Advanced treatment of WWTP effluent; no use or reuse?


* Department of Sanitary Engineering, Faculty of Civil Engineering and Geoscience, Delft University of Technology, P.O. Box 5048 GA Delft, The Netherlands (E-mail: s.m.scherrenberg@tudelft.nl; j.h.j.m.vandergraaf@tudelft.nl)
** Witteveen + Bos Consulting Engineers, P.O. Box 233, 7400 AE Deventer, The Netherlands (E-mail: h.menkveld@witteveenbos.nl; d.schuurman@witteveenbos.nl)
*** The Rijnland District Water Control Board, P.O. Box 156, 2300 AD Leiden, The Netherlands (E-mail: jeffrey.elzen@rijnland.net)

Abstract From 2006 until 2008 a research project is executed at wastewater treatment plant (WWTP) Leiden Zuid-West (The Rijnland District Water Control Board). The research focus is on the removal of nitrogen, phosphorous, heavy metals and priority hazardous substances from WWTP effluent with various treatment techniques to reach the effluent quality which is required by the Water Framework Directive 2000/60/EC. The semi-practical installation at WWTP Leiden Zuid-West consists of small full scale installations and has a maximum capacity of 150 m$^3$/h. The installation is divided into two parallel streets. The first street consists of flocculation tanks and a continuous sand filter. The second street consists of a continuous sand filter, flocculation tanks and a dual media filter. The continuous sand filters are denitrifying filters. The results of this research up to now show that continuous sand filtration has the ability of removing total nitrogen and total phosphorous to MPR values (maximum permissible risk) at high filtration rates. This means that continuous sand filtration is suitable as pre-treatment for ultrafiltration and reverse osmosis to produce ultra pure water. The advanced treatment of WWTP effluent is not only good to reach a better water quality but makes also the possibility of reuse easier.

Keywords Advanced treatment; WWTP effluent; Reuse; Nutrients removal; Water Framework Directive; Dual Media Filter; Continuous Sand Filter

INTRODUCTION

From 2006 until the end of 2008 an extensive research project is executed at wastewater treatment plant (WWTP) Leiden Zuid-West (The Rijnland District Water Control Board). The research focus is on the removal of nutrients (nitrogen and phosphorous), heavy metals and priority hazardous substances from WWTP effluent with various treatment techniques to reach the effluent quality which is required by the Water Framework Directive 2000/60/EC.

Legislation

In 1998 the Ministry of Transport and Water Management introduced the fourth Memorandum on Water Management (in Dutch Vierde Nota Waterhuishouding or NW4) which contains the water policy for the Netherlands until 2006 (Beesen, 1998). In the NW4 the norms for the water quality are defined and discharge points, which need to be improved, are set out. The NW4 contains norms for Maximum Permissible Risk (MPR) (in Dutch Maximaal Toelaatbaar Risiconiveau, MTR) and target values. The MPR values are related to the minimum water quality which needs to be reached by the end of 2015. The MPR values are preliminary and it is not known yet which final values will become effective. The target values are set as a norm to prevent negative ecological effects on a longer time scale (Scherrenberg, 2006).

In 2000 the European Union introduced the Water Framework Directive (WFD). The aim of the WFD is to have an ecological and biological balance for all surface waters, coastal waters, transitional waters and groundwater in Europe effectively working in 2015. The ecological quality
status of water bodies is based on the status of biological, hydromorphological and physicochemical quality elements (Borja, 2005). The European Commission identified 22 priority substances and their maximum allowable concentrations, FHI-values (Fraunhofer Institute) which are specified in the Annex of the WFD. The WFD focuses on catchment areas rather than on individual countries which means that catchment areas may cross country borders.

Since the introduction of the Dutch NW4 in 1998 and the European WFD in 2000, water authorities are obligated to fulfil more stringent discharge limits. To meet the deadline in 2015, water authorities in all river basin districts in Europe are obliged to a coherent program of measures by 2009. In case a river basin district includes more than one member state, a trans-boundary management plan must be developed. The Netherlands is involved in management plans for four trans-boundary river basin districts: the districts Rhine, Meuse, Scheldt and Ems. In the meanwhile the values identified in the NW4, the MPR values, are still leading as well as the European guidelines for Swimming Water Quality and Dangerous Substances.

**Advanced treatment of WWTP effluent**
Most current WWTPs do not have the ability to remove priority substances to the required low levels and therefore advanced treatment will be necessary. With advanced treatment of WWTP effluent low values for nutrients (phosphorous and nitrogen), heavy metals and priority hazardous substances are expected to be reached in the discharged water. These advanced techniques can also be used for production of process water for the industry or agricultural proposes itself or as pre-treatment technique for ultrafiltration and reverse osmosis to produce ultra pure water. With the removal of nutrients and different priority hazardous substances also natural organic matter (NOM) will be removed. Therefore practical research information about these techniques is interesting for both fields of interest.

**Effluent quality WWTP Leiden Zuid-West**
The yearly average effluent quality for WWTP Leiden Zuid-West of 2006 compared with the MPR values are given in table 1. The parameters are Nitrate (NO₃-N), Kjeldahl nitrogen (N_Kj), total nitrogen (N-total), total suspended solids (TSS), total-phosphorous (P-total), chemical oxygen demand (COD) and biochemical oxygen demand (BOD). Note that the average P-total values are extremely low, the mean value for WWTPs in The Netherlands is 0.5 – 1 mg P-total/L (STOWA, 2006).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Average MPR</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO₃-N</td>
<td>mg/L</td>
</tr>
<tr>
<td>N_Kj</td>
<td>mg/L</td>
</tr>
<tr>
<td>N-total</td>
<td>mg/L</td>
</tr>
<tr>
<td>TSS</td>
<td>mg/L</td>
</tr>
<tr>
<td>P-total</td>
<td>mg/L</td>
</tr>
<tr>
<td>COD</td>
<td>mg/L</td>
</tr>
<tr>
<td>BOD</td>
<td>mg/L</td>
</tr>
</tbody>
</table>

This research focuses on the removal of nutrients, heavy metals and priority hazardous substances from WWTP effluent with different treatment techniques to reach the effluent quality which is required by the Water Framework Directive 2000/60/EC. Until now the focus is mainly on phosphorous and nitrogen removal, in a later stage of the research removal of heavy metals and priority hazardous substances will also be investigated. The Rijnland District Water Control Board has set three main goals for this research; firstly to investigate if the MPR values can be reached,
secondly, finding the design parameters for full scale installations and thirdly, to gain experience with continuous sand filtration and dual media filtration.

METHODS
In 2006 The Rijnland District Water Control Board started semi-practical pilot investigations at WWTP Leiden Zuid-West in cooperation with Witteveen+Bos Consulting Engineers and Delft University of Technology. The STOWA (Dutch Foundation for Applied Water Research) is participating in this demonstration project. The project covers STOWA’s objectives to carry out research for the improvement of water management. The project is financially supported by the LIFE program of the European Union. WWTP Leiden Zuid-West treats the water of 126,000 inhabitants living in the area of Leiden Zuid-West, Voorschoten and Zoeterwoude-Dorp. The average daily flow is 24,000 m$^3$. At the WWTP first of all removal of coarse solids takes place followed by nitrification and denitrification combined with chemical phosphorous removal and finally sedimentation.

The semi-practical installation at WWTP Leiden Zuid-West consists of small full scale installations with a maximum capacity of 150 m$^3$/h. The semi-practical installation is divided into two streets, which are shown in figure 1. Street A consists of two flocculation tanks and a continuous sand filter (CFA). Street B consists of a continuous sand filter (CFB), two flocculation tanks and a dual media filter. Activated carbon can be placed after both streets for the removal of priority substances.

![Diagram of semi-practical installation at WWTP Leiden Zuid-West](image)

**Figure 1.** The semi-practical installation at WWTP Leiden Zuid-West

Both continuous sand filters are denitrifying filters, methanol (MeOH) is used as a carbon source. The grain size of the quartz sand is 1.2 – 2 mm (Menkveld, 2006b). The flow of CFA is 75 m$^3$/h which results in a filtration rate of 25 m/h. The flow of CFB is 52.5 m$^3$/h which results in a filtration
rate of 17.5 m/h. The dual media filter installation contains two filtration layers (Menkveld, 2006b), the top layer is anthracite (800 mm height, 2.0 – 4.0 mm grain diameter and a density of 1400 kg/m$^3$), the bottom layer is quartz sand (400 mm height, 1.2 – 2.2 mm grain diameter and a density of 2600 kg/m$^3$). The dual media filter is used for phosphorous removal only. The flow is 30 m$^3$/h (filtration rate 10 m/h) with filtration runs up to 24 hours. The dual media filter is back-flushed with air and filtrate water which is collected in a filtrate buffer.

Online analysers for total-phosphorous, ortho-phosphorous (PO$_4$-P or P-ortho), suspended solids, NO$_x$-N (nitrate and nitrite) and oxygen are placed in the WWTP effluent, in the filtrate of the CFA and in the filtrate of the dual media filter. The online analysers for P-total and P-ortho measure every 6 minutes alternating between the P-ortho and the P-total value. The sample is not filtered over 0.45 µm which means that the measured values for P-ortho are the sum of P-ortho and metal bound phosphorous. These online analysers are used to control the methanol and coagulant dosage.

For methanol dosing ratios of 3-5 kg MeOH/kg NO$_x$-N are used. Coagulant dosage is tested on the CFA and on the dual media filter. As coagulant ferric(III)chloride (FeCl$_3$) or polyaluminiumchloride (PACl) is used. The coagulant is dosed in-line before the flocculation tanks. Initial mixing takes place by a static mixer (G-value ±1000 s$^{-1}$). According to Tchobanoglous (2003) and Bratby (2006) a G-value of ±1000 s$^{-1}$ is optimal for initial mixing. The flocculation tanks can be bypassed, used separately or used in series. With the flocculation tanks a flocculation time of 35 minutes can be set when a filtration rate of 10 m/h is used. Process parameters such as dosing ratio, flocculation time and G-values during flocculation have been tested on lab scale with a jartester before tested full scale. For CFA a continuous dosage of 0.5 – 2 mg Al$^{3+}$/L is used, the flocculation tanks are by-passed. The FeCl$_3$ dosage for the dual media filter is based on the metal-PO$_4$-P ratio, which varies between 3 and 10 mol/mol. PACl dosage is tested with metal-PO$_4$-P ratios of 2 – 15 mol/mol. It needs to be taken into account that the metal-PO$_4$-P dosing ratios are based on the P-ortho concentration in the WWTP effluent and that the P-ortho measurement is the sum of P-ortho and metal bound phosphorous, which was already explained in the previous paragraph. As a consequence of this P-ortho measurement, and because of the preliminary continuous (biological) sand filtration, only an estimation of the metal-PO$_4$-P ratio can be made.

RESULTS

Denitrification

During the start up period the filtration rates of CFA are increased to the hydraulical maximum of 25 m/h (75 m$^3$/h). A methanol dosing ratio of 5 kg MeOH/kg NO$_x$-N and 0.8 kg MeOH/kg O$_2$ is used, without coagulant dosage. In figure 2 the results are shown for NO$_x$-N removal with a filtration rate of 25 m/h. These filtration rates are very high according to STOWA (2006), which describes different researches with continuous sand filtration in the Netherlands, Germany and Norway. It is shown that removal rates for NO$_x$-N of about 80% can be reached. Grab samples and 24 hours samples show that MPR values of 2.2 mg/L for total nitrogen can be reached at stable ingoing nitrogen loads for a longer period.

The flow of the CFB is 52.5 m$^3$/h (17.5 m/h), the MeOH dosing ratio is equal to street A, with this flow rate removal rates for NO$_x$-N are comparable with CFA (75 m$^3$/h) when no coagulant dosage is used. MPR values of 2.2 mg N-total/l can be reached. For filtration rates of 17.5 m/h the mean conversion rate is 1.9 kg NO$_x$-N/m$^3$.day, for filtration rates of 22.5 m/h the mean conversion rate is 1.8 kg NO$_x$-N/m$^3$.day.
Simultaneous nitrogen and phosphorous removal
For simultaneous removal of nitrogen and phosphorous, MeOH and PACl are dosed at a filtration rate of 25 m/h without using flocculation tanks. The methanol dosing ratio is decreased from 5 to 3.5 kg MeOH/kg NO\textsubscript{X}-N. During simultaneous nutrient removal no negative effects on the denitrification are observed.

Figure 2. Results of NO\textsubscript{X}-N removal of street A at a filtration rate of 25 m/h

Figure 3. Phosphorous removal at a filtration rate of 25 m/h and a continuous coagulant dosage of 1 and 0.5 mg Al\textsuperscript{3+}/l
In figure 3 results are shown for phosphorous removal with a continuous dosage of 1 mg Al$^{3+}$/L and 0.5 mg Al$^{3+}$/L. The mean metal-PO$_4$-P dosing ratios are respectively 6.5 mol/mol and 5.7 mol/mol. The P-total and P-ortho concentrations in the filtrate water remain stable after the coagulant dosage is decreased. The removal rate is about 65% for P-total and 78% for P-ortho. The online data show that the MPR value of 0.15 mg P-total/L can be reached.

The minimal metal-PO$_4$-P dosing ratio needed to reach the MPR value for P-total is 4 mol/mol. If the P-ortho concentration in the WWTP effluent increases, the metal-PO$_4$-P ratio will decrease and the MPR value can not be reached. The conclusion is that continuous sand filtration with denitrification and a continuous low coagulant dosage can reach MPR values for the filtrate water at low P-total concentrations (0.2 - 0.3 mg/L) in the WWTP effluent.

**Phosphorous removal**

Jar tests which were executed before the start up of the dual media filter gave the best removal efficiencies for ferric(III)chloride compared to aluminium(III)chloride or polyaluminiumchloride. Based on the results, full scale tests with dual media filtration started with FeCl$_3$. The tests which are conducted are given in table 2.

**Table 2.** Process parameters phosphorous removal tests with dual media filter

<table>
<thead>
<tr>
<th>Coagulant</th>
<th>Flow rate (m/h)</th>
<th>Metal-PO$_4$-P (mol/mol)</th>
<th>Flocculation time (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeCl</td>
<td>10</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>FeCl</td>
<td>6</td>
<td>5</td>
<td>60</td>
</tr>
<tr>
<td>PACl</td>
<td>10</td>
<td>5</td>
<td>35</td>
</tr>
<tr>
<td>PACl</td>
<td>10</td>
<td>10</td>
<td>35</td>
</tr>
<tr>
<td>PACl</td>
<td>10</td>
<td>15</td>
<td>35</td>
</tr>
</tbody>
</table>

In figure 4 results are shown for phosphorous removal with a continuous dosage of 1 mg Al$^{3+}$/L and 0.5 mg Al$^{3+}$/L. The mean metal-PO$_4$-P dosing ratios are respectively 6.5 mol/mol and 5.7 mol/mol. The P-total and P-ortho concentrations in the filtrate water remain stable after the coagulant dosage is decreased. The removal rate is about 65% for P-total and 78% for P-ortho. The online data show that the MPR value of 0.15 mg P-total/L can be reached.

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**Figure 4.** Phosphorous removal for street B, Fe$^{3+}$ dosing ratio of metal-PO$_4$-P 5 mol/mol, a filtration rate of 6 m/h and a flocculation time of 35 minutes
The first experiment is executed without using flocculation tanks (flocculation time about 10 minutes), with a metal-PO\textsubscript{4}-P ratio of 5 mol/mol and a filtration rate of 10 m/h. Removal rates are about 50% for P-total resulting in average filtrate concentrations of 0.28 mg P-total/L. Secondly a test is executed with a flocculation time of 60 minutes after initial coagulation, with an equal dosing ratio and a filtration rate of 6 m/h. The G-values in the flocculation tanks are ± 60 s\textsuperscript{-1}. In figure 4 the results are shown of street B for P-total and P-ortho removal by using two flocculation tanks. Removal rates decreased for total phosphorous to 44% resulting in an average filtrate concentration of 0.32 mg P-total/L.

Due to unsatisfying results and breakthrough of FeCl\textsubscript{3}, the ferric dosage is replaced for PACl which was the second best coagulant regarding the jartests. The filtration rate is 10 m/h, de dosing ratio metal-PO\textsubscript{4}-P is 5 mol/mol (see figure 5). Both flocculation tanks are in use (flocculation time 35 minutes) with a G-value of ±60 s\textsuperscript{-1}. The removal efficiencies for P-total varies between 30-50%, which is no improvement compared to FeCl\textsubscript{3}. Breakthrough of Al\textsuperscript{3+} is not as high as for FeCl\textsubscript{3} and the turbidity of the filtrate water is 1.5 NTU (nephelometric turbidity units). The feed water of the dual media filter is 2.5 NTU.

Increasing the metal-PO\textsubscript{4}-P ratio from 5 to 10 mol/mol improves the removal efficiency for total-phosphorous up to 40%. Concentrations for total phosphorous in the filtrate water are near to or sometimes complying with the MPR value of 0.15 mg P-total/L. The metal-PO\textsubscript{4}-P ratio was further increased to 15 mol/mol. At the same moment the total and ortho-phosphorous concentrations in the WWTP effluent became extremely low (P-total 0.20 mg /L). It is possible that due to the low concentrations the removal efficiencies decreased, for ortho-phosphorous this was 10% and for total phosphorous 20%.

P-ortho measurements (filtrated over 0.45 µm) show for all the executed filtration tests that the P-ortho is completely bound after the initial coagulation. This means that the ortho-phosphorous measured in the filtrate water is mainly bound phosphorous and that the relatively low removal efficiencies are caused by either floc breakage in the filter medium or an insufficient floc growth.

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**Figure 5.** Phosphorous removal for street B, Al\textsuperscript{3+} dosing ratio of metal-PO\textsubscript{4}-P 5 mol/mol and a filtration rate of 10 m/h
CONCLUSIONS
Removal rates for NO\textsubscript{x}-N are comparable for CFA and CFB. This means that flow rates up to 75 m\textsuperscript{3}/h (25 m/h) have no negative effect on the conversion of NO\textsubscript{x}-N at stable nitrogen loads. Combined with a low (0.5 mg Al\textsuperscript{3+}/L) continuous coagulant dosage, high removal rates for total phosphorus can be reached in spite of the low total phosphorous concentrations in the WWTP effluent.

Removal rates for total phosphorous with dual media filtration are not enough satisfying. The use of flocculation tanks do not enough increase the removal efficiencies of the dual media filter for total phosphorous. Changing coagulant from FeCl\textsubscript{3} to PACI with the used configuration has no significant effect on the removal efficiencies.

The results of this research up to now show that continuous sand filtration has the ability of removing total nitrogen and total phosphorous to MPR values at high filtration rates of 25 m/h. This means that continuous sand filtration is suitable as pre-treatment for ultrafiltration and reverse osmosis to produce ultra pure water. The relatively low removal rates for total phosphorous with the dual media filtration will be investigated further. Two options will be tested, namely a different coagulant and replacing the filter media for smaller grain sizes. In a later stage of the research the removal of priority hazardous substances and heavy metals will be investigated.

The advanced treatment of WWTP effluent is not only good to reach a better surface water quality but makes also the possibility of reuse easier.

REFERENCES
Tchobanoglous, G. (2003) Wastewater Engineering : Treatment and Reuse, Metcalf & Eddy, Inc. , 4\textsuperscript{th} Edition