



Effect of long-term organic removal on ion exchange properties and performance during sewage tertiary treatment by conventional anion exchange resins



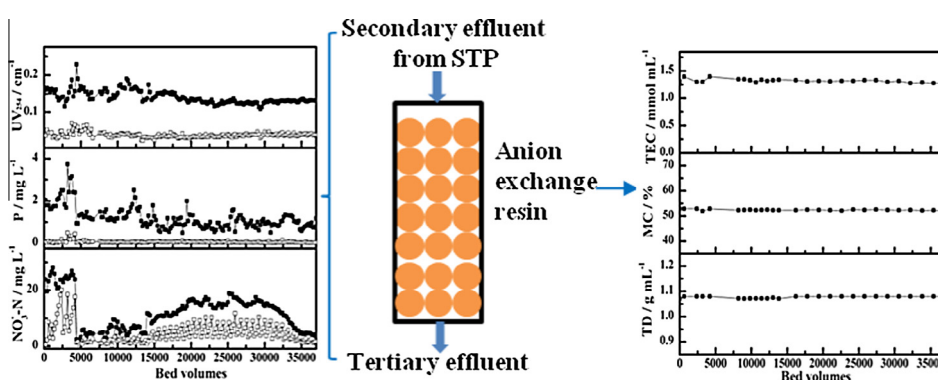
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HIGHLIGHTS

- Long-term pilot-scale evaluation on sewage tertiary treatment by conventional resin.
- Anion exchange resin stably removed DOM from secondary effluent.
- DOM fouling insignificantly influenced anion removal and resin properties.
- Ecological hazard still occurred due to residual PAHs in the tertiary effluent.
- This process produces high quality reclaimed water at low operational costs.

GRAPHICAL ABSTRACT



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ABSTRACT

This study evaluated the long-term dissolved organic matter (DOM), phosphorus and nitrogen removal performance of a commercially available conventional anion exchange resin (AER) from actual secondary effluent (SE) in a sewage treatment plant based on a pilot-scale operation (2.2 m³ d⁻¹, 185 cycles, 37,000 bed volume, 1.5 years). Particular emphasis was given to the potential effect of DOM fouling on the ion exchange properties and performance during the long-term operation. Despite the large range of COD (15.6–33.5 mg L⁻¹), BOD₅ (3.0–5.6 mg L⁻¹), DOC (6.5–24.2 mg L⁻¹), and UV₂₅₄ (UV absorption at 254 nm) (0.108–0.229 cm⁻¹) levels in the SE, the removal efficiencies of the AER for the aforementioned parameters were 43 ± 12%, 46 ± 15%, 45 ± 9%, and 72 ± 4%, respectively. Based on three-dimensional fluorescence excitation–emission matrix data, i.e., the fluorescence intensities of four regions (peaks A–D), all organic components of the SE were effectively removed (peak A 74%, peak B 48%, peak C 55%, and peak D 45%) following the adsorption. The AER effluent still has considerable polycyclic aromatic hydrocarbons' ecological hazard on freshwater fishes when they were significantly removed from SE. The obvious DOM fouling on the AER, identified by color change, had no significant influence on the long-term removal of the representative inorganic anions (averaging 95 ± 4% phosphate, 100 ± 0% SO₄²⁻, and 62 ± 17% NO₃⁻) and AER properties (including total exchange capacity, moisture content, and true density). The conventional AER can produce high quality reclaimed water from SE at a low operational cost.

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1. Introduction

Anion exchange resins (AERs) have traditionally been used to remove various inorganic anions (e.g. NO_3^- (Chabani et al., 2006), and perchlorate (Gu et al., 2007)) from raw drinking water by the ion exchange process. In many cases, coagulation–flocculation, adsorption with active carbon, and even oxidation processes have been proposed to remove organic matter in advance, to protect the resins against organic fouling (Beril Gönder et al., 2006) which can result in considerable loss of ion exchange capacity (Shuang et al., 2013). Since the 1970s, numerous studies have demonstrated the potential of AERs for natural organic matter (NOM) removal from drinking water sources (Bolto et al., 2002; Cornelissen et al., 2008). For example, ~90% of dissolved organic carbon (DOC) removal (Bolto et al., 2002; Humbert et al., 2005) can significantly reduce the formation of disinfection by-products during the subsequent chlorination process (Singer and Bilyk, 2002; Fearing et al., 2004). In the 1980s, MIEX[®], a magnetic strong-base AER with iron oxide integrated into a macroporous polyacrylic matrix, was marketed specifically for NOM removal from raw drinking water by Orica Watercare of Victoria, Australia (Allpike et al., 2005). NOM consists largely of hydrophobic acids (i.e. humic substances consisting of humic acids and fulvic acids) (Matilainen and Sillanpää, 2010), which account for ~70% of the DOC in raw drinking water (Liu et al., 2008). The most important mechanism of NOM removal using an AER is the ion-exchange process (e.g. the exchange in NOM hydrophobic acids and chloride ions) (Bolto et al., 2002; Cornelissen et al., 2008). Therefore, no NOM accumulated on AER and further interfered the ion-exchange process, which explains the absence of organic fouling of AERs used for removing organic matter from raw drinking water.

The secondary effluent (SE) from sewage treatment plants is characterized by low levels of dissolved organic matter (DOM), nitrogen (mostly as NO_3^- -N), and phosphorus (mostly as orthophosphate) (Zheng et al., 2011). Approximately 75% of DOM in SE has a low molecular weight (MW) of <1000 Da (Gong et al., 2008), whereas about 54% of NOM in drinking water sources has a MW of >1000 Da (Kim and Yu, 2005). Furthermore, unlike NOM in raw drinking water (mainly humic substances), the DOM in SE is a complex organic mixture that includes NOM, soluble microbial products, and various harmful refractory chemicals that pose a potential ecological hazard (Shon et al., 2006). It is speculated that both physical adsorption and ion-exchange occur simultaneously during the AER adsorption of DOM in wastewater (Bassandeh et al., 2013). In previous reports, the DOM fractions retained by AER included 76% hydrophilic and 55% hydrophobic compounds from SE (Ahmad et al., 2012), or 57% hydrophobic acid, 44% transphilic acid and 18% hydrophilics from paper mill effluent (Ciputra et al., 2010). In recent years, there has been increasing interest in investigating the potential of AERs for DOM removal from SE by methods including: a fluidised bed packed with AER (Purolite A500PS) (Ahmad et al., 2012), MIEX[®] in batch test (Zhang et al., 2006), or a MIEX[®] fluidised bed contactor (Zhang et al., 2008; Tien Vinh et al., 2011). Besides, several investigations of DOM removal from landfill leachate (Bashir et al., 2010; Boyer et al., 2011), paper mill effluent (Ciputra et al., 2010; Bassandeh et al., 2013) and olive mill wastewater (Victor-Ortega et al., 2015) by AER also have been conducted recently. It is noted that the aforementioned studies were conducted during short-term experimental periods with 100 (Zhang et al., 2008), 172 (Tien Vinh et al., 2011), or 800 (Ahmad et al., 2012) bed volumes (BVs) and 4 (Tien Vinh et al., 2011), 5 (Zhang et al., 2008), or 11 (Boyer et al., 2011) adsorption–regeneration cycles.

Besides DOM, phosphorus is the major limiting nutrient for algal production (Sengupta and Pandit, 2011), and phosphorus

removal technology has become a significant part of sewage tertiary treatment processes in recent years (Martin et al., 2013). Furthermore, the presence of NO_3^- is a potential public health hazard that causes infant methaemoglobinaemia and produces nitrosamine (Kioussis et al., 2000). Therefore, removal of DOM, NO_3^- -N, and phosphorus from SE should simultaneously be involved during the sewage tertiary treatment (Zheng et al., 2011). Both NO_3^- -N and phosphorus can be removed by the ion exchange process. However, conventional AERs lack greater selectivity for phosphorus or NO_3^- -N over competing substances including inorganic anions (e.g. SO_4^{2-}) and organic acids (e.g. humic acid) (Paul Chen et al., 2002). Furthermore, organic fouling of conventional AERs results in a considerable loss of ion-exchange capacity (Shuang et al., 2013). Therefore, in recent decades, some phosphorus-selective resins, e.g., polymeric ligand exchange (Zhao and Sengupta, 1998) and hybrid anion-exchange resins (Blaney et al., 2007), have been developed to replace conventional AERs for the selective removal of phosphorus from SE. Our previous investigation found that the conventional AER could achieve stable adsorption efficiencies of $\geq 71\%$ of UV-absorbing organic substances, $\geq 89\%$ phosphorus, and $\geq 95\%$ NO_3^- -N in SE for five adsorption–regeneration cycles by fixed-bed column tests (Zheng et al., 2011). This was the first investigation of the simultaneous DOM, phosphorus, and NO_3^- -N removal performance of conventional AERs for SE (Zheng et al., 2011). Compared to the phosphorus-selective resins and MIEX[®], conventional AERs may be the commercially available adsorbents for simultaneous DOM, NO_3^- -N and phosphorus removal from SE. However, like other aforementioned studies on the potential of AER for DOM removal from wastewater (Zhang et al., 2008; Boyer et al., 2011; Tien Vinh et al., 2011; Ahmad et al., 2012), our previous investigation on conventional AER for sewage tertiary treatment was also conducted during short-term experimental periods with 5 adsorption–regeneration cycles (Zheng et al., 2011). It seems that the amount of UV-absorbing organic substances accumulated on the conventional AER increased gradually from 0.4 to 1.2 g mL⁻¹ during the five adsorption–regeneration cycles (Zheng et al., 2011). Therefore, although AERs can remove NOM from drinking water sources by the ion exchange process, some uncertainties remain regarding the long-term application of AERs to remove DOM from SE. For example, due to possible DOM accumulation on AER (Zheng et al., 2011), whether the potential organic fouling on AER will result in a considerable loss of the AER ion exchange properties and performance during long-term operation is unknown, which is the most important factor determining the lifespan of AER in terms of stable phosphorus, NO_3^- -N, and DOM removal from SE. Additionally, the potential ecological hazards of AER effluent is unknown even if those harmful refractory chemicals in SE have been effectively removed by AER.

Based on a pilot-scale operation (2.2 m³ d⁻¹, 185 cycles, 37,000 BV, 1.5 years), this study evaluated the long-term performance of a commercially available conventional AER for removal of DOM in SE, with particular emphasis on the potential effect of DOM fouling on the ion exchange properties and performance during the long-term operation. In addition, the potential ecological effect of tertiary effluent (TE) on freshwater fishes, an overall water quality evaluation on the TE produced by the AER process, and the application of the results of this investigation were also discussed.

2. Materials and methods

2.1. The AER and the operation of the pilot plant

In this study, the conventional AER in the chloride form, 201 × 4 resin (16–50 mesh. Referred resin products: Amberlite IRA-401,

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