



## Micropollutant removal by advanced oxidation of microfiltered secondary effluent for water reuse



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### ABSTRACT

The removal of micropollutants (MPs) from secondary municipal wastewater by an advanced oxidation process (AOP) based on UV irradiation combined with hydrogen peroxide (UV/H<sub>2</sub>O<sub>2</sub>) has been assessed through pilot-scale experiments incorporating microfiltration (MF) and reverse osmosis (RO). Initial tests employed low concentrations of a range of key emerging contaminants of concern, subsequently focusing on the highly recalcitrant compound metaldehyde (MA), and the water quality varied by blending MF and RO permeate.

Under optimum H<sub>2</sub>O<sub>2</sub> and lamp power conditions, AOP achieved significant removal (>99%) of N-nitrosodimethylamine (NDMA) and endocrine disrupting compounds (EDCs) for all waters. Pesticide removal, in particular metaldehyde, atrazine and 2,4,5-trichlorophenoxyacetic acid, was dependent on water transmittance (UVT), and levels of TOC and other hydroxyl radical (<sup>•</sup>OH) scavengers. Further analysis of MA removal showed UVT, hydraulic retention time and H<sub>2</sub>O<sub>2</sub> dose to be influential parameters in determining degradation as a function of UV dose.

A cost assessment revealed energy consumption to account for 65% of operating expenditure with lamp replacement contributing 25%. A comparison of three unit process sequences, based on MF, RO, AOP and activated carbon (AC), revealed MF–RO–AOP to be the most cost effective provided management of the RO concentrate stream incurred no significant cost. Results demonstrated AOPs to satisfactorily reduce levels of the more challenging recalcitrant MPs to meet stringent water quality standards for wastewater reuse, but that practical limitations exist and the cost penalty is significant.

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### 1. Introduction

Over the past 20 years, there has been increased awareness of the incidence of certain micropollutants (MPs) in the environment and a concomitant increase in the study of their fate and removal by water treatment technologies. This is particularly germane to recovered and reused wastewater, which demands implementation of advanced treatment technologies to attain water of a quality comparable to treated raw waters with respect to the MP levels.

MPs can cause potentially adverse health effects at concentrations in the milligram/nanogram per litre range [14]. They may originate from natural or anthropogenic sources, such as industry, agriculture and domestic households. Environmental quality standards for several organic MPs that may be released into surface waters have been promulgated via regulations such as the European Water Framework Directive (WFD) [8]. However, there are

several other emerging chemicals of concern that are currently unregulated and not monitored, particularly in wastewater effluents. Specifically, the difficulty in effectively managing and reliably treating diffuse sources of pollution poses a significant challenge to wastewater reclamation, especially if intended for human consumption.

Research into removal or fate of MPs has mainly focused on organic compounds often classified as disinfection by-products (DBPs), pharmaceuticals, pesticides, and endocrine disrupting compounds (EDCs). EDCs are widespread in the environment and consist of natural oestrogens (estrone (E1) and 17β-estradiol (E2)) along with synthetic xenoestrogens (17 α-ethynylestradiol), phthalates and many more, known to affect/interfere with the action of hormones in the endocrine system [15]. Sewage effluents have been identified as being a major source of natural estrogenic chemicals in the aquatic environment [7]. Research conducted over the past 15–20 years has highlighted the limited removal capability of conventional wastewater treatment systems to levels that are deemed acceptable.

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The use and type of pesticides for wide-ranging applications within the agricultural sector has developed significantly over the past 50 years, and these can readily contaminate raw drinking water sources through run-off. Regulatory limits for pesticide levels in EU drinking waters are  $0.1 \mu\text{g L}^{-1}$  for an individual pesticide and  $0.5 \mu\text{g L}^{-1}$  in total. Among those of concern in drinking waters are atrazine and, most recently, metaldehyde (MA). The latter has been shown to be widely detected in UK water sources [38] and is not readily removed by conventional water treatment technologies [2].

Advanced oxidation processes (AOPs), characterised by the generation of highly reactive, non-selective hydroxyl radicals ( $\cdot\text{OH}$ ), offer a promising alternative to conventional treatment for removing organic constituents in contaminated waters. The UV/ $\text{H}_2\text{O}_2$  process is amongst the most studied AOP and reported to be effective at degrading recalcitrant MPs such as NDMA [25] and various pharmaceuticals and EDCs [4,20,34] for indirect potable reuse (IPR) and, in the case of surface water treatment generally, metaldehyde [1]. Such a process installed downstream of the membrane filtration step of an IPR scheme to remove traces of compounds permeating the RO membrane [31] can potentially achieve full mineralisation of MPs [23], whereas reverse osmosis (RO) leaves MPs unchanged in the waste stream thus demanding further management. However, both RO and AOPs are energy and chemically intensive and thus incur a significant cost. Whilst previous studies of AOPs at bench scale have elucidated degradation mechanisms and  $\text{H}_2\text{O}_2$  dosing requirements (or hydroxyl rate constants) for MP removal [32,29,18,3], representative costs and performance from larger scale demonstration against real waters have not been quantified.

This study aims to determine both the effectiveness and cost of a UV/ $\text{H}_2\text{O}_2$  process for treating MPs of emerging concern, and the most highly recalcitrant of these in particular - MA, which currently presents a significant challenge to the water industry. Impacts of key parameters, specifically feedwater transmittance, are assessed and process costs compared with those of other candidate processes recently demonstrated on the same large pilot scale.

## 2. Materials and methods

### 2.1. Pilot plant and chemical reagents

The pilot plant (Fig. 1), details of which are provided elsewhere [27], treated  $600 \text{ m}^3 \text{ d}^{-1}$  of final effluent from a conventional activated sludge (CAS)-based wastewater treatment works. The process consisted of a pre-filter, microfiltration (MF), reverse osmosis (RO) and an advanced oxidation process (AOP), based on a combination of UV irradiation and  $\text{H}_2\text{O}_2$  dosing (UV/ $\text{H}_2\text{O}_2$ ), downstream of the MF (AOP1) and the RO (AOP2).

Each AOP comprised a flow-through UV reactor (Trojan UVPhox™, Model 12AL30) equipped with 12 low-pressure/high-output (LP/HO) amalgam lamps with nominal output power from the lamps varies from 60% to 100%. AOP1 reactor was fitted with an automatic sleeve wiping and clean-in-place (CIP) system, the

latter using citric acid for iron fouling mitigation or scaling on the quartz sleeves (i.e. fouling).  $\text{H}_2\text{O}_2$  was dosed upstream of both UV reactors with static mixers in-line. UV transmittance (at 254 nm), intensity and power were monitored online for each reactor, along with temperature, and independent flowmeters fitted to each stream. Selected MPs (Table 3) were dosed at concentrations generally between  $0.2$  and  $2 \mu\text{g L}^{-1}$  into the feed via glass ampoules and/or pipetted from solutions of the compounds dissolved in water/acetone, the latter adding up to 15% to the organic carbon but assumed to be inert under the AOP operating conditions employed, based on previous studies [13]. All MPs were of an analytical grade and provided by Sigma Aldrich, with the exception of the EDC compounds (E1, E2 and EE2) which were from QMX Laboratories (Essex, UK).

### 2.2. Experimental procedure

Feed water quality was varied by testing either the MF, RO or an equal blend of the two permeate streams (Table 1). MPs were then dosed at concentrations based on those typically arising in secondary wastewater, as identified in the literature [21,22] and regulatory guidelines [37], or else constrained by their limit of detection as advised by the laboratory (Thames Water Analytical Laboratories, Reading) to allow %removal values to be accurately calculated. Operating conditions for each AOP stream (Table 2) were determined from the optimum UV lamp input power and  $\text{H}_2\text{O}_2$  dose for each AOP stream for efficient removal of target MPs to the required log reduction, based on outcomes of initial scoping trials.

Chemicals were prepared to a required concentration and dosed directly into a  $1 \text{ m}^3$  storage tank, fitted with a submersible mixing pump, for holding the MF/RO permeate or blend. This feedwater was pumped to the UV unit with in-line  $\text{H}_2\text{O}_2$  dosing upstream of a static mixer at flow rates of  $1\text{--}3 \text{ m}^3 \text{ h}^{-1}$ , providing residence times of 120–180 s. An experimental test comprised feed solution mixing for 20 min, to ensure a homogeneous solution, followed by once-through AOP treatment for 20 min. Sampling of the outlet stream was timed to allow for the above residence time.

A daily 5 wt% citric acid CIP was performed on AOP1 to minimise sleeve fouling. The reactor was flushed through with the normal permeate stream following each experiment. Experiments were conducted separately for NDMA and MA to ensure no competition between the two sets of reactions; the EDCs, pesticides and herbicides were combined in the same bulk feedwater solution as applied in other studies [22]. Duplicate samples for each MP compound were taken from the feed tank and triplicate samples collected post-AOP at 5, 10, and 15 min intervals. Tests were repeated for any apparent anomalies.

Sampling and analysis was conducted for MPs along with standard sanitary determinants. All compounds were analysed by Thames Water Laboratories (Reading, UK) using standard methods (APHA, 2005). EDCs, atrazine and terbutryn were analysed by liquid chromatography with mass spectrophotometric detection (LS-MS-MS), ion chromatography (IC) was used to analyse NDMA,

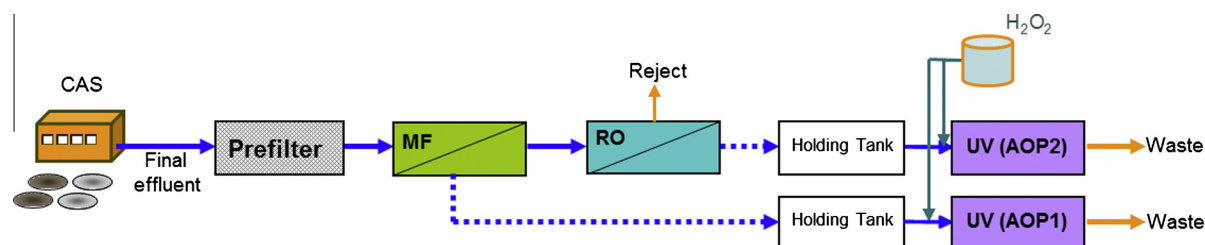


Fig. 1. Pilot plant schematic.

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