Science of the Total Environment 461–462 (2013) 480–498

Contents lists available at SciVerse ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Treatment of micropollutants in municipal wastewater: Ozone or powdered activated carbon?

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HIGHLIGHTS

- Micropollutants are efficiently removed by both ozone and powdered activated carbon.
- Specific substances were removed more efficiently by ozone.
- Powdered activated carbon effectively removed a wider range of pollutants.
- Both treatments significantly reduced the toxicity of WWTP effluent.
- Both treatments are feasible for use in municipal WWTPs.

ARTICLE INFO

Article history:
Received 12 February 2013
Received in revised form 9 May 2013
Accepted 9 May 2013
Available online 8 June 2013

Editor: Simon James Pollard

Keywords:
Organic micropollutant
Pharmaceutical
Wastewater treatment
Ozone
Powdered activated carbon
Effluent toxicity

ABSTRACT

Many organic micropollutants present in wastewater, such as pharmaceuticals and pesticides, are poorly removed in conventional wastewater treatment plants (WWTPs). To reduce the release of these substances into the aquatic environment, advanced wastewater treatments are necessary. In this context, two large-scale pilot advanced treatments were tested in parallel over more than one year at the municipal WWTP of Lausanne, Switzerland. The treatments were: i) oxidation by ozone followed by sand filtration (SF) and ii) powdered activated carbon (PAC) adsorption followed by either ultrafiltration (UF) or sand filtration. More than 70 potentially problematic substances (pharmaceuticals, pesticides, endocrine disruptors, drug metabolites and other common chemicals) were regularly measured at different stages of treatment. Additionally, several ecotoxicological tests such as the Yeast Estrogen Screen, a combined algae bioassay and a fish early life stage test were performed to evaluate effluent toxicity. Both treatments significantly improved the effluent quality. Micropollutants were removed on average over 80% compared with raw wastewater, with an average ozone dose of 5.7 mg O_3 l^{-1} or a PAC dose between 10 and 20 mg l^{-1}. Depending on the chemical properties of the substances (presence of electron-rich moieties, charge and hydrophobicity), either ozone or PAC performed better. Both advanced treatments led to a clear reduction in toxicity of the effluents, with PAC-UF performing slightly better overall. As both treatments had, on average, relatively similar efficiency, further criteria relevant to their implementation were considered, including local constraints (e.g., safety, sludge disposal, disinfection), operational feasibility and cost. For sensitive receiving waters (drinking water resources or recreational waters), the PAC-UF treatment, despite its current higher cost, was considered to be the most suitable option, enabling good removal of most micropollutants and macropollutants without forming problematic by-products, the strongest decrease in toxicity and a total disinfection of the effluent.

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http://dx.doi.org/10.1016/j.scitotenv.2013.05.034

1. Introduction

About 3000 pharmaceutical compounds and more than 300 pesticides and biocides are commercially available in Switzerland (OPBio, 2005; OPPh, 2010; Swissmedic, 2012). They can enter urban sewer systems via human excretion in urine and feces, by improper disposal, or through leaching of pesticides and biocides from urban areas during rain events. In conventional wastewater treatment plants (WWTPs), many of these hydrophilic organic compounds are poorly removed (Choubert et al., 2011; Deblonde et al., 2011), and are thus characterized by a relatively constant input at low concentrations (ng l^{-1} to μg l^{-1}) into the aquatic environment. As most of these substances are designed to be biologically active, they can affect sensitive aquatic organisms even at very low concentrations (Santos et al., 2010), hence the name "micropollutant". For instance, endocrine effects on fish and mussel populations such as intersex, reproductive disruption or feminization of males have been observed in rivers downstream of municipal WWTP outfalls (Alan et al., 2008; Gagné et al., 2011; Tetreault et al., 2011; Tyler and Jobling, 2008; Vethaak et al., 2005; Woodling et al., 2006). These effects were attributed to the release of endocrine-active chemicals such as the synthetic estrogen 17α-ethinylestradiol (found in contraceptive pills), natural estrogens estrone and 17β-estradiol or nonylphenol. Furthermore, as lakes and rivers are used in many places for drinking water supply, pharmaceuticals and pesticides can therefore be found in tap water at very low concentrations, even after drinking water treatment (Huerta-Fontela et al., 2011; Mompelat et al., 2009; Stackelberg et al., 2007). Acute human health effects are not expected (Webb et al., 2003), but effects of long term exposure are unknown and, therefore, the release of these compounds into the environment should be avoided.

Effluents of WWTPs are the main source of pharmaceuticals in the aquatic environment (Bartelt-Hunt et al., 2009; da Silva et al., 2011). Since it is unrealistic to limit the consumption of pharmaceuticals, additional steps during wastewater treatment are one of the best options to reduce the release of these compounds into surface waters. Currently, two main technologies with a potential for large-scale application in terms of efficiency, costs and energy requirements have been identified (Abegglen and Siegrist, 2012; Joss et al., 2008): oxidation of micropollutants with ozone or adsorption onto activated carbon.

Through the strong oxidative properties of ozone and of the hydroxyl radicals produced spontaneously in its decomposition, ozonation was found to degrade efficiently most micropollutants present in treated wastewater with a dose of 3–8 mg O_3 l^{-1} (Hollender et al., 2009; Lee et al., 2012; Nakada et al., 2007; Reungoat et al., 2010, 2012; Rosal et al., 2010). A potential disadvantage of this process is the formation of unknown reactive by-products due to partial oxidation of the compounds and reaction with matrix components (von Gunten, 2003a). For example, undesirable toxic oxidation by-products such as nitrosamines N-Nitrosodimethylamine (NDMA), bromate or formaldehyde can be formed (Hollender et al., 2009; Richardson, 2003; Wert et al., 2007), potentially increasing the toxicity compared to non-ozonated wastewater (Petala et al., 2006, 2008; Stalter et al., 2010a, 2010b). These oxidation products are usually more easily biodegradable and can be partially removed during biological post-filtration (Hollender et al., 2009; Richardson et al., 1999; Stalter et al., 2010a, 2010b).

Activated carbon allows removal of a broad spectrum of micropollutants via adsorption to its high specific surface area and is thus widely used in drinking water treatment (Snyder et al., 2007; Westerhoff et al., 2005). As organic matter present in wastewater effluent can compete for adsorption sites, larger amounts of activated carbon are required. The efficiency of granular activated carbon (GAC) filtration to remove micropollutants has been studied in some WWTPs, showing a mitigated efficiency depending on the compound and the frequency of GAC regeneration/replacement (Grover et al., 2011; Nguyen et al., 2012; Reungoat et al., 2010, 2012; Snyder et al., 2007). Powdered activated carbon (PAC) adsorption, with a dosage of 10–20 mg l^{-1}, has been proposed as a more efficient alternative compared to GAC treatment (Boehler et al., 2012; Metzger et al., 2005; Nowotny et al., 2007; Serrano et al., 2011). However, to date, very few large scale studies evaluating the efficiency of micropollutant removal via PAC treatment in municipal wastewater have been reported.

In order to find a feasible and efficient solution for the removal of pharmaceuticals and pesticides in wastewater, a global pilot study was conducted at the municipal WWTP of Lausanne, Switzerland. The goals were to evaluate and compare the efficiency of ozonation and PAC adsorption (i) to remove a broad range of micropollutants in WWTP effluents, and (ii) to reduce ecological impacts of the effluent. Finally, we aimed to determine the feasibility of these advanced treatments at the WWTP scale in terms of operation, energy consumption and costs.

2. Materials and methods

2.1. Lausanne wastewater treatment plant

The municipal WWTP of Lausanne, Switzerland, is the largest in the Lake Geneva watershed and treats on average 95,000 m^3 d^{-1} of wastewater representing a population equivalent (PE) of 220,000 individuals. The sewer system is only partially separated, collecting a significant amount of urban runoff during rain events. The watershed includes a major hospital and several clinics, which are a potential source of specific pharmaceuticals. The wastewater treatment consists of pre-treatments (grit removal and screening at 1 cm), primary clarifiers, biological activated sludge treatment (AS, sludge age of 2 d) without nitrification, or, for 5% of the flow, a moving bed bioreactor (MBBR) with partial to complete nitrification (<1 mg N-NH_4 l^{-1}). In both treatments, phosphorus is removed by precipitation with iron chloride. Treated wastewater (WWTP effluent) is then discharged in Lake Geneva, which is the main drinking water reservoir for more than 600,000 inhabitants (www.cipel.org, last accessed 7 May 2013).

2.1.1. Ozonation pilot plant

The pilot plant for ozonation was designed to treat a maximum flow rate of 100 l s^{-1} (13,000 PE) and consisted of a plug flow reactor (volume of 129 m^3) separated into four chambers (nine compartments) in series (Fig. 1a) to assure optimal hydraulic conditions and a minimal reaction time of 20 min. Characteristics of the feed water (effluent of the conventional WWTP) are presented in Table 1. Ozone-containing gas (2–14% w/w) was continuously produced by an ozone generator (Effizon SMO 600 from ITT Wedeco, Wallisellen, Switzerland) fed with pure oxygen. 60% of the gas was injected counter currently into the 1st or 2nd chamber depending of the water flow rate and 40% in the 3rd chamber. The reaction time in the reactor ranged between 20 and 60 min. The ozone dosage was automatically adjusted to the water quality (oxidative demand) by varying the gas flow to maintain a constant residual concentration of dissolved ozone (around 0.1 mg O_3 l^{-1}), measured with an online sensor (AMI codes II, from Swan, Hinwill, Switzerland), and confirmed with a second probe (AquaTector from Mesin, Winterthur, Switzerland) at the outlet of the 3rd chamber. Corresponding initial ozone doses varied between 2 and 13 mg O_3 l^{-1}, with on average 5.7 mg O_3 l^{-1}. Ozone concentrations in the feed and off gas were continuously measured with BMT 964 probes (Berlin, Germany). The transfer efficiency of ozone into the dissolved phase was from 70 to over 90% depending on the gas flow. In this paper, the ozone dose refers to the amount of gaseous ozone injected and not to the ozone dissolved into the water. The remaining gaseous ozone was catalytically converted to oxygen before its release into the atmosphere. The effluent of the ozone reactor was then filtered through a rapid sand filter (flux of 8 m h^{-1}, characteristics described in the next section) with biological activity to remove reactive oxidation products.