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Comparison of two PAC/UF processes for the removal of micropollutants from wastewater treatment plant effluent: Process performance and removal efficiency

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ABSTRACT

Two hybrid membrane processes combining powdered activated carbon (PAC) adsorption with ultrafiltration (UF) were investigated regarding operational performance and efficiency to remove organic micropollutants from municipal wastewater treatment plant effluent. A pressurized PAC/UF (pPAC/UF) and a submerged PAC/UF (sPAK/UF) system were operated continuously over a period of six months.

Both UF membrane systems showed good compatibility with the application of PAC showing no abrasion or other negative impacts. The pPAC/UF system reached permeability values up to 290 L/(m^2 h bar) at high fluxes of 80 L/(m^2 h) compared to the sPAC/UF with a permeability of up to 200 L/(m^2 h bar) at fluxes of up to 23 L/(m^2 h). Surface analysis of both membranes with scanning electron microscopy revealed no membrane deterioration after the six-month period of operation. On the surface of the pressurized membrane the formation of a PAC layer was observed, which may have contributed to the high permeability by forming a protective coating.

Five micropollutants, i.e. sulfamethoxazole, carbamazepine, mecoprop, diclofenac and benzotriazole in ambient effluent concentrations were investigated. Both PAC/UF systems removed 60–95% of the selected micropollutants at a dosage of 20 mg PAC/L and 4 mg Fe^{3+}/L. However, extreme peak loads of sulfamethoxazole with concentrations of up to 30 μg/L caused a considerable performance decrease for more than a week.

Abbreviations: AIB, Amt für industrielle Betriebe Basel-Landschaft; BZT, Benzotriazole; CBZ, Carbamazepine; CEB, Chemical enhanced backwash; DCF, Diclofenac; DOC, Dissolved organic carbon; FOEN, Federal Office for the Environment; GAC, Granular activated carbon; LC-OCD, Liquid chromatography organic carbon detection; LMW, Lower molecular weight; MDL, Method detection limit; MEC, Mecoprop; MRM, Multiple reaction monitoring; PAC, Powdered activated carbon; pPAC/UF, Pressurized PAC/UF system; SBR, Sequencing-batch-reactor; SMX, Sulfamethoxazole; sPAC/UF, Submerged PAC/UF system; SEM, Scanning electron microscopy; TMP, Transmembrane pressure; TSS, Total suspended solids; UF, Ultrafiltration; WWTP, Wastewater treatment plant.

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1. Introduction

Micropollutants have been detected in the range of ng/L to μg/L in the urban water cycle and natural water bodies, presenting a rising concern in protection of the aquatic environment and conservation of water resources used for drinking water supply (Gälli et al., 2009; Götz et al., 2010). Although mostly occurring in concentrations far below their effectiveness in humans, these substances are potential hazards particularly for the receiving environmental compartments (Daughton and Ternes, 1999; Ternes, 1998; Roig, 2010). Organic micropollutants found in the aquatic environment typically include pharmaceuticals, personal care products, industrial chemicals from consumer products and pesticides. Several comprehensive studies identified the effluent of wastewater treatment plants (WWTP) as the main origin of micropollutants entering natural water bodies (Gälli et al., 2009).

In Switzerland ambitious efforts are underway to reduce the discharge of micropollutants into natural water bodies (FOEN, 2012a). Ort et al. (2009) evaluated the impact of effluent from 742 WWTPs on micropollutant loading throughout all Swiss river catchments. Their study suggests upgrading of 173 WWTPs if diclofenac cannot be reduced at source. Subsequently, within the next 20–25 years around 100 of the currently operating WWTPs are supposed to be upgraded with an additional treatment step to reduce the discharge of micropollutants (Gälli et al., 2009). Consequently, the identification of suitable and economically feasible process combinations has become a major focus of environmental research for instance in Switzerland and Germany over the last years.

Several treatment technologies can remove or break down micropollutants from WWTP effluent very effectively. In particular, powdered activated carbon (PAC) adsorption has been identified as powerful and easily adjustable technology to remove micropollutants (Snyder et al., 2007; Federal Office of the Environment FOEN, 2012b; Zwickenpflug et al., 2010). Margot et al. (2013) confirmed the high potential of PAC to reduce the environmental impact of effluent discharge with regard to ecotoxicity. However, in comparison to granular activated carbon (GAC), PAC is typically incinerated or dumped while GAC can be regenerated (Aktas and Cecen, 2007).

PAC adsorption has been investigated for the removal of micropollutants from effluent in lab-, pilot- and even in technical scale (Zwickenpflug et al., 2010; Metzger, 2010; FOEN, 2012b; Margot et al., 2013). The highest efficiency can be achieved when PAC is dosed to the effluent after biological treatment. While the dosage and mixing of PAC can be implemented quite easily, often using existing infrastructure, separating PAC efficiently prior to the final discharge of the treated effluent remains a technical challenge.

While sedimentation and sand filtration are the most common methods to separate PAC from the effluent stream, ultrafiltration (UF) offers several advantages such as complete PAC and bacteria retention alongside high virus removal and less space demand (Menzel, 1997; Metzger, 2010; Remy et al., 2009). Membrane filtration processes in combination with upstream PAC adsorption are already being used in drinking water treatment, in which PAC has shown positive effects on the operation performance of the membrane stage. The main reason for that has been reported to be the adsorption of a certain fraction of the dissolved organic carbon (DOC) upstream of the membrane to mitigate fouling. DOC removal of up to 70% has been reported for the PAC/UF process (Jeong et al., 2012).

Though the general applicability has been proven, open questions remain particularly regarding the impact of PAC on membrane operation such as the influence of PAC on membrane fouling or the transmembrane pressure (TMP) development.

Studies on the topic of PAC/UF processes show that the efficiency of the process is depending on many factors such as membrane material properties, operating condition and characteristics of the micropollutants as well as the water matrix (Campos et al., 2000; Matsui et al., 2001; Vigneswaran et al., 2003; Summers et al., 2011).

While some studies showed a positive effect of PAC dosage on membrane performance (Remy et al., 2009; Li et al., 2011; Jeong et al., 2012; Campinas and Rosa, 2010), others mention negative effects causing for example a lower filtration flux (Stoquart et al., 2012) or less effective backwash if higher amounts of PAC are used (Tomaszewska and Mozia, 2002). An evaluation of the available literature on the topic indicates that a beneficial effect of PAC on membrane processes is achievable if PAC type and membrane system are adjusted to the respective water matrix.

Building on the promising findings of recent studies, the work reported here employs two ultrafiltration methods, namely pressure driven membranes operated in inside-out mode and submerged UF membranes operated in outside-in mode. The technical feasibility and micropollutant removal efficiency were investigated for both processes operated in parallel. Furthermore, preferential operating conditions were identified by adjusting filtration, backwash and cleaning intervals. The tests were conducted at the WWTP Birs in Birsfelden, Canton Basel-Landschaft, Switzerland.

The addition of PAC was performed at a target concentration of 20 mg/L which was found to be sufficient to achieve the envisaged degree of micropollutant removal, based on previous studies on PAC adsorption on WWTP effluent (Metzger, 2010; Margot et al., 2013). The two different PAC/UF processes were studied for an extended period of six months. The removal of five micropollutants, namely sulfamethoxazole (SMX), carbamazepine (CBZ), mecoprop (MEC), diclofenac (DCF) and benzotriazole (BZT) was investigated. Furthermore, the influence of PAC on the membrane material was examined with regard to potential damage of the membrane material for instance due to abrasion by PAC.

2. Material and methods

2.1. *Feed water characteristics*

The raw water used to feed the PAC/UF units was taken from the effluent of the municipal WWTP Birs (Birsfelden, Basel-Landschaft, Switzerland). The WWTP with a design capacity of 150,000 population equivalent is operated by the public utility Amt für Industrielle Betriebe (AIB) of the Canton. The