



# Integration of advanced oxidation processes at mild conditions in wet scrubbers for odourous sulphur compounds treatment



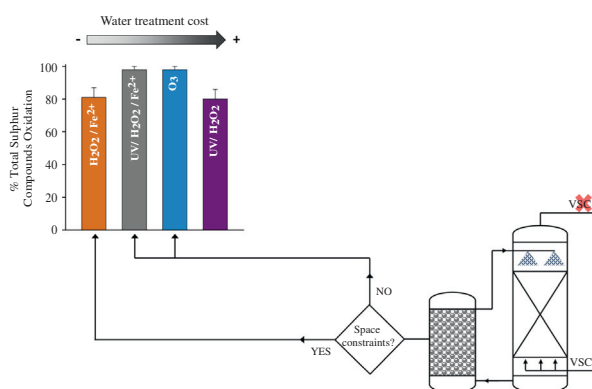
Esther Vega\*, Maria J. Martin, Rafael Gonzalez-Olmos

LEQUIA, Institute of the Environment, University of Girona, Campus Montilivi, E-17071 Girona, Catalonia, Spain

## HIGHLIGHTS

- All the evaluated AOPs demonstrated to be suitable to remove the studied VSC.
- O<sub>3</sub> and photo-Fenton are the most efficient treatments for VSC oxidation.
- The increase in SO<sub>4</sub><sup>2-</sup> indicates an important mineralisation degree.
- Fenton treatment has lower total cost than the other studied AOPs.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The effectiveness of different advanced oxidation processes on the treatment of a multicomponent aqueous solution containing ethyl mercaptan, dimethyl sulphide and dimethyl disulphide (0.5 mg L<sup>-1</sup> of each sulphur compound) was investigated with the objective to assess which one is the most suitable treatment to be coupled in wet scrubbers used in odour treatment facilities. UV/H<sub>2</sub>O<sub>2</sub>, Fenton, photo-Fenton and ozone treatments were tested at mild conditions and the oxidation efficiency obtained was compared. The oxidation tests were carried out in magnetically stirred cylindrical quartz reactors using the same molar concentration of oxidants (hydrogen peroxide or ozone). The results show that ozone and photo-Fenton are the most efficient treatments, achieving up to 95% of sulphur compounds oxidation and a mineralisation degree around 70% in 10 min. Furthermore, the total costs of the treatments taking into account the capital and operational costs were also estimated for a comparative purpose. The economic analysis revealed that the Fenton treatment is the most economical option to be integrated in a wet scrubber to remove volatile organic sulphur compounds, as long as there are no space constraints to install the required reactor volume. In the case of reactor volume limitation or retrofitting complexities, the ozone and photo-Fenton treatments should be considered as viable alternatives.

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## 1. Introduction

During the last years, the emission of unpleasant odours produced in waste water treatment plants, among other facilities,

has become a public concern. One of the main causes of malodour episodes in these facilities is the emission of volatile sulphur compounds (VSCs), especially in the sludge management areas (Dincer and Muezzinoglu, 2008).

One of the most extended treatments for the removal of odourous compounds, especially VSCs, is the use of oxidative wet scrubbers using hypochlorite (Van Durme et al., 1992; Kastner

\* Corresponding author. Tel.: +34 972418162; fax: +34 972418150.

E-mail address: [esther.vega@udg.edu](mailto:esther.vega@udg.edu) (E. Vega).

et al., 2003; Sanchez et al., 2006; Vilmain et al., 2013). However, the formation of chlorinated by-products in the oxidative process has been reported as a negative aspect by several authors (Myslinski et al., 2000; Lin and Chang, 2005). Thus, other oxidants such as hydrogen peroxide (Charron et al., 2004; Couvert et al., 2006; Biard et al., 2009) and ozone (Hwang et al., 1994; Biard et al., 2009), which can promote advanced oxidation processes (AOPs), have been tested. Gaseous VSC removal integrating AOPs with wet scrubbers involves mass transfer with simultaneous reaction due to the generation of highly reactive oxidants, such as hydroxyl radicals. These radicals have a higher oxidation potential (2.8 eV) than other oxidising species such as hydrogen peroxide or chlorine (1.78 and 1.36 eV, respectively) (Metcalf, 2003). Because the VSC concentration in the liquid phase decreases as it is degraded, VSC mass transfer from the gas phase to the liquid phase is expected to be continuous (Tokumura et al., 2012).

The reported studies about coupling AOPs with wet scrubbers have been mainly focused on the removal of chlorinated solvents (Dewulf et al., 2001; Feitz et al., 2002) and aromatic compounds (Tokumura et al., 2012). Concerning sulphur compounds, the coupling of wet scrubber with AOPs has been limited to photocatalysis with  $\text{TiO}_2$  (Liu et al., 2010; Lu et al., 2012) and the use of  $\text{O}_3/\text{H}_2\text{O}_2$  (Biard et al., 2009, 2011). However, there is a lack of comparative studies about which AOP is the most suitable to be integrated in wet scrubbers, and about the efficiency of AOPs to remove VSC from aqueous solution.

Processes such as Fenton ( $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ), photo-Fenton ( $\text{UV}/\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ),  $\text{UV}/\text{H}_2\text{O}_2$  or ozonation are included among the most studied AOPs (Parsons, 2004). The effectiveness of most of  $\text{H}_2\text{O}_2$ -based AOPs is usually improved by acidic conditions. Nevertheless, several authors stated that these processes can be carried out at neutral pH as long as the iron is in low concentration and water contains dissolved organic matter which facilitates that an iron fraction remains in solution due to formation of complexes (Wang, 2008; Vermilyea and Voelker, 2009; De la Cruz et al., 2012). Thus, saving consumption reagents in the pH adjustment leads to a decrease in the process operational costs.

The main objective of the present work was to assess which is the optimal AOP that can be coupled into a wet scrubber for odour treatment. Thus, the advanced oxidation of an aqueous multicomponent mixture of VSC composed by ethyl mercaptan (ETM), dimethyl sulphide (DMS) and dimethyl disulphide (DMDS) was investigated. This study was focused on the use of  $\text{UV}/\text{H}_2\text{O}_2$ ,  $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ,  $\text{UV}/\text{H}_2\text{O}_2/\text{Fe}^{2+}$  and  $\text{O}_3$  at mild conditions (room temperature and pressure and pH around neutral). A comparative assessment regarding the oxidation efficiency and the total cost of each treatment was carried out.

## 2. Materials and methods

### 2.1. Chemicals

All sulphur compounds used in this study (ETM, DMS and DMDS) were supplied by Acros Organics (Belgium) and Merck (Germany) with purities higher than 99%. Fenton and photo-Fenton oxidation were carried out using iron chloride ( $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ ) and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) of 30% (w/w) purchased from Panreac and Scharlau (Spain), respectively.

### 2.2. Experimental setup

The experimental set-up, shown in Fig. SM-1 of supplementary material (SM), was used in all oxidation experiments. The steel container can hold up to 6 cylindrical quartz reactors (diameter = 1.8 cm; height = 17.5 cm) which are magnetically stirred and

is equipped with a thermostatic bath. The device can also be fitted with a UV lamp in the centre when required. After adding 25 mL of a synthetic multicomponent solution containing  $0.5 \text{ mg L}^{-1}$  of each VSC (ETM, DMS and DMDS) in distilled water, the cylindrical quartz reactors were capped with a rubber septum, leaving a headspace volume (HSV) of 5 mL. The gas tightness of the cylindrical reactors was checked. Neither leakage nor permeation were detected since the concentration of the VSC in the HSV remained constant for 30 min. Since the ratio gas-to-liquid volume is low and the solution is stirred, the partition of the VSC among the liquid and gas phases was fast, achieving steady concentration in the HSV is less than 2 min. Furthermore, the analysis of HSV sample according to the previous gas phase calibrations (Vega et al., 2013) revealed that only a small fraction of the VSC initially in solution was transferred to the gas phase (0.4% for ETM and DMDS and 0.3% for DMS). In all oxidation experiments, the oxidant was injected through the rubber septum once the partitioning between liquid and gas phase had reached the steady state.

Photoirradiation experiments ( $\text{UV}/\text{H}_2\text{O}_2$  and  $\text{UV}/\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ) were carried out using a low-pressure Hg lamp TNN 15/32 Heraeus (15 W) with a wavelength emission at 254 nm and an irradiation intensity of  $0.07 \text{ W cm}^{-2}$ . The filled reactors were distributed equidistant around the UV lamp (at 4 cm) and the reactor area belonging to gas phase was covered with aluminium foil in order to avoid the UV oxidation of the VSC in gas phase during the experiments.

A reactor was taken off from the container every two minutes and the concentration of VSC was determined as explained in analytical procedures. Triplicate experiments were carried out at  $15 \text{ }^\circ\text{C}$  and at free pH ( $6.7 \pm 0.2$ ). In order to compare the oxidative efficiency of the studied treatments, the same molar concentration of both hydrogen peroxide and ozone ( $0.20 \text{ mM}$ ) was used. This corresponds to the stoichiometric amount of hydrogen peroxide needed for the complete oxidation of the synthetic multicomponent solution.

The influence of hydrogen peroxide concentration ( $0.2\text{--}4 \text{ mM}$ ) in the VSC oxidation was tested with  $\text{UV}/\text{H}_2\text{O}_2$  treatment. In the Fenton treatment ( $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ), the effect of  $\text{Fe}^{2+}$  concentration ( $0.004\text{--}0.018 \text{ mM}$ ) was also studied. In the Fenton treatment, the reactors were covered with aluminium foil to avoid the incidence of the natural light during the reaction. Photo-Fenton treatment was performed in a similar way as Fenton treatment but in the presence of UV light and without aluminium foil covering the liquid phase.

The ozone treatment was carried out using the ozone stream, which was generated by ozone generator (Anseros COM-AD-02, Actualia) coupled with Actualia HCM 2003 oxygen concentrator. An ozone flow of  $10 \text{ g O}_3 \text{ h}^{-1}$  was bubbled in water during a certain time until the amount of ozone in solution, which was measured by the colorimetric indigo method, was  $9.8 \text{ mg L}^{-1}$  ( $0.20 \text{ mM}$ ). After that, the ozone solutions were immediately used in order to avoid the decomposition of the ozone (Lovato et al., 2009). The experimental conditions used for each treatment are summarised in Table SM-1.

### 2.3. Analytical procedure

The VSCs were analysed by gas chromatography (CP-3800, Varian) using a pulse flame photometric detector (PFPD). Further details about the analytical methodology are presented in Table SM-2. For the calibration, 6 standard solutions of VSCs were prepared. The solutions were placed in the quartz reactors with the same headspace-to-liquid ratio under identical temperature and stirring conditions of the oxidation experiments. Once the steady state was reached (2 min),  $100 \text{ } \mu\text{L}$  of the HSV were sampled and injected into the GC/PFPD. The calibration curves were obtained by plotting the GC/PFPD response against the standard solution

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