Ravensburg</td>
<td>x</td>
<td></td>
<td>184.000</td>
<td>PAC</td>
<td>$Q_{\text{max}} = 4000 \text{ m}^3/\text{h}$</td>
</tr>
<tr>
<td>Sindelfingen</td>
<td>x</td>
<td></td>
<td>250.000</td>
<td>PAC</td>
<td>$Q_{\text{max}} = 4000 \text{ m}^3/\text{h}$</td>
</tr>
<tr>
<td>Stockacher Aach</td>
<td>x</td>
<td></td>
<td>43.000</td>
<td>PAC</td>
<td>$Q_{\text{max}} = 900 \text{ m}^3/\text{h}$</td>
</tr>
<tr>
<td>Stuttgart Mühlhausen</td>
<td>x</td>
<td></td>
<td>1.200.000</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
| Ulm (Steinhäule)  | x           |                    | 440.000                             | PAC        | $Q_{\text{max}} = 5000 \text{ m}^3/\text{h}$ 
|                   |             |                    |                                     |            | Future $Q_{\text{max}} = 9400 \text{ m}^3/\text{h}$ |
| Wendlingen        | x           |                    | 170.000                             | PAC        | -                             |
| Westerheim        | x           |                    | 5.500                               | GAC        | -                             |
| **WWTP in Switzerland** |             |                    |                                     |            |                               |
| Dübendorf (Neugut)| x           |                    | 105.000                             | Ozone      | Dry weather $\text{avg}: 950 \text{ m}^3/\text{h}$ 
|                   |             |                    |                                     |            | Rainweather $\text{avg}: 1900 \text{ m}^3/\text{h}$ |

Based on the large scale research estimates can be made on the costs which are involved in the Netherlands to remove micropollutants from effluents of municipal wastewater treatment plants. First the functionality, operation and maintenance aspects of the techniques ozonisation, powdered activated carbon adsorption and granular activated carbon filtration are further explained in chapter 3. In chapter 4 the design criteria of these three techniques will be discussed. The costs will be calculated in chapter 5.
3 Ozonation and activated carbon treatment of wwtp effluent

3.1 Ozonation

3.1.1 Functionality

Ozone is the triatomic form of oxygen, this means that it is composed of three oxygen atoms. Ozone’s chemical symbol is O$_3$. Under normal conditions ozone is unstable and quickly decomposed to the more stable gaseous oxygen O$_2$. Ozone is capable of oxidizing micropollutants either by a direct reaction with ozone or indirectly after formation of hydroxyl radicals. As a result, the micropollutants are transformed into other compounds and not completely removed from the effluent (see figure 3).

![Diagram of Oxidation patterns through ozonation of wastewater](image)

*Figure 3 - Oxidation patterns through ozonation of wastewater [33]*

As can be seen from figure 3, dissolved organic matter is oxidized by ozone or hydroxyl radicals. This significantly reduces oxidant exposure to micropollutants. This means that the dosage of ozone is dependent on the amount of dissolved organic matter in the wastewater. Also inorganic compounds, such as nitrite, can exert an ozone demand.

The dissolved organic matter is expressed as DOC (Dissolved Organic Carbon). The term 'DOC' is used for the fraction of organics that pass through a 0.45 μm pores size membrane. Overall DOC removal was found to be independent from the DOC concentration in raw wastewater, the removal is rather determined by the composition of the DOC. DOC is therefore not correlated to removal of other organic pollution parameters, such as BOD and COD. Typical removals of DOC are 1 % in the primary treatment, 63% in the secondary treatment, and 69% in the overall treatment, resulting in a DOC content in the effluent of wwtp of 7 – 15 mg/l in Western Europe [45].

DOC consists mostly biopolymers (including extracellular polymeric substances, i.e., mainly proteins and polysaccharides), humic substances, low-molecular-weight acids and low-molecular-weight neutrals [64]. The distribution of these substances are changed through ozonation and PAC-treatment [45]. The higher the DOC content of the effluent of wwtp, the
higher the dosage of ozone (and also PAC) must be, to achieve comparable removal rates for micropollutants [32][44][45][46][47][49] (see chapter 4).

The formation of intermediates by ozonation of wwtp effluent, which are sometimes more toxic than the parent substances are reason for concern. The degradation pathways for several substances have been studied extensively in laboratory research. An example is given in figure 4 for the substance diclofenac. The diclofenac parent substance is partly transformed into other equally toxic intermediates, under nowadays applied ozone dosages and hydraulic retention times and is not completely mineralized [34].

Figure 4 - Degradation of diclofenac in metabolites [34]

Recent studies assessing the effect of ozonation on wastewater toxicity have been inconclusive [35][36][37][39] though biodegradability usually increases after ozonation [32][38][39]. It is however concluded that through ozonation of effluents of wwtp that intermediates are formed, under the current design conditions with dosages around 0.6-1.0 g O₃/g DOC and hydraulic retention times of 20-30 minutes. The DOC content after ozonation is about 90-99% from the DOC content before ozonation, which means that the parent substances and metabolites are not (fully) mineralized [38][39]. Because these intermediates cannot be measured, as the substances are unknown, it is unknown what their toxicological effects are in the cocktail of treated wwtp effluent. Therefore it is advised to implement a biological sand filtration after ozonation [32][40][41][42]. At the wwtp Neugut in Dübendorf, Switzerland, where a full scale ozonation has been implemented, ecotoxicological measurements will be carried out on the effluent after ozonation and also after the different types of post treatment of this ozonated effluent, being a sandfilter, fluidized sand bed filter and granular activated carbon filter [42].

3.1.2 Operation and maintenance
The ozone is produced from pure oxygen through electrical discharge. The produced ozone can then be mixed with the effluents of wwtp through air diffusers in a contact basin. This contact
basin is air-tight as remaining ozone in gaseous form has to be treated. The effluent of the contact basin is then passed through a sandfilter to remove any biodegradable metabolites (see figure 5).

![Figure 5 - Ozonation of effluent of a wastewater treatment plant](image)

The ozone can be also be mixed in by injectors instead of diffusers. The injector type has not been realized much. At wwtc Bad Sassendorf researchers compared treatment by diffusers and injectors. In this study it was concluded that the injectors do not have any extra benefit on removal of micropollutants, compared to diffusers. The energy consumption was slightly higher of the injectors compared to the diffusers [38]. Therefore in this report costs will be calculated for the diffuser type.

Because ozone is unstable and cannot be stored successfully, it must be generated at the point of application. Most simply, ozone can be generated by passing oxygen, or air containing oxygen, through an area having an electrical discharge or spark. To generate a sufficient quantity of ozone for a wastewater treatment plant, ozonators developing a corona discharge are used. These ozonators have two large metal electrodes separated by an air gap. An alternating electric current is applied to the electrodes creating an electrical discharge. At the same time air or oxygen is passed through the air gap. As the air or oxygen flows through the air gap and the electrical discharge, a portion of the oxygen is converted to ozone [56].

A side product from the corona discharge is the generation of a large amount of heat. The air or oxygen flow in the air gap is not large enough to cool the electrodes. Since high temperatures cause ozone to very rapidly decompose to oxygen, it is necessary to provide a cooling system for the electrodes. The formation of oxides of nitrogen also takes place in the corona discharge. Oxides of nitrogen react with water to form nitric acid which would in time attack the materials inside the ozonator. To avoid this problem and extend the useful life of the ozonator, the air or oxygen flowing through the air gap in the ozonator must be moisture free. This can be accomplished by cooling or drying the gas to remove the moisture before allowing it to enter the air gap [56].

The Maximum Allowable Concentration (MAC) of ozone in the Netherlands in air is 0.06 ppm by volume for continuous human exposure during 8 hours per day, 5 days a week. The maximum exposure during 15 minutes is 0.3 ppm \(^2\). The threshold odor of ozone is 0.01 ppm. This means a person working near an ozone-handling area should be able to detect the presence of ozone at levels far below the MAC. The odor of ozone has been described as

\(^2\) Databank Grenswaarden Stoffen op de Werkplek (GSW)
similar to that of cloves, new mown hay, nitric acid, etc., depending on the concentration. Concentrations greater than 1 ppm are extremely pungent and are considered unsafe for prolonged human exposure, and therefore should be avoided. In accordance to worker safety guidelines, ozone detectors and warning systems should be present in buildings and installations, where ozone is produced and used in large quantities.

3.2  Activated Carbon

3.2.1  Functionality

Activated carbons are produced from materials including wood, coconut shell, peat, lignite, bituminous coal and petroleum residues. A schematic is given in figure 6. Treatment is generally produced from medium volatile bituminous coal or lignite. The carbon medium is “activated” by subjecting it to steam and high temperatures (circa 1200 °C). The effectiveness of activated carbon for the removal of organic pollutants from water by carbon adsorption is enhanced by its large surface area, an important factor in the adsorption process. The surface area of activated carbon typically can range from 500 to 1400 m^2/g [58].

![Figure 6 - Schematic illustrating the adsorption onto activated carbon particle [52]](image)

Granular activated carbon (GAC) has been used as a common measure for drinking water purification in the past and has also been studied in WWTP’s. Advantages of GAC include its simple application and the possibility for regeneration/reuse of exhausted GAC. However, GAC efficiency might be significantly reduced by the presence of competing organic matter in WWTP effluents. Alternatively, powdered activated carbon (PAC) can be applied in a tertiary treatment step or dosed directly to the biological stage of a WWTP. Due to its smaller particle size, PAC is typically superior in regard to adsorption kinetics and is more efficient compared to GAC [58].

Activated carbon has been tested in numerous applications as an advanced wastewater treatment step. As substances attach to the surface of the adsorbent, these adsorbed compounds are completely removed from the wastewater. If more than one contaminant is present, hydrophobic contaminants are easily and strongly adsorbed to the carbon and will be removed in greater quantities than contaminants that are less adsorbed. This is called competitive adsorption. Micropollutants which are not well removed by WWTP are mostly hydrophilic, which are less adsorbed than hydrophobic substances. The removal rate of these
hydrophilic substances by activated carbon adsorption is therefore greatly influenced by the presence of organic matter [47] [58].

Concerning competition with organic matter, removal efficiencies of PAC are mostly influenced by Dissolved Organic Carbon (DOC) [38] [39] [43] [44] [47] [49] [51], whereas removal efficiencies of GAC are influenced by both DOC and particulate organic matter. DOC will compete with micropollutants for the available adsorption surfaces while particulate organic matter will clog the macropores of the GAC, through which the adsorption surfaces cannot be reached, see figure 6 [47] [51].

3.2.2 Operation and maintenance

**Powdered activated carbon (PAC)**

Most activated carbon research on a larger scale is the treatment of wwtp effluent through a separate system consisting of a contact tank, settling tank and a sand filter. In the contact tank PAC, polymers and metal solutions are dosed. The sludge form the settling tank is recycled to the contact tank. Optionally it can also be recycled to the aeration tank. Because of remaining PAC particles a post treatment to separate these PAC particles from the wwtp effluent is necessary through e.g. sand filtration (see figure 7).

![Figure 7 - PAC-treatment of wwtp effluent; adapted from [32]](image)

Alternatively, PAC can be dosed in the aeration tank of wwtp or in the inlet of already existing sandfilters. These implementations reduce the investment costs. As the separate system with a PAC contact tank is most researched in full scale as depicted in figure 7, the costs of this system are further investigated in this report.

PAC is normally stored in silos which have a minimum size of a lorry transport. From this silo, the PAC is fed into a dosage system, where it is mixed with water. The dissolved PAC is then dosed into the contact tank. Clogging of the carbon slurry transport pipes can occur. The problem is mostly caused by undersized piping, short radius bends, insufficient velocity, and lack of cleanouts in the carbon transport system. Abrasion wear of slurry transport pipes is also a common problem in unlined mild steel and fiberglass reinforced plastic (FRP) piping, particularly at sharp bends. Increasing the size of the piping (a minimum pipe diameter of 2 inches is recommended), transporting a more dilute carbon slurry, using long radius piping, and providing a sufficient number of cleanouts helps to minimize the clogging problem. Abrasion of the pipes can be reduced significantly by using glass or rubber lined steel piping or coated cast iron piping for carbon slurry transport. The use of long radius piping and extra-heavy elbows and tees is recommended [58].
Due to the high surface area and porosity of powdered activated carbon, it can react with oxygen, releasing heat. This means the PAC can cause explosions under conditions, where the PAC is forming dust and sparks can occur. The storage and dosage system should therefore be designed in accordance to Ex-regulations, with proper monitoring equipment. The formation of dust should be avoided through airtight installation design and appropriate air treatment [58].

By dosing PAC, polymers and metal solutions to the wastewater effluent, the sludge production is increased by approximately 10% -20% at normally applied dosages of 10-20 mg PAC/l.

**GAC**

For GAC treatment, a filtration step like a normal sand filtration is realized in a fixed bed filter. Only the filter is filled with Granular Activated Carbon instead of sand or anthracite. The granular carbon remains stationary in a cylindrical tank while the water flows downward through the granular medium under the force of gravity and is removed from the bottom. Pressurized air is added to the incoming water, to enhance biological degradation. The GAC filter has to be flushed periodically to remove organic matter and prevent blockage. This so-called “backwash water” has to be treated at the wastewater treatment plant (see figure 8). Periodically the GAC has to be replaced, once the removal efficiency of the targeted compounds begins to drop.

![Figure 8 - GAC-treatment of wastewater effluent](image)

Operation and maintenance of the GAC filter are relatively simple. When all adsorption sites on the activated carbon are filled with contaminants, the filter is saturated and has reached its capacity. At this point, contaminants may not be adsorbed and some may move from the carbon back into the water. This is called breakthrough since the contaminants “break through” the filter and enter the treated water. The most time consuming task is when the spent carbon has to be removed after this breakthrough by shutting down and draining the tank, and new or reactivated carbon is added as a new batch. Drainage systems should be well designed to perform this task in a comfortable manner[58].

Clogging of backwash and surface wash nozzles can be a problem. This is caused by migration of carbon and solids to the underdrains where they are picked up by the incoming backwash water and clog the distribution nozzles. Screens installed at the bottom of the carbon bed prevent media migration to the underdrains. Frequent backwashing, especially after loading the carbon, removes the fines from the bed, thus decreasing the clogging of the nozzles. Organic particles can clog the GAC filter easily. If the settling tank of the wastewater treatment plant
not function well, which means particle concentrations of more than 10 mg/l are present in the effluent, the GAC-filter should be bypassed. If clogging of the GAC-filter occurs and backwashing with water is not solving this problem, the GAC-filter can be flushed with pressurized air [58].

3.3 Effectiveness of ozonation, PAC and GAC treatment of wwtp effluent

Implementation of a post treatment of wwtp effluent with ozonation, PAC or GAC are all capable of removing most micropollutants to a similar degree. Zoomed in at different substances, the techniques differ in removal rates because of their functionality in combination with the properties of the substances which have to be removed.

Typical removal rates of selected compounds during ozone, PAC and GAC treatment are illustrated in figures 9, 10 and 11 respectively. As can be seen from figures 9 and 10 the dosage of ozone and PAC influences the removal rate of the different micropollutants. For GAC the removal rate depends on the service life of the activated carbon (this is the time in between replacement of the activated carbon). For both ozone and activated carbon the amount of organic matter also influences the removal rates. The reasons why this occurs were explained under paragraphs 3.1.1 for ozone and 3.2.1 for activated carbon. Next to ozone and PAC dosages and standing time of GAC filters, there are several other important design criteria, which influence the removal efficiencies of different micropollutants. These are further discussed in chapter 4.

![Figure 9 - Typical removal efficiencies of substances from wwtp effluents at different ozone dosages [31]](image-url)
Figure 10 - Typical removal efficiencies of substances from WWTP effluents at different PAC dosages [49]

Figure 11 - Typical removal efficiencies of substances from WWTP effluents; standing time GAC 6 months [48]
4 Design criteria and operating conditions

4.1 Removal efficiencies of micropollutants

As explained in chapter 3, the removal efficiencies differ considerably for different substances and techniques and are moreover influenced by operating conditions and design criteria. A higher ozone or PAC dosage will lead to a higher removal efficiency as does a more often replacement of GAC for most micropollutants. To accurately design a post treatment and to estimate the costs, a very important design criterium is the minimum removal efficiency, which should be obtained for the different micropollutants.

Minimum removal efficiencies or effluent demands for all micropollutants in effluents of wwtp have not been decided upon in Germany. Different substances are removed in different degrees through different treatments at the different wwtp. For metabolites and transformation products, the situation is unknown, since it is unknown which intermediates are formed.

For cost estimates in Switzerland five substances have been chosen for which a minimum removal of 80% should be achieved [32]:

- Diclofenac (painkiller)
- Carbamazepine (anticonvulsant)
- Sulfamethoxazole (antibiotic)
- Benzotriazole (corrosion inhibitor)
- Mecoprop (herbicide)

These substances were chosen based on the following criteria [32]

- Presence in surface waters
- Use of the substances
- Wastewater composition
- Poor removal in wastewater treatment plants (wwtp’s)

Because of the absence of a minimum demand on removal efficiencies, post treatments in Germany in North Rhine-Westphalia (NRW) and Baden-Württemberg (BW) wwtp are designed differently. Up until 2014, the dimensioning of the installations is mostly determined by what can be achieved with available financing and with optimal use of the installations which are now in use and not used at the present time because of e.g. overcapacity. This is why many ozone experiments and PAC experiments are conducted in constructions which are partly out

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1 Based on the removal efficiency of the concentration in the outflow of the presettling tank and the effluent of the posttreatment: biological degradation of substances in the activated sludge system is thus incorporated in this removal rate. The removal rate is based on incoming and outflowing yearly average loads.

4 In December 2014 a new proposal on indicator substances has been issued by BAFU [67]. In this proposal 12 substances are mentioned. These substances are divided into 2 groups: 1. very well removable substances and 2. well removable substances. Group 1 consists of Amisulprid, Carbamazepine, Citalopram, Clarithromycin, Diclofenac, Hydrachloridthicid, Metoprolol, Venlafaxin. Group 2 consists of Benzotriazole, Candesartan, Irbesartan, Mecoprop. An average removal rate of more than 80% (based on yearly average incoming and outflowing loads) must be established for a minimum of 4 substances of group 1 and a minimum of 2 substances of group 2. Presumably this new approach will be implemented by summer 2015.
of use, like sand filters. Removal efficiencies of substances are not predetermined, but measured as the research is carried out. So ozone and PAC dosages differ significantly per project. Per wwtp an assessment is made of guide parameters to be monitored. For the future, in NRW efforts are made to find a more consistent approach by the Ministry for Climate Protection, Environment, Agriculture, Conservation and Consumer Protection of the State of North Rhine-Westphalia (MKULNV) [41][55] .

For this report it is important to give an insight in and fix the removal efficiencies which can be obtained through different post treatment techniques and the costs involved. To do this, data has to be available on removal efficiencies, but also on the height of investment and operational costs of the different techniques to establish these removal efficiencies. In this study, it was found that throughout all the Swiss and German research, the removal of a limited number of substances has been extensively researched for the different techniques ozonation, PAC and GAC on a large pilot scale ( > 200 m³/h) or full-scale (see table 2).

**Table 2 - Extensively researched substances in Germany NRW and BW and Switzerland**

<table>
<thead>
<tr>
<th>Painkillers/ antiinflamm</th>
<th>Betablockers</th>
<th>Antibiotics</th>
<th>X-ray contrast media</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ibuprofen</td>
<td>Carbamazepine</td>
<td>Ciprofloxacin</td>
<td>Amidotrizoic acid</td>
<td>Corrosion inhibitor and de-icing agent: Benzotriazole; metabolites 4-Methylbenzotriazole and 5-Methylbenzotriazole</td>
</tr>
<tr>
<td>Diclofenac</td>
<td>Metoprolol</td>
<td>Clarithromycin</td>
<td>Lopamidol</td>
<td>Herbicide: Mecoprop</td>
</tr>
<tr>
<td>Moxifloxacin</td>
<td>Bisoprolol</td>
<td>Metronidazol</td>
<td>Diatrizoic acid</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sulfamethoxazole; metabolite N4-Acetylsulfamethoxazole</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The removal efficiencies reported in the studies mentioned in table 2 are strongly influenced by operating and design conditions. To estimate costs of the post treatments, fixed assumptions on these operating and design conditions for different techniques have to be established. The goal is to fix these conditions in such a way, that comparable removal efficiencies for the substances mentioned in table 2 are achieved. Therefore design criteria operating conditions and removal efficiencies were compared for the pilot scale studies and full scale constructed installations. Then latest insights and experience on design criteria in Germany and Switzerland were combined with this knowledge. The result is given in table 3 based on [33][38][39][41][42][50][65] and expert judgement.

**Table 3 - General design criteria in Germany and Switzerland for removal of micropollutants from municipal wwtp effluent.**

<table>
<thead>
<tr>
<th>Subject</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ozonation</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dosage ozone</td>
<td>g O₃ / g DOC</td>
<td>0.6 – 0.9</td>
</tr>
<tr>
<td>Dosage ozone</td>
<td>mg O₃/l⁺</td>
<td>4 – 14</td>
</tr>
<tr>
<td>Hydraulic Retention Time Contact Tank</td>
<td>minutes</td>
<td>15 - 30</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(reactor 10-25 min; Removing remaining ozone 5 min)</td>
</tr>
<tr>
<td>Power consumption</td>
<td>kWh/kg O₃ * h</td>
<td>10</td>
</tr>
<tr>
<td>Power consumption</td>
<td>W/treated m³</td>
<td>45</td>
</tr>
</tbody>
</table>
Based on the available data which have been compiled in the references shown in tables 2 and 3, it is now possible to calculate costs for removal of micropollutants from effluents of WWTP as follows. The working method, which has been followed in this study to do this, is visualized in figure 12.

1. First removal rates, which presumably can be established for the different substances are fixed under specific design and operating conditions as summarized in tables 4,5 and 6. These operating conditions are chosen based on the latest insights on average design parameters for full scale plants, under which it should be possible to achieve the removal rates mentioned in tables 4 to 6 [30] [31] [32] [38] [39] [40] [41] [42] [43] [44] [48] [49] [50] [51] [59] [60]. These removal rates are based on the concentrations in the outflow of the presettling tank and the effluent of the posttreatment including sand filtration. Biological degradation of substances in the activated sludge system is thus incorporated in this removal rate.

2. Secondly, the design flow is fixed for the Dutch situation (see chapter 4.2).

3. Thirdly, investment costs are calculated based on dimensioning of civil structures like (settling) tanks and piping, mechanical equipment like pumps, ozone generators, mixing and aeration devices and electrical and automation equipment. Operational costs are calculated based on assumptions on power consumption, consumption of chemicals and/or activated carbon, sludge production and costs for sludge processing, required personell and maintenance etcetera (for cost assumptions see chapter 5.1).

4. Finally, as an example costs are calculated for three scales of wastewater treatment plants which are common in the Netherlands: 20.000 p.e, 100.000 p.e and 300.000 p.e.
The removal rates mentioned in figure 12, are based on interpretation of data of references mentioned in table 2 and the design criteria in table 3, which result in the following fixed average operating conditions:

- **Ozonation**: dosage 0.7 g O$_3$/g DOC or 7.7 mg O$_3$/l; total retention time contact tank 25 minutes
- **PAC**: dosage 1.1 g PAC/g DOC or 12 mg/l; total retention time contact tank 35 minutes
- **GAC**: Empty Bed Contact Time: 30 minutes and standing time coal 6 months (bedvolumes: 8.800)

These fixed operating and design conditions lead to comparable removal efficiencies for the selected substances as displayed in tables 4-6 for the different techniques.

Assumed DOC concentration wwtp effluent: 11 mg/l
### Table 4 – Assumed removal rates ozonation + sand filtration*

<table>
<thead>
<tr>
<th>&lt; 30%</th>
<th>30-60%</th>
<th>60-80%</th>
<th>&gt; 80%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diatrizoic acid</td>
<td>Ciproflaxine, Iopamidol, Mecoprop, Ibuprofen</td>
<td>Benzetrazol, Methyl-benzetrazol, Metoprolol</td>
<td>Carbemepazepin, Clarithromycine, Diclofenac, Sulfametoxazol, Acetylsulfametoxazol, 17β estradiol</td>
</tr>
</tbody>
</table>

* Removal rates based on the outflow of the presetting tank and the effluent of the posttreatment and interpreted data from [30][31][32][38][39][40][41][42][44][50]; ozondosage 0,7 g O₃/g DOC, contact time 25 minutes and other design criteria as mentioned in table 3; assumed DOC concentration wwtp effluent: 11 mg/l

### Table 5 – Assumed removal rates PAC + sand filtration*

<table>
<thead>
<tr>
<th>&lt; 30%</th>
<th>30-60%</th>
<th>60-80%</th>
<th>&gt; 80%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diatrizoic acid</td>
<td>Iopamidol, Acetyl-sulfametoxazol, Mecoprop</td>
<td>Ciproflaxine, Diclofenac, Sulfametoxazol</td>
<td>Carbemepazepin, Clarithromycine, Ibuprofen, Metoprolol, 17β estradiol, Benzetrazol, Methyl-benzetrazol</td>
</tr>
</tbody>
</table>

* Removal rates based on the outflow of the presetting tank and the effluent of the posttreatment and interpreted data from [30][32][38][39][40][41][43][44][49][50]; PAC-dosage 12 mg/l or 1,1 g PAC/g DOC, contact time 35 minutes and other design criteria as mentioned in table 3; assumed DOC concentration wwtp effluent: 11 mg/l

### Table 6 – Assumed removal rates GAC*

<table>
<thead>
<tr>
<th>&lt; 30%</th>
<th>30-60%</th>
<th>60-80%</th>
<th>&gt; 80%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diatrizoic acid</td>
<td>Iopamidol, Acetyl-sulfametoxazol, Sulfametoxazol, Ciproflaxacine</td>
<td>Diclofenac, Mecoprop, Ibuprofen</td>
<td>Carbemepazepin, Clarithromycine, Metoprolol, 17β estradiol, Benzetrazol, Methyl-benzetrazol</td>
</tr>
</tbody>
</table>

* Removal rates based on the outflow of the presetting tank and the effluent of the posttreatment and interpreted data from [32][40][41][42][48][50][51][59][60], max bed volumes GAC 8.800 and other design criteria as mentioned in table 3; assumed DOC concentration wwtp effluent: 11 mg/l

At the same DOC-content of the wwtp effluent, for substances which are now in the “60-80% removal range”, a higher removal rate > 80% can be achieved by dosing more ozone or PAC or by decreasing the amount of treated bed volumes for GAC. The same applies for the substances in the “30-60% removal range”; it is assumed that they will be removed for 60-80%.

As explained in chapter 3, the removal rates of PAC, GAC and ozonation are strongly influenced by the amount of DOC in the wwtp effluent. The higher the DOC content, the higher the dosages of PAC or ozone must be to achieve comparable removal rates. For GAC not only dissolved organic matter, reduces the effectiveness of the GAC considerably but also organic particulate matter.

Increasing the removal rates of substances (higher ozone and PAC dosage and lower amount of GAC bed volumes) and/or a higher DOC-content than assumed in tables 4, 5 and 6 will lead to
higher operational costs. A sensitivity analysis on the calculated costs will therefore be conducted in chapter 5, to quantify these influences.

4.2 Treated wwtp effluent flow

The dimensioning of the wwtp effluent flow, which has to be treated is heavily discussed in Germany. If the wwtp treats wastewater coming from a combined sewer system, then the rain weather flow can be up to 4 times as high as the dry weather flow in the Netherlands. Per European region this can differ considerably. In the Netherlands a RWF/DWF ratio\(^6\) of 3 is very normal, whereas in Switzerland this is on average lower than 2 and in Germany it differs a lot per region. As this RWF only occurs in the Netherlands about 10-15% of the time of the year, the investment costs are generally considered too high to treat this. Especially because after about 2-4 hours the incoming flow will be very diluted. The first couple of hours however, micropollutants are present in the RWF in assumably high numbers and concentrations. In the Netherlands this is called “The First Flush”.

To date there is no agreement on the amount of wwtp effluent which has to be treated in Germany. In Switzerland the removal efficiencies are calculated based on yearly loads of the effluent of the wwtp and the outflow of the presettling tank. In the first Swiss full scale installation (wwtp Neugut) it was chosen to design the retention time of the ozonation contact tank on dry weather flow and monitor if the lower contact time under rain weather flow is sufficient to remove the selected micropollutants \(^7\) to more than 80%. These results are not known yet at publication of this report.

The DWF should be treated, every expert agrees on that. But still this leaves margins for discussion. One can design the post treatment on the average DWF (24h-average or 16h-average between 07.00 and 23.00h) or the peak of the DWF. Or one can make a frequency graph of the total flow coming in and then select the wastewater flow which occurs for 70 or 80% of the time of a year (again aiming at treating the DWF). The different principles and terms are very confusing as they do not match up internationally. Furthermore at the large pilot scale installations, another approach was chosen in Germany, to treat that amount of flow, which could be handled by existing process units which were not in use, like sand filters, buffer tanks, etc..

For the estimation of costs, the treated flow of wwtp effluent is a very important parameter, as it strongly influences investment and operational costs. To estimate these costs, a well-defined design flow must be chosen. For this report, it is chosen to follow Dutch design rules for other wwtp installations, which have to treat Dry Weather Flow (DWF). These are mostly designed on the design peak of the dry weather flow. This design peak of the dry weather flow corresponds to roughly 115% of the actual dry weather flow peak (see figure 13). By treating this amount, around 80% of the total incoming wastewater amount, will be treated \(^8\).

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\(^6\) RWF = Rain Weather Flow; DWF = Dry Weather Flow
\(^7\) Diclofenac, Carbamazepine, Sulfamethoxazole, Benzotriazole, Mecoprop
\(^8\) Based on a RWF/DWF ratio of 3
4.3 Process automation

Ozonation and PAC

Ozone dosage can be adjusted based on the incoming flow and the ozone content of the off-gas. PAC dosages are mostly adjusted only on the incoming flow to the post treatment. Based on these parameters dosages of ozone or PAC which are too high or too low, can easily occur. As discussed in chapter 3, the effectiveness of ozonation and PAC are strongly influenced by the DOC-concentration of the effluent. This means that the dosage of ozone and PAC not only has to be adjusted on the amount of wastewater flowing through the post treatment, but ideally also on the DOC-concentration of to be treated effluent.

Measuring DOC-concentration online at full scale installations proves to be very difficult and is not accurate enough to control the ozone dosage. Instead of measuring this, the loss of UV light absorption at 254 nm (UVA\textsubscript{254}) seems to be a suitable control parameter and is investigated at several wwtp. A close correlation between relative removal of micropollutants with ozone / PAC and corresponding reduction of UVA\textsubscript{254} has been reported in various studies [44][46]. As UVA\textsubscript{254} is easy to measure online, the parameter can be used as a surrogate for the removal of micropollutants, through which the right dosage can be applied. The results of the accuracy of controlling ozone and PAC dosages through UVA\textsubscript{254} are inconclusive. Retention time in the contact tank of the ozonation or PAC seems to influence the reduction in UVA\textsubscript{254} too, as well as removal of micropollutants. As the retention time in the contact tanks for ozone and PAC differ depending on the incoming flow, this is a difficult issue to cope with. Further research is needed to come up with an accurate answer to control the ozone and PAC dosage [38][41][42][50].

Figure 13 - Typical pattern of Dry Weather Flow during the day of a Dutch wwtp (Mulder, M. 2015; [68])
**GAC**

The control of a GAC filter is based on maintaining a continuous upflow velocity through the filter and thus volume based. Clogging of the GAC-filter can be measured through pressure differences, on which a water or air flushing can be automatically started. At certain time intervals the influent and effluent concentrations of micropollutants must be measured to establish the break-through patterns and the necessity to replace the activated carbon.
5 Estimated costs

5.1 General cost principles

Based on the information given in chapters 3 and 4, investment and operational costs are calculated for a post treatment with ozonation, PAC and GAC-treatment, for the following three scales of wwtp in the Netherlands:

- Small wwtp of 20,000 p.e. 150 g TOD\(^9\), design flow post treatment 200 m\(^3\)/h
- Average wwtp of 100,000 p.e. 150 g TOD, design flow post treatment 1,050 m\(^3\)/h
- Large wwtp of 300,000 p.e. 150 g TOD, design flow post treatment 3,100 m\(^3\)/h

The design flow is based on treating 115% of the actual dry weather flow peak, through which around 80% of the total incoming wastewater amount, will be treated\(^10\) (see chapter 4.2).

For the investment costs it is assumed that the post treatment step has to be fully invested for with the exception of the availability of land. This means that at the wwtp there is space to build a post treatment, but that there are no facilities for a post treatment like out of order structures, piping, water or sludge buffers, pumps and so on. It is assumed that the effluent of the wwtp has to be pumped for 200 meters in length and up to 5 meters in height for all techniques (Ozonation, PAC and GAC). For the electrical and automation installations it is assumed that there is no spare room in the available electrical power supply or automation equipment and that new transformers and cables for electricity have to be realized.

Investment and operational costs are furthermore calculated based on the design criteria in table 3 and the following average operating conditions (see chapter 4.2):

- Ozonation: dosage 0.7 g O\(_3\)/g DOC or 7.7 mg O\(_3\)/l\(^{11}\); total retention time contact tank 25 minutes
- PAC: dosage 1.1 g PAC/g DOC or 12 mg/l\(^{10}\); total retention time contact tank 35 minutes
- GAC: Empty Bed Contact Time: 30 minutes and standing time coal 6 months\(^9\)
  (bedvolumes: 8,800)

Other cost principles are:

- Investment costs
  - Technical lifetime
    - civil works: 30 years
    - machinery and electrical equipment: 15 years
    - automation: 5 years
  - Interest: 4% 
  - Depreciation based on interest annuity
  - Accuracy of investment costs: ± 35%
- Realization and project costs: 65% of investment
  - Engineering: 12%
  - Insurances, permits and other building costs: 15%
  - Projectmanagement and construction supervision: 8%
  - Temporary installations during build and start-up: 5%

\(^9\) 1 p.e. 150 g TOD = (COD + 4,57*NKj)/150
\(^{10}\) Based on a RWF/DWF ratio of 3
\(^{11}\) Assumed DOC concentration wwtp effluent: 11 mg/l
• Training personnel: 2%
• Communication: 2%
• VAT: 21%
• Maintenance (% of investment)
  • civil works: 0.5%
  • machinery, electrical equipment and automation: 3%
• Yearly personnel costs
  • € 25,000 for a small wwtp (20,000 p.e.), € 50,000 for an average wwtp (100,000 p.e.) and € 75,000 for a large wwtp (300,000 p.e.)
• Variable costs (including VAT of 21%)
  • Electricity: € 0.10/kWh
  • Pure oxygen: € 0.20/kg
  • Powder Activated Carbon: € 1.500 / m³
  • Granular Activated Carbon: € 1.200 / m³
  • Coagulant: € 250 / ton
  • Polymer: € 3 / kg (40% active)
  • Sludge treatment (dewatering + thermal processing): € 350 / ton dry matter

Specific cost assumptions per post treatment technique:

Ozonation
• Contact time: 25 minutes
• Ozonation through diffusors
• Depth contact tank: 5 metres
• Ozone dosage: 0.7 g O₃ / g DOC
• Electrical consumption ozone generation: 10 kWh / kg produced ozone per hour
• Electrical consumption other equipment: 45 W/m³ treated wwtp effluent including sand filtration

PAC
• Contact time: 35 minutes
• Depth contact tank: 5 metres
• Surface load settler: 2 m/h
• PAC dosage: 1.1 g PAC / g DOC
• Recycle factor PAC: 0.8
• Dosage of coagulant: 5 mg/l
• Dosage of polymers: 0.2 mg 100% active/l
• Electrical consumption: 60 W/m³ treated wwtp effluent including sand filtration

12 Improvements of sludge dewaterability of the sludge have been reported if PAC is dosed and treated together with the wwtp secondary sludge, but they are inconclusive. So for the cost calculations the processing of the extra amount of PAC-sludge is taken into account at € 350 / ton dry matter
GAC
- Maximum TSS wwtp effluent: 10 mg/l
- Empty Bed Contact Time: 30 minutes
- Bed Height Activated Carbon: 2.5 m
- Filtrate back wash (20 m$^3$-h / treated m$^3$-h)
- Backwash water: 10%
- Electrical consumption: 40 W/m$^3$ treated wwtp effluent

Sand filtration
- Upflow velocity: 12 m/h
- Backwash water: 5%

1-STEP© filtration
- Replacement of coal every 12 months
- Upflow velocity: 10 m/h
- Backwash water: 10%
- Further investment and operational costs according to [54]

Hence: The sand and 1-STEP filters are capable of removing phosphorus and nitrogen when respectively metal solutions and a carbon source are dosed. For these calculations these dosages including investments on storage and dosage units were not taken into account for the cost estimates.

5.2 Cost calculations

The investment and operational costs of ozonation, PAC and GAC-treatment are calculated based on the cost principles given in paragraph 5.1. The breakdown of the total realization, investment and operational costs is detailed in appendix 2.

To give an insight in the differences per technique the costs are divided by the total amount of treated effluent. These costs per treated m$^3$ effluent are given in figures 14, 15 and 16 for respectively ozonation, PAC and GAC treatment.
Figure 14 - Ozonation followed by sandfiltration: Costs per m³ treated WWTP effluent based on yearly costs including capital and operational costs.

Figure 15 - PAC followed by sandfiltration: Costs per m³ treated WWTP effluent based on yearly costs including capital and operational costs.
Based on comparable removal rates for micropollutants, it can be concluded from figures 14-16 that ozonation of effluent of wwtp is less expensive than PAC-treatment, both followed by sand filtration. GAC-treatment is most expensive. The differences in costs can be explained as follows:

- The investments costs for ozonation and PAC-treatment are in the same range. The investment costs for GAC treatment are much lower than for ozonation and PAC-treatment, because of the simplicity of the GAC-installation.

- Although ozonation uses 2 times more energy than PAC-treatment and even 12 times more than GAC-treatment, the overall variable costs for ozonation are the lowest. For PAC this is caused by the extra costs for coal, chemicals and sludge treatment. For GAC-treatment this is caused by the extra costs for coal.

- The variable costs of GAC-treatment are 5 times higher per m³ than for PAC-treatment. This is mainly caused by the fact that the costs for coal in GAC-treatment are much higher than for PAC-treatment, even if the extra costs for sludge processing are taken into account. PAC is more effective per g of coal. Also the GAC has to be replaced fully after 6 months, because some of the micropollutants break through, whereas other micropollutants still adsorb. Therefore more granular activated carbon is needed than powdered activated carbon, to achieve the same removal rates.

Figure 16 - GAC: Costs per m³ treated wwtp effluent based on yearly costs including capital and operational costs

<table>
<thead>
<tr>
<th>Capacity ruwi (i.e. 150 g TZW)</th>
<th>€/m³ treated effluent</th>
</tr>
</thead>
<tbody>
<tr>
<td>20,000</td>
<td>€0.30</td>
</tr>
<tr>
<td>100,000</td>
<td>€0.25</td>
</tr>
<tr>
<td>300,000</td>
<td>€0.20</td>
</tr>
</tbody>
</table>

- **Variable costs GAC**
- **Variable costs pumping**
- **Personell GAC**
- **Maintenance GAC**
- **Capital costs GAC**
5.3 Sensitivity analysis

The costs summarized in paragraph 5.2 are subject to variations, mainly due to

- Higher or lower DOC-concentration of wwtp effluent: 15 or 7 mg DOC/l instead of 11 mg DOC/l
- Higher demand for removal efficiencies: 1,0 g O3 / g DOC and 1,8 g PAC /g DOC instead of 0,7 g O3 / g DOC and 1,1 g PAC /g DOC
- Further removal of metabolites of ozonation: 1-STEP© filtration instead of sand filtration [54]

In this sensitivity analysis these factors are taken into account. The results are given in table 7.

Table 7 – Sensitivity analysis: Relative decrease or increase in costs

<table>
<thead>
<tr>
<th></th>
<th>Ozonation incl. sand filtration</th>
<th>PAC incl. sand filtration</th>
<th>GAC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average costs</td>
<td>100%</td>
<td>100%</td>
<td>100%</td>
</tr>
<tr>
<td>Higher or lower DOC</td>
<td>80% - 120%</td>
<td>85% - 115%</td>
<td>85% - 115%</td>
</tr>
<tr>
<td>Higher removal at same DOC</td>
<td>120%</td>
<td>115%</td>
<td>115%</td>
</tr>
<tr>
<td>Higher DOC and higher removal</td>
<td>140%</td>
<td>130%</td>
<td>130%</td>
</tr>
<tr>
<td>1-STEP® filtration instead of sand filtration after ozonation</td>
<td>135%</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Summarizing this paragraph, the costs mentioned in table 8 can be expected to implement a post treatment step for micropollutant removal at wwtp in the Netherlands, in which most of the micropollutants mentioned in table 2, except for the x-ray contrast media, will be removed for more than 30-80%.

Table 8 – Costs / treated m³ of wwtp effluent for micropollutant removal in the Netherlands; assumed DOC concentration 7-15 mg/l, average removal

<table>
<thead>
<tr>
<th>Capacity wwtp -&gt;</th>
<th>20,000 p.e.</th>
<th>100,000 p.e.</th>
<th>300,000 p.e.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ozonation + sand filtration</td>
<td>€ 0,22 ± € 0,04</td>
<td>€ 0,18 ± € 0,03</td>
<td>€ 0,16 ± € 0,03</td>
</tr>
<tr>
<td>PAC + sand filtration</td>
<td>€ 0,26 ± € 0,04</td>
<td>€ 0,20 ± € 0,03</td>
<td>€ 0,18 ± € 0,03</td>
</tr>
<tr>
<td>GAC</td>
<td>€ 0,29 ± € 0,04</td>
<td>€ 0,27 ± € 0,04</td>
<td>€ 0,26 ± € 0,04</td>
</tr>
</tbody>
</table>

If a higher removal is desired, in accordance with the latest guidelines on removal of different substances by Switzerland \[13\], the costs will be higher and are summarized in table 9.

---

\[13\] In December 2014 a new proposal on indicator substances has been issued by BAFU [67]. In this proposal 12 substances are mentioned instead of the substances mentioned in BAFU [32] being Diclofenac, Carbamazepine, Sulfamethoxazole, Benzotriazole, Mecoprop. The new substances are divided into two groups: 1. very well removable substances and 2. well removable substances. Group 1 consists of Amisulprid, Carbamazepine, Citalopram, Clarithromycin, Diclofenac, Hydrachloridthiacid, Metoprolol, Venlafaxin. Group 2 consists of Benzotriazole, Candesartan, Irbesartan, Mecoprop. An average removal rate of more than 80% (based on yearly average incoming and outflowing loads) must be established for a minimum of 4 substances of group 1 and a minimum of 2 substances of group 2. Presumably this new approach will be implemented by summer 2015.
Table 9 – Costs / treated m³ of wwtp effluent for micropollutant removal in the Netherlands; assumed DOC concentration 7- 15 mg/l; higher removal

<table>
<thead>
<tr>
<th>Capacity wwtp -&gt;</th>
<th>20.000 p.e.</th>
<th>100.000 p.e.</th>
<th>300.000 p.e.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ozonation + sand filtration</td>
<td>€ 0,26 ± € 0,05</td>
<td>€ 0,22 ± € 0,04</td>
<td>€ 0,19 ± € 0,03</td>
</tr>
<tr>
<td>PAC + sand filtration</td>
<td>€ 0,30 ± € 0,04</td>
<td>€ 0,23 ± € 0,04</td>
<td>€ 0,21 ± € 0,03</td>
</tr>
<tr>
<td>GAC</td>
<td>€ 0,33 ± € 0,05</td>
<td>€ 0,31 ± € 0,04</td>
<td>€ 0,30 ± € 0,04</td>
</tr>
</tbody>
</table>

Not all micropollutants will be removed by this post treatment. Persistent micropollutants, like x-ray contrast media and complexing agents like EDTA, will remain in the effluent. Also metabolites will not be removed at the same rate. To enhance the removal of the metabolites an activated carbon step can be implemented integrated in the sand filter as is being done in the 1-STEP© concept [54]. This step can be included after ozonation and will increase yearly costs by circa 35%.

5.4 Comparison with German and Swiss cost studies

First of all before any cost comparison can be made, one should realize that there are a few important differences between Dutch, Swiss and German cost and wastewater treatment structures:

1. The design capacity of a wwtp in population equivalents is calculated differently for the Dutch, Swiss and German waste water treatment plants. In Germany a calculation method is used based on the total inflow of BOD\textsuperscript{14}, in Switzerland based on COD\textsuperscript{15} and in the Netherlands based on COD an Nitrogen\textsuperscript{16}. These calculations methods lead to different design capacities based on population equivalents (p.e) (see table 10). This means that the costs calculated for a wwtp of 100.000 p.e in the Netherlands correspond to a German wwtp of 70.000 p.e. and a Swiss wwtp of 87.000 p.e (see table 10).

Table 10 – Difference in calculation of population equivalents

<table>
<thead>
<tr>
<th>The Netherlands Population equivalents</th>
<th>Germany Population equivalents</th>
<th>Switzerland Population equivalents</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 p.e 150 g TOD 1,0</td>
<td>0,70</td>
<td>0,87</td>
</tr>
<tr>
<td>20.000 p.e. 150 g TOD 20.000</td>
<td>14.000</td>
<td>16.000</td>
</tr>
<tr>
<td>100.000 p.e. 150 g TOD 100.000</td>
<td>70.000</td>
<td>87.000</td>
</tr>
<tr>
<td>300.000 p.e. 150 g TOD 300.000</td>
<td>210.000</td>
<td>261.000</td>
</tr>
</tbody>
</table>

2. Costs calculated in Swiss Francs should not be recalculated due to exchange rates, but rather to references in building costs. Based on the latest German cost study 1,00 euro for building and operating a wastewater treatment plant is equal to 0,50 Swiss Francs. [65]

5.4.1 Comparison with German cost studies

From tables 8 and 9 it can be concluded that the costs for a post treatment in the Netherlands for the removal of micropollutants are € 0,16 – 0,33 / m³ treated effluent, depending on the technique, the DOC-concentration of the wwtp effluent and the desired removal efficiencies of different substances.

\textsuperscript{14} 1 p.e in Germany = 60 g BOD
\textsuperscript{15} 1 p.e in Switzerland = 120 g COD
\textsuperscript{16} 1 p.e in the Netherlands = 150 g TOD = COD + 4,57 *NkJ
In Germany a recent study [65] calculated the costs for posttreament. In this study the costs for the removal of micropollutants are separated from the installations needed to build a post filtration treatment, including piping and pumping of effluents to the posttreatment and to the discharge point. The total costs for the removal of micropollutants were based on the large scale studies in Germany and show a large variation in costs (see figure 17.) The costs for the post treatment by a filter and the necessary costs for pumps and piping were based on expert judgement. The results are summarized in table 11.

Table 11 – Costs calculated in Germany for micropollutant removal per m³ incoming wastewater to the wwtp [65]

<table>
<thead>
<tr>
<th>Capacity wwtp Geman p.e (60 g BOD)</th>
<th>Micropollutant removal</th>
<th>Pumping and posttreatment in filter</th>
<th>Total costs</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.000 p.e.</td>
<td>0,108</td>
<td>0,08</td>
<td>0,188</td>
</tr>
<tr>
<td>35.000 p.e.</td>
<td>0,092</td>
<td>0,08</td>
<td>0,172</td>
</tr>
<tr>
<td>75.000 p.e.</td>
<td>0,079</td>
<td>0,08</td>
<td>0,159</td>
</tr>
<tr>
<td>150.000 p.e.</td>
<td>0,069</td>
<td>0,05</td>
<td>0,119</td>
</tr>
<tr>
<td>350.000 p.e.</td>
<td>0,059</td>
<td>0,05</td>
<td>0,109</td>
</tr>
<tr>
<td>750.000 p.e.</td>
<td>0,051</td>
<td>0,05</td>
<td>0,101</td>
</tr>
</tbody>
</table>

In this German study the whole incoming wastewater flow is taking into account, whereas mostly only 85% of the effluents of the researched wastewater treatment plants has been treated in the cost studies [65]. The costs per treated m³ of wwtp effluent are therefore on average 18% higher than shown in figure 17.

Furthermore one can see from figure 17 that there is a significant deviation from the average reported costs mentioned in table 11. The 95% confidence interval shows a variation in costs of ± € 0,08 per m³. Also the GAC-treatment has not been tested as much as ozonation and PAC-treatment and gives significantly higher costs than the average costs reported in table 11. Therefore, for the comparison of the costs reported in this report and [65], only ozonation and PAC-treatment are considered. Ozonation seems to be a little bit cheaper than PAC-treatment. These cost differences have also been concluded in this report (see paragraph 5.2 and 5.3).
Furthermore the achieved removal efficiencies in the German studies, do not always meet the removal efficiencies which have been taken into account to calculate the costs in tables 8 and 9 for the Dutch situation. As no legal or other design criteria were in place, one could choose at which PAC or ozone dosages the experiments were conducted. The explanation of these differences is beyond the scope of this report. Therefore only the difference between the calculation of population equivalents and treated wastewater amount is taken into account for this study and the German cost study. For the comparison the costs calculated for average removal as mentioned in table 8 are compared to the German costs. The difference in results is given in table 12.

Finally there are slight differences in the cost calculations for the Dutch and the German situation. The most influential cost variables are:

- Capital costs are 15% higher in the Netherlands than in Germany
- Electricity is 30% cheaper in the Netherlands than in Germany
- Labour is 50% more expensive in the Netherlands than in Germany
- Overall these factors imply that in the Dutch situation the costs of treatment of wastewater effluent is 10% higher than in Germany. This higher price level is neglectable if one takes the inaccuracy into account of the costs calculated in the Netherlands and Germany (see table 12).

From table 12 it can be concluded that the calculated costs in this report are well within the range of the calculated German costs in [65].

Table 12 – Comparison of calculated costs in Germany and the Netherlands for micropollutant removal including post treatment in a (sand)filter

<table>
<thead>
<tr>
<th>Capacity wwtp Dutch p.e.</th>
<th>Capacity wwtp German p.e.</th>
<th>Costs this study micropollutant removal + post treatment</th>
<th>Costs Germany study micropollutant removal + post treatment</th>
<th>Costs Germany study micropollutant removal + post treatment</th>
</tr>
</thead>
<tbody>
<tr>
<td>(150 g TOD)</td>
<td>(60 g BOD)</td>
<td>Per treated m$^3$ of wastewater effluent</td>
<td>Per treated m$^3$ of wastewater effluent</td>
<td>Per m$^3$ treating 85% of total incoming wastewater*</td>
</tr>
<tr>
<td>20,000 p.e.</td>
<td>14,000 p.e.</td>
<td>€ 0,22 - € 0,26 ± 0,05</td>
<td>€ 0,21 ± 0,08</td>
<td>€ 0,18 ± 0,08</td>
</tr>
<tr>
<td>100,000 p.e.</td>
<td>70,000 p.e.</td>
<td>€ 0,18 – € 0,20 ± 0,05</td>
<td>€ 0,19 ± 0,08</td>
<td>€ 0,16 ± 0,08</td>
</tr>
<tr>
<td>300,000 p.e.</td>
<td>210,000 p.e.</td>
<td>€ 0,16 - € 0,18 ± 0,05</td>
<td>€ 0,14 ± 0,08</td>
<td>€ 0,12 ± 0,08</td>
</tr>
</tbody>
</table>

* See figure 17 and table 11

5.4.2 Comparison with Swiss cost studies

In Switzerland cost scenarios are calculated in [66]. In this cost calculation it is stated, that implementation of a fourth step for the removal of micropollutants from effluents of wastewater treatment plants will cost 133 million Swiss Francs. This amount is based on:

- Removal efficiencies of indicator substances$^{17}$ as proposed by BAFU [32] > 80%.
- Post treatment of 80% of all Swiss wastewater which means that 4.5 million of Swiss population equivalents will be treated (design capacity at 120 g COD/p.e.).

The total costs are CHF 29.60 or € 14.30 per Swiss population equivalent (120 g COD/p.e).

$^{17}$ Diclofenac, Carbamazepine, Sulfamethoxazole, Benzotriazole, Mecoprop
For this study it was concluded that the costs for implementing a post treatment for the same indicator substances as issued by BAFU are € 0,16 – 0,20 / m³ treated effluent through treatment of ozonation or PAC \(^\text{18}\) for wwtp with a capacity of more than 100.000 p.e, see table 8. These costs can be calculated back to treated population equivalents based on the scenario’s calculated for the Dutch waste water treatment plants in 2011 [28]. Based on these scenarios the costs per p.e. (150 g TZV) can be calculated in comparison with the Swiss situation (see table 13).

Table 13 – Cost comparison Switzerland – The Netherlands for micropollutant removal per m³ incoming wastewater to the wwtp, based on removal of indicator substances as issued by BAFU 2012 (Diclofenac, Carbamazepine, Sulfamethoxazole, Benzotriazole, Mecoprop)

<table>
<thead>
<tr>
<th>Treated capacity: &gt; 80% of wastewater</th>
<th>Total costs</th>
<th>Costs per Swiss p.e (120 g COD)</th>
<th>Costs per Dutch p.e. (150 g TOD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4,500,000 p.e. CH</td>
<td>€ 66.5 million</td>
<td>€ 14,30</td>
<td>€ 12,40</td>
</tr>
<tr>
<td>13,500,000 p.e. NL</td>
<td>€ 150 – 190 million</td>
<td>€ 12,80 – 16,20</td>
<td>€ 11,10 – 14,10</td>
</tr>
</tbody>
</table>

The calculated costs for the Swiss and the Dutch situation are comparable. Especially if the amount of wwtp’s is taken into account, which already have a post treatment in the form of (sand) filtration. In the report for the Swiss calculations this was assumed at 40% of the total amount of the incoming wastewaters to Swiss wwtp’s [66]; in the Netherlands less than 10% of the wwtp effluent is post treated by a sand filter.

5.4.3 Other remarks

There are many more examples of cost calculations. One should be aware of the above mentioned factors, when translating these costs to the Dutch wastewater treatment practice. For example, in pilot studies, lower ozone and PAC dosages and lower replacement intervals of GAC are tested then are nowadays advised for the design. This will cut costs on investment and operation. Also cost calculations in Germany are based on the total amount of wastewater treated by the wwtp, which is generally more than treated by the post treatment plant. This can amount to 50% more, through which the reported costs are 50% lower if only the treated wastewater amount is taken into account.

Overall it can be concluded that the calculations done for the wwtp in the Netherlands correspond to the costs reported in German and Swiss literature, if differences in calculation methods of population equivalents, treated amount of effluent, use of already existing process units like sand filters and cost structures are accounted for.

\(^\text{18}\) Based on > 80% removal of indicator substances as proposed by BAFU[32]
6 Conclusions and recommendations

6.1 Considerations

The calculated costs for micropollutant removal from wastewater treatment plant (WWTP) effluent are highly dependent on:

1. Dissolved Organic Carbon (DOC)
2. Applied ozone and PAC dosage and replacement intervals of GAC
3. Treated wastewater flow

Ad 1. The DOC-content of Dutch WWTP effluent is not known. For this study it was assumed that the DOC-content is the same in the Netherlands as in Switzerland and Germany. The average DOC-content is fixed in this study on 11 mg DOC/l WWTP effluent. An increase to 15 mg DOC/l WWTP effluent increases costs by 15-20%, depending on the technique used.

Ad 2 The desired removal rates of different substances are not fixed for the Dutch situation. In Switzerland five guide parameters were defined for the cost estimates, for which removal through the WWTP should be more than 80% \(^{19}\). In Germany, a screening of to be removed micropollutants is first carried out. Next, the desired removal rate is established and then the appropriate technique and operating parameters are chosen.

Different techniques lead to different removal rates of different compounds. For example, the Swiss and German common guide parameter sulfamethoxazole can be removed well by ozonation (> 80%), but less by activated carbon (> 60%). Another common guide parameter, benzotriazole will be removed well by activated carbon (> 80%) and less by ozonation (> 60%) \(^{20}\). In both cases, an enhanced dosage of ozone or PAC or more frequently replacing GAC, will lead to higher removal rates, but also to higher costs. For the Dutch situation these guide parameters are not yet available. In this report often used Swiss and German guide parameters are taken into account.

Furthermore, for metabolites and transformation products the situation is unknown, as they can often not be measured. Studies on this subject are inconclusive. Tested circumstances are very different concerning ozone dosages, hydraulic retention times and influence of post treatment steps like sand filtration, natural lagoons and so on. By applying the current design criteria, transformation products will be formed through ozonation of WWTP effluent. This is inextricably linked to the functionality of ozonation. Whether these metabolites cause harm in the (aquatic) environment and if they are adequately enough removed through a biological sand filtration remains a topic of discussion. First experiments will be conducted in 2015 at ARA Neugut in Switzerland, comparing a sand filtration, GAC filter and fluidized sand filter after ozonation of WWTP effluent. For this report, costs are calculated based on ozonation followed by sand filtration. Another type of filtration with for instance activated carbon will increase costs.

Ad 3. In the Netherlands, many WWTP treat wastewater from combined sewage systems. Generally, after a dry period, a higher flush of contaminants will arrive at the WWTP during the first hours of a rain period. This is presumably also the case for micropolllutants. Because of the

---

\(^{19}\) Based on the removal efficiency of the concentration in the outflow of the presettling tank and the effluent of the posttreament: biological degradation of substances in the activated sludge system is thus incorporated in this removal rate.

\(^{20}\) Removal rates based on average design parameters as stated in chapter 4.2; tables 4-6
very high investment costs, post treatment of wwtp effluent is in this study based on treating the hourly peak of the Dry Weather Flow. This means that about 80% of the total amount of wastewater is treated for the Dutch situation.

6.2 Conclusions

The information on the design and operation of the large and full scale systems for the removal of micro pollutants from the effluent of wwtp in Germany and Switzerland is sufficient to give an insight in the costs involved in implementing post treatment of wwtp effluent for micropollutant removal in the Netherlands. The techniques which are extensively researched on a large scale on wwtp effluent are:

- Ozonation
- Powdered Activated Carbon (PAC) dosage
- Granular Activated Carbon (GAC) filtration

The estimated costs for implementing a post treatment of wwtp effluent for micropollutant removal in the Netherlands are summarized in table 14. Most of the micropollutants, except for persistent substances like x-ray contrast media, will be removed by this post treatment for more than 30-80%.

Table 14 – Costs / treated m³ of wwtp effluent for micropollutant removal in the Netherlands; assumed DOC concentration 7-15 mg/l, average removal

<table>
<thead>
<tr>
<th>Capacity wwtp -&gt;</th>
<th>20.000 p.e. 150 g TOD</th>
<th>100.000 p.e. 150 g TOD</th>
<th>300.000 p.e. 150 g TOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ozonation + sand filtration</td>
<td>€ 0,22 ± € 0,04</td>
<td>€ 0,18 ± € 0,03</td>
<td>€ 0,16 ± € 0,03</td>
</tr>
<tr>
<td>PAC + sand filtration</td>
<td>€ 0,26 ± € 0,04</td>
<td>€ 0,20 ± € 0,03</td>
<td>€ 0,18 ± € 0,03</td>
</tr>
<tr>
<td>GAC</td>
<td>€ 0,29 ± € 0,04</td>
<td>€ 0,27 ± € 0,04</td>
<td>€ 0,26 ± € 0,04</td>
</tr>
</tbody>
</table>

These costs are in correspondence with German and Swiss studies, when calculation methods of population equivalents, treated amount of effluent, re-use of existing process units like sand filters and differences in cost structures are taken into account, as well as design parameters on the treated effluent flow and operating parameters are standardized on dosages of ozone and PAC and bed volumes of GAC.

Removal efficiencies of micropollutants differ greatly per wwtp and per post treatment. Depending on the substance, the technique and the way in which the technique is implemented, different substances will have different removal rates. From the large scale and full scale systems for post treatment of effluent in Germany and Switzerland, it can be concluded that in general persistent micropollutants like complexing agents and x-ray contrast media will almost not be removed. Also metabolites and transformation products will be not be removed at the same rate.

For ozonation, the formation of toxic transformation products is a topic of discussion. In Germany and Switzerland it is advised to implement a biological sand filtration step after ozonation, to remove any biodegradable metabolites formed in the ozonation. Whether this sand filtration after ozonation is adequate enough is not known. For PAC dosage, sand filtration is necessary to remove small PAC particles, not because of the formation of metabolites. To reduce the risk of discharging toxic metabolites into the environment, the Dutch 1-STEP© concept can be implemented [54]. In this case the sand filter after ozonation will be filled with activated carbon, through which more metabolites and transformation
products presumably will be removed, but this will increase costs per treated m$^3$ of effluent by 35%.

6.3 Recommendations

1. The DOC-content of effluents of wwtp in The Netherlands is unknown. As the DOC-content significantly influences the costs of micropollutant removal from effluents, it is recommended to do research at (variations in) DOC-content of effluent of Dutch wwtp.

2. No guide parameters have been established for the Dutch situation, from which minimum removal efficiencies of different micropollutants can be determined. Different techniques and design criteria lead to different removal rates of different compounds. For the Dutch situation policy concerning guide parameters should be established through which post treatment of wwtp effluent can be accurately designed.

3. The costs in this report are based on a capacity for the posttreatment which is equal to the design peak of the Dry Weather Flow. This means that 15% of the total amount of yearly incoming wastewater will not be posttreated. It is not known to what extent micropollutants are present in this “Rain Weather Flow”. Research is therefore recommended on micropollutant concentration during Dry Weather Flow and Rain Weather Flow in the Netherlands and design criteria on the to be treated wastewater flow.

4. Through ozonation of wwtp effluent unknown transformation products are formed. It is recommended to do more research on the formation of these transformation product, their environmental impact and the effectiveness of different post treatments like sand filtration, 1-STEP®- or GAC-treatment.

5. An adequate control of PAC and ozone dosages can reduce costs. This can be done through the optimization of process automation. On this subject, research has just started on the full scale installations in Germany and Switzerland. More research is recommended to come up with an effective and efficient automation strategy.
References


[17] Figure by Mertsch, V. (2013)


[33] HydroIngenieure (2012), Machbarkeits Spurenstoffelimination Kläranlage Harsewinkel, Machbarkeitsstudie zur Erläuterungsbericht Düsseldorf, November 2012


[41] Unpublished information on design criteria, Kompetenzcentrum Mikroschadstoffe NRW (2015)


Zwickenpflug (2010), Einsatz von Pulveraktivkohle zur Elimination von Mikroverunreinigungen aus kommunalem Abwasser. Abschlussbericht, EAWAG Dübendorf, September 2010


http://water.me.vccs.edu


At November 17th 2014, the parliament of the Netherlands has agreed on a motion in which the government is requested to present an approach on the prevention and removal of residuals of pharmaceuticals in surface water before summer 2015.


Appendices

1. Overview research on the removal of micropollutants in Germany
2. Costs breakdown
Appendix 1 – Overview research on micropollutants in Germany

Based on information from

http://www.masterplan-wasser.nrw.de/karte/
http://www.koms-bw.de/klaeranlage/
Mikroschadstoffentfernung in kommunalen Kläranlagen in NRW
(Stand 01/2015)

Anzahl Kläranlagen in NRW

<table>
<thead>
<tr>
<th>Kategorie</th>
<th>Anzahl der Anlagen</th>
<th>Anschlussgröße (H)</th>
<th>Auslaufgröße (H)</th>
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<tr>
<td>&gt; 6 Mio.</td>
<td>293</td>
<td>329.920</td>
<td>93,3%</td>
</tr>
<tr>
<td>10 Mio. - 12 Mio.</td>
<td>64</td>
<td>17,731.580</td>
<td>22,952.168</td>
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<tr>
<td>&lt; 10 Mio.</td>
<td>894</td>
<td>27.935.090</td>
<td>35.986.915</td>
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<tr>
<td>Gesamt</td>
<td>1.096</td>
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<td></td>
</tr>
</tbody>
</table>

Die vollständige Legende befindet sich auf der nächsten Seite.

Die aktuelle Karte befindet sich auf www.masterplan-wasser.nrw.de
Mikroschadstoffentfernung in kommunalen Kläranlagen in NRW
(Stand 01/2015)

Großtechnische Untersuchungen

- **Aachen-Soers:** Abwasserozonation - Wasserverband Eifel-Rur
- **Bartrup:** Elimination von Mikroschadstoffen durch PAK und Abtrennung der Feststoffe unter Einsatz des Fuzzy Filters
- **Detmold:** Untersuchungen zum Einsatz von Ozon mit nachgeschaltetem GAK-Filter - nach der vorhandenen Filtration
- **Dinslaken:** Technikum auf der Kläranlage Emschermindeung
- **Dülmen:** Den Spurenstoffen auf der Spur - Untersuchungen des Aktivkohleinsatzes auf der KA (Teil 3) - Lippeverband
- **Düren-Merken:** Untersuchung an einer bestehenden Filterzelle mit dem Einsatz der Aktivkohle zur Entfernung organischer Restverschmutzung - Wasserverband Eifel-Rur
- **Düsseldorf-Süd:** Elimination organischer Spurenstoffe aus kommunalem Abwasser unter Einsatz von Aktivkohleschlammen aus Trinkwasserwerken - STEB Düsseldorf
- **Herford:** Untersuchungen zum Einsatz von Pulveraktivkohle unter Nutzung der vorhandenen Actiflow-Anlage
- **Köln-Rodenkirchen:** Umrüstung der Kölner BIOFOR-Filtrationsanlagen auf Spurenstoffelimination - Phase 1 - STEB Köln
- **Paderborn:** Untersuchungen zur Verfahrenskombination von Ozonierung und Aktivkohlefiltration unter Nutzung der vorhandenen Filtration
- **Wuppertal-Buchenhofen:** Technische Erprobung des Aktivkohleinsatzes zur Elimination von Spurenstoffen in Verbindung mit vorhandenen Filteranlagen - Wupperverband

Machbarkeitsstudien

- **Aachen-Soers**
- **Ahaus**
- **Ahlen**
- **Altenberge**
- **Bad Lippspringe**
- **Bad Oeynhausen**
- **Bartrup**
- **Bleifeld-Brake**
- **Borken**
- **Büren**
- **Detmold**
- **Drensteinfurt**
- **Duisburg-Hochfeld**
- **Dülken**
- **Emmerich**
- **Emsdetten-Austum**
- **Ennigenlohe**
- **Espelkamp**
- **Gescher-Harwick**
- **Greven**
- **Gütersloh**
- **Harsewinkl**
- **Heiden**
- **Herford**
- **Hopsten**
- **Höxter**
- **Ibbenbüren-Püsselbüren**
- **Isselburg**
- **Lage**
- **Legden**
- **Lemgo**
- **Lengerich**
- **Lichtenau-Grundstein**
- **Löhne**
- **Lübbecke**
- **Metelen**
- **Mettlingen**
- **Minden-Leteln**
- **Münster**
- **Münster-Geist**
- **Neubekum**
- **Neuenkirchen-Wettringen**
- **Neuss-Ost**
- **Obere Lutter**
- **Oelde**
- **Ochtrup**
- **Paderborn**
- **Rheina-Wiedenbrück**
- **Rheine-Nord**
- **Rietberg**
- **Saerbeck**
- **Salzkotten-Verne**
- **Sassenberg**
- **Sassenberg-Füchtorf**
- **Schöppingen**
- **Sendenhorst**
- **Stadtlohn**
- **Südlohn**
- **Velen**
- **Verl-Sende**
- **Verl-West**
- **Warburg**
- **Warendorf**
- **Wesel**
- **Wesseling**

Kläranlagenausbau

- **Aachen-Soers (in Planung)**
- **Bad Oeynhausen (in Planung)**
- **Bad Sassendorf**
- **Bartrup (in Planung)**
- **Detmold (in Planung)**
- **Duisburg-Vierlinden**
- **Dülmen (im Bau)**
- **Emmerich (in Planung)**
- **Espelkamp (in Planung)**
- **Gütersloh (Teilbetrieb)**
- **Harsewinkel (in Planung)**
- **Neuss-Ost (in Planung)**
- **Obere Lutter**
- **Rietberg (im Bau)**
- **Rheina (in Planung)**
- **Schwerte**
- **Warburg (in Planung)**

Die aktuelle Karte befindet sich auf www.masterplan-wasser.nrw.de
Kläranlagen mit einer Reinigungsstufe zur gezielten Spurenstoffelimination in Baden-Württemberg

Zustand
- in Planung
- in Bau
- in Betrieb

Verfahrenswahl
- Pulveraktivkohle
- granulierte Aktivkohle
- Ozon
- Verfahren offen

Hinweis:
JAM = Jahresabwassermenge

**Standort**

<table>
<thead>
<tr>
<th>Stadt</th>
<th>Einwohnerzahl (EW)</th>
<th>Q&lt;sub&gt;max. ads.&lt;/sub&gt;</th>
<th>behandelte JAM</th>
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<td>300 L/s</td>
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<td>Bönnau</td>
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<tr>
<td>Öhringen</td>
<td>46.700</td>
<td>150 L/s</td>
<td>&gt; 95 %</td>
</tr>
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<td>Hechingen</td>
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<td>&gt; 85 %</td>
</tr>
<tr>
<td>Sindelfingen</td>
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<td>&gt; 85 %</td>
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<td>Karlsruhe</td>
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<td></td>
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<tr>
<td>Freiburg</td>
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<td>Lahr</td>
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<td>440.000</td>
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<td>Lüchingen</td>
<td>35.000</td>
<td>150 L/s</td>
<td>&gt; 95 %</td>
</tr>
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<td>Öhringen</td>
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<td></td>
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</tr>
<tr>
<td>Emmingen-Liptingen</td>
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<td>Stockacher Aach</td>
<td>43.000</td>
<td>250 L/s</td>
<td>&gt; 85 %</td>
</tr>
<tr>
<td>Kressbronn</td>
<td>24.000</td>
<td>250 L/s</td>
<td>100 %</td>
</tr>
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**Projektsteckbrief**

Stand 02 / 2015
Appendix 2 – Costs Breakdown
Breakdown costs average removal and average DOC (11 mg DOC/l)

<table>
<thead>
<tr>
<th>Capacity wastewater (150 g TOD)</th>
<th>20,000</th>
<th>100,000</th>
<th>300,000</th>
</tr>
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<tbody>
<tr>
<td>Design capacity post treatment (m³/h)</td>
<td>200</td>
<td>1,050</td>
<td>3,100</td>
</tr>
<tr>
<td>Treated volume (m³/year)</td>
<td>1,140,000</td>
<td>5,980,000</td>
<td>17,660,000</td>
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</tbody>
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Realization costs

<table>
<thead>
<tr>
<th></th>
<th>1,300,000</th>
<th>5,800,000</th>
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<tbody>
<tr>
<td>Ozonation</td>
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<td>9,300,000</td>
<td>25,000,000</td>
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<tr>
<td>Ozonation + 1-STEP filtration</td>
<td>1,800,000</td>
<td>7,100,000</td>
<td>18,400,000</td>
</tr>
<tr>
<td>GAC</td>
<td>700,000</td>
<td>3,200,000</td>
<td>8,300,000</td>
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Yearly costs (capital + operational costs)

<table>
<thead>
<tr>
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<th>210,000</th>
<th>900,000</th>
<th>2,400,000</th>
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<tbody>
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<td>Ozonation</td>
<td>250,000</td>
<td>1,100,000</td>
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<tr>
<td>Ozonation + sand filtration</td>
<td>390,000</td>
<td>1,600,000</td>
<td>4,400,000</td>
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<td>Ozonation + 1-STEP filtration</td>
<td>300,000</td>
<td>1,300,000</td>
<td>3,300,000</td>
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<tr>
<td>GAC</td>
<td>330,000</td>
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Capital costs

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<td>230,000</td>
<td>880,000</td>
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<td>Ozonation + 1-STEP filtration</td>
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<tr>
<td>GAC</td>
<td>66,000</td>
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Maintenance costs

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Other operational costs

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Breakdown costs high removal and DOC (15 mg DOC/l)

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<tr>
<th>Capacity wwt (150 g TOD)</th>
<th>20.000</th>
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<th>300.000</th>
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<tr>
<td>Design capacity post treatment (m³/h)</td>
<td>200</td>
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<td>3.100</td>
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<tr>
<td>Treated volume (m³/year)</td>
<td>1.140.000</td>
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<td>17.660.000</td>
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</table>

**Realization costs**

<table>
<thead>
<tr>
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<th>Ozonation + sand filtration</th>
<th>Ozonation + 1-STEP©filtration</th>
<th>PAC + sand filtration</th>
<th>GAC</th>
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<td></td>
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<td>2.500.000</td>
<td>3.200.000</td>
<td>2.100.000</td>
<td>670.000</td>
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<td>11.000.000</td>
<td>12.900.000</td>
<td>8.400.000</td>
<td>3.000.000</td>
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<tr>
<td></td>
<td>25.100.000</td>
<td>29.100.000</td>
<td>34.600.000</td>
<td>22.200.000</td>
<td>8.000.000</td>
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**Yearly costs (capital + operational costs)**

<table>
<thead>
<tr>
<th></th>
<th>Ozonation</th>
<th>Ozonation + sand filtration</th>
<th>Ozonation + 1-STEP©filtration</th>
<th>PAC + sand filtration</th>
<th>GAC</th>
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</thead>
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<td>350.000</td>
<td>480.000</td>
<td>370.000</td>
<td>420.000</td>
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<td>1.600.000</td>
<td>2.100.000</td>
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<td>2.100.000</td>
</tr>
<tr>
<td></td>
<td>3.600.000</td>
<td>4.100.000</td>
<td>5.600.000</td>
<td>4.400.000</td>
<td>6.000.000</td>
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**Capital costs**

<table>
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<tr>
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<th>Ozonation</th>
<th>Ozonation + sand filtration</th>
<th>Ozonation + 1-STEP©filtration</th>
<th>PAC + sand filtration</th>
<th>GAC</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>210.000</td>
<td>240.000</td>
<td>480.000</td>
<td>200.000</td>
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</tr>
<tr>
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<td>900.000</td>
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**Maintenance costs**

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<th>Ozonation + sand filtration</th>
<th>Ozonation + 1-STEP©filtration</th>
<th>PAC + sand filtration</th>
<th>GAC</th>
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**Other operational costs**

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