An integrated wastewater reuse concept combining natural reclamation techniques, membrane filtration and metal oxide adsorption

A. Sperlich, X. Zheng, M. Ernst and M. Jekel

ABSTRACT

In a Sino-German research project, a sustainable water reclamation concept was developed for different applications of municipal water reuse at the Olympic Green 2008 in Beijing, China. Results from pilot-scale experiments in Beijing and Berlin show that selective nutrient removal by adsorption onto granular ferric hydroxide (GFH) after a membrane bioreactor (MBR) can maintain a total phosphorus concentration of <0.03 μg L⁻¹ P, thus preventing eutrophication of artificial lakes. Operation time of GFH adsorption columns can be extended by regeneration using sodium hydroxide solution. A subsequent ultrafiltration (UF) membrane after bank filtration creates an additional barrier for pathogens and allows for further urban reuse applications such as toilet flushing. Short term bank / bio-filtration prior to UF is shown to effectively remove biopolymers and reduce membrane fouling.

Key words | fouling control, membrane filtration, metal oxide adsorption

INTRODUCTION

The 2008 Summer Olympics will be held in Beijing, China. Green Olympics is one concept of the Beijing Olympic Games and states the determination and ambition for environmental protection. However, the environmental problems of the mega-city Beijing are still tremendous. Above all the water sector is struggling with quality problems and declining groundwater levels. As a consequence the City of Beijing is striving for long term solutions for wastewater treatment and its reuse with a challenging and Olympic-oriented water program. The Olympic Green is intended to serve as a demonstration area for innovative, sustainable wastewater treatment and reuse technologies. The Park covers a total area of 1,215 ha, located north of the city centre. An area of 60 ha of constructed artificial lakes and channels will be filled with treated wastewater in the northern area of the Park. In the central area, a smaller lake of 15 ha with superior water quality will be operated with fountains; all waters supplied will be advanced treated municipal wastewaters. Further reuse applications are envisaged, including water for irrigation (not only in the Park, but also in neighboring areas) and urban water applications like toilet flushing and washing waters.

The present study summarizes the results of the Sino-German research cooperation project on a sustainable water concept and its application for the Olympic Games 2008 in the subproject “wastewater treatment and reuse”. Project partners are the Beijing Drainage Group, the Tsinghua University in Beijing and the Technical University of Berlin. Moreover, a German small and medium-sized enterprise (SME) and a consulting company are involved. A combination of advanced wastewater treatment and nature-orientated technologies were realized in pilot-scale at the Beijing wastewater treatment plant (WWTP) Beixiaohe. The main objectives included advanced nutrient removal and eutrophication control of scenic impoundments, reuse for irrigation and urban reuse.

Based on the plans for the Olympic Green and the extension of the WWTP Beixiaohe, a reuse concept has
been developed (Ernst et al. 2007), including multiple barriers for contaminants and pathogens. Effluent of a membrane bio-reactor (MBR) with in-situ P-precipitation will be treated to two different qualities, depending on the use. In a first treatment step, phosphate as essential nutrient is removed by adsorption onto granular ferric hydroxide (GFH) and the water is pumped into an artificial lake. Here, the water is used for scenic impoundment, but also further treated by biological processes. Finally, lake bank filtration (BF) further removes organic compounds and serves as an additional pathogen barrier and effective pre-treatment step for a subsequently operated membrane filtration. An ultrafiltration (UF) ensures a sufficient water quality for safe non-potable (urban) reuse.

Research on the developed treatment scheme focussed around two major questions. First, selective phosphorus removal by adsorption onto GFH and eutrophication control in the artificial lake was studied intensively. In previous laboratory-scale experiments, GFH has shown high capacities for phosphate removal from MBR filtrates (Genz et al. 2004) and wastewater effluents (Ernst et al. 2007), despite potential competition by other wastewater constituents. A particle free influent and the goal of very low phosphorus concentrations in the effluent favors adsorption compared to other options of phosphorus removal, e.g. coagulation. Second, fouling of the ultrafiltration membrane and its control by biofiltration was investigated. Fouling is generally acknowledged as one of the main problems restraining the application of membrane filtration (AWWA 1998a). Polysaccharides and proteins have been found to be major foulants (Jarusutthirak & Amy 2006; Laabs et al. 2006). Removal of these substances by biofiltration might therefore significantly reduce membrane fouling.

**METHODS**

The developed treatment scheme (Figure 1) was realized in pilot-scale at the Beijing WWTP Beixiaohe. Effluent of a pilot membrane bioreactor, operated by the Tsinghua University, was used as influent for two GFH fixed-bed columns. Since MBR effluent P concentrations varied substantially, a flocculation step using poly-aluminium chloride (PAC, dosage of 8–16 mg L\(^{-1}\) Al) followed by sedimentation and rapid sand filtration was introduced before GFH adsorption. In large-scale application, this step could be substituted by in-situ precipitation or biological P removal in the MBR. The resulting modified treatment scheme for pilot operation in Beijing consisted of MBR effluent – flocculation/sedimentation – rapid sand filtration – GFH adsorption – artificial lake – bank filtration – ultrafiltration.

To achieve effluent concentrations of less than 30 μg L\(^{-1}\) P, two fixed-bed adsorption columns were operated. Operational conditions and parameters are given in Table 1. To simulate the Olympic Lakes, a 30 m\(^2\) large artificial lake was constructed and operated. At start-up, the lake was filled with 95 % drinking water, supplemented by 5% secondary

![Figure 1](https://iwaponline.com/wst/article-pdf/57/6/909/438777/909.pdf)

**Table 1 | Operational parameters of two GFH fixed-bed columns at WWTP Beixiaohoe**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Operation in series</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow rate [m(^3) d(^{-1})]</td>
<td>9</td>
</tr>
<tr>
<td>Empty bed contact time [min]</td>
<td>2 × 6</td>
</tr>
<tr>
<td>Empty bed volume [m(^3)]</td>
<td>2 × 0.038</td>
</tr>
<tr>
<td>Column diameter [m]</td>
<td>2 × 0.25</td>
</tr>
<tr>
<td>Bed depth [m]</td>
<td>2 × 0.76</td>
</tr>
<tr>
<td>Filtration rate [m h(^{-1})]</td>
<td>7.6</td>
</tr>
<tr>
<td>Backwash</td>
<td></td>
</tr>
<tr>
<td>Filtration rate [m h(^{-1})]</td>
<td>25</td>
</tr>
<tr>
<td>Flow rate [m(^3) h(^{-1})]</td>
<td>1.2</td>
</tr>
<tr>
<td>Volume [BV/m(^3)]</td>
<td>2/0.075</td>
</tr>
</tbody>
</table>
effluent. In order to facilitate the settlement of planktonic organisms, plankton samples were taken from four lakes in Beijing and the lake was inoculated. The dimensions of the lake were 5.5 m × 5.5 m × 2.5 m, the applied filtration materials were sand \((d = 2–4 \text{ mm})\) and limestone \((d = 3–6 \text{ mm})\) which were set up in two separated containers to form embankments. On the surface of each bank, a layer of textile was situated to ensure stability. The BF was operated with a filtration passage 1.7 m in depth, a filtration velocity of 1.55 m d\(^{-1}\) and a travelling time of about 1.1 to 1.5 days.

An UF pilot plant (W.E.T., Germany) was operated with a membrane module (INGE, Germany, membrane material: PESM, MWCO: 100 kD, dead-end filtration, membrane surface 4.5 m\(^2\), operational flux: 60 – 140 L/m\(^2\)h).

Following the pilot experiments in Beijing, a similar set-up was realized in Berlin’s WWTP Ruhleben, Germany. Here, fine-screened secondary effluent was applied directly to four biofilters (slow sand filtration), operated at different hydraulic loading rates between 0.02 and 0.5 m h\(^{-1}\). The filtration area was 0.018, 1 and 2 m\(^2\), respectively. Filter height was approximately 0.72 m. As filter material, sand was chosen. The subsequently operated UF pilot plant was the one operated in Beijing before.

Water quality analysis included pH, turbidity, ammonia, total phosphorus, total coliforms, UV absorption at 254 and 436 nm, respectively. All analyses were performed according to Standard Methods (AWWA 1998b). Dissolved organic carbon in the water was characterized by size exclusion chromatography (LC-OCD with DOC/UVA online detection).

**RESULTS AND DISCUSSION**

Operation of the pilot adsorption columns was not constant during the operation period (July 28th to October 31st 2005). Start-up of the MBR pilot unit upstream resulted in strong variations of the influent concentration as well as shut-down periods. The measured effluent concentrations show a reliable and almost complete removal of phosphate by adsorption onto GFH. Despite highly variable influent concentrations (between 0.008 and 1.62 ppm P), the average effluent concentration was 0.023 ppm P, but always below 0.07 ppm P. After approximately 12,000 BV, breakthrough of the adsorber occurs (Figure 2). Following this, the first of two in-series operated adsorbers was regenerated using 1 molar sodium hydroxide solution, thereby extending the operation time to more than 16,000 BV or 96 days, respectively. For regeneration, 400 L, equivalent to 10 bed volumes, of 1 molar sodium hydroxide solution were used. After a throughput of 100 L regenerant at 20 L h\(^{-1}\), regeneration was paused for 12 h. The remaining 300 L regenerant was applied at 45 L h\(^{-1}\).

Figure 3 shows that after four bed volumes the major part of the bound phosphate is already desorbed. A total of 10.5 g P per kg dry matter GFH material could be desorbed. Corresponding lab-scale batch experiments show an average regeneration efficiency of 86%, and marginal influence of operational time and pH.

By the effective removal of phosphate, which is the limiting nutrient for microbial growth, eutrophication of the artificial

![Breakthrough curve of GFH pilot columns at WWTP Beixiaohe.](https://iwaponline.com/wst/article-pdf/57/6/909/438777/909.pdf)

![Phosphate desorption using 1M NaOH as regenerant for fixed-bed GFH columns in WWTP Beixiaohe.](https://iwaponline.com/wst/article-pdf/57/6/909/438777/909.pdf)
The mean concentrations in the operation period for chlorophyll A and total phosphorus were 2.4 mg m\(^{-3}\) and 25.1 mg m\(^{-3}\), respectively. Following the Vollenweider/OECD model (Vollenweider & Kerekes 1982), the artificial lake can be classified as mesotrophic.

Table 2 shows the average water quality parameters of the lake water, sand filtrate and UF permeate. The reuse water quality after UF generally meets the Chinese reuse water standard (Ernst et al. 2007), in which chlorination is mandatory. This demonstrates the advantage of UF removing particles and bacteria in wastewater reuse.

During an operational period of more than 3 months (July–Nov. 2005) trans-membrane pressure (TMP) of UF varied in different patterns depending on the characteristics of influents through different filtration media (Figure 4). In the first phase (July 27th to Aug. 15th) of the experiment, the influent to UF was a 50:50 mixture of sand and limestone filtered water. Because of impurities in the used limestone, effluent pH after limestone filtration was always higher than 10 and serious scaling was observed. In this phase scaling and organic fouling caused a steep TMP increase. In phase 2 (Aug.16th to Sep.12th), the influent to UF was exclusively effluent from sand filtration. TMP increased quickly, probably due to organic fouling. In this phase, organic foulants were not effectively removed, which can be explained by an insufficient adaptation of the biofilter. TMP increased much slower in phase 3 (Sep.12th to Nov. 7th) compared to preceding phases. During this period, influent to UF was effluent from sand filtration only. Less organic fouling lead to a longer operation time and a moderate TMP increase of around 400 mbar within 55 days.

Figure 5 shows a LC-OCD diagram of the artificial lake water, bank filtration (limestone/sand) effluent and ultrafiltration permeate. The liquid chromatography unit separates organic compounds according to their molecular size through the size exclusion column. Online detection of organic carbon leads to three characteristic peaks, representing biopolymers, humic substances, and low molecular weight acids. It is shown that the artificial lake water contains high concentrations of biopolymers (first peak at a retention time of 40 min), but sand filtration effectively removes the biopolymer peak. Limestone
filtration shows only slight reduction in biopolymer concentration, which explains the TMP development of the UF pilot plant (Figure 4). After an adaptation time (phase 2), biological processes in the filter material remove biopolymers.

Further investigations on the removal of biopolymers by biofiltration in Berlin’s WWTP Ruhleben indicate a direct correlation between biopolymer concentration, filtration rate and fouling (Zheng et al. 2007). Figure 6 shows the removal of biopolymers by sand filtration at different filtration rates. The complete absence of a biopolymer peak in the UF permeate indicates that biopolymers are major foulants. Sand filtration reduces the biopolymer concentration, with lower filtration rates showing better removal and a higher filterability of the water sample.

Figure 7 shows the biopolymer removal by biofiltration for an operation time of 7 months (Nov. 2006 to May 2007), confirming that the filtration rate influences biopolymer removal. Biopolymer removal rate is improved from 32 % to 50 % on average when filtration rate is decreased from 0.5 m h\(^{-1}\) to 0.25 m h\(^{-1}\), but as filtration rate sinks from 0.1 m h\(^{-1}\) to 0.05 m h\(^{-1}\), the corresponding biopolymer removal rate increases only from 62 % to 65 % on average. This implies a need for technical optimization of the biofiltration rate and the performance of the subsequent UF.

CONCLUSIONS

- The Beijing pilot experiments have shown that the proposed reuse concept is a reasonable and feasible treatment option.
- The produced water quality is sufficient for scenic impoundment reuse (after phosphate removal) and urban reuse purposes (after membrane filtration).
- GFH adsorption shows high capacities for phosphate removal and its application produces effluents of very low phosphate concentrations for use in artificial lakes, thus preventing eutrophication.
- GFH material can be regenerated using sodium hydroxide solution, resulting in longer operation times of fixed-bed systems before the material needs to be replaced.
- Short term bank/bio-filtration prior to UF reduces the fouling potential by removing biopolymers, thus reducing the operational costs of UF membranes.
ACKNOWLEDGEMENTS

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