

**Intensification of ozonation processes in a novel, compact, multi-orifice oscillatory baffled column**

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## **Abstract**

A novel approach for the intensification of ozonation of water and wastewater is presented using a highly efficient and compact Multi-Orifice Oscillatory Baffled Column (MOBC) ozonation contactor. The MOBC uniquely yielded full (i.e. 100%) use of the ozone supplied with a very short (2.25 min) liquid contact time under continuous operation and reducing the need of further gas-liquid contacting equipment downstream from the MOBC. The increased performance of the MOBC ozonation reactor was benchmarked against a bubble column (BC) design and resulted in 20% increase on the rate of *p*-hydroxybenzoic acid (*p*-HBA) degradation, 75% increase in the rate of mineralization of *p*-HBA per mole of ozone consumed, and 3.2-fold increase in the rate of mineralization of *p*-HBA per mole of ozone supplied. This results from the very small size of bubbles (few hundreds of microns) and enhanced gas-liquid mass transfer and hold-up generated in the presence of small fluid pulsations and orifice baffles.

**Keywords:** Multi-orifice oscillatory baffled column; ozonation; *p*-hydroxybenzoic acid; process intensification; water and wastewater treatment.

## 1. Introduction

Ozonation is a mature advanced oxidation process increasingly applied to the treatment and purification of water and wastewater, including the oxidation of micro-contaminants and the inactivation of virus and bacteria [1-7]. Nevertheless, current ozonation technologies can be economically unattractive when they are applied at industrial scale due to the high rate of ozone supply needed to treat contaminated water, the extended contacting time required in the reactor and the bulky size of the installed equipment. Ozone contacting often relies on conventional gas-liquid reactor designs such as bubble columns, packed columns, stirred tank reactors, air-lift reactors, or static mixers or venturi injectors systems usually combined with very bulky gas-liquid holding tanks [8-13]. These approaches require large footprint equipment and high ozone supply demands.

Oscillatory Baffled Columns (OBCs) are an emerging multiphase mixing technology that can effectively intensify many multiphase chemical and biological processes [14-16]. In OBCs, the eddy mixing obtained with small fluid pulsations and periodic restricting baffles distributed in an elongated column, results in the generation of strong radial mixing within the chambers defined by successive baffles. The intense mixing generated is very effective in tuning the bubble size of the dispersed phase and in increasing the contact between immiscible phases (gas-liquid or liquid-liquid) [16,17]. Previous dissolution studies of oxygen, carbon dioxide and ozone in water [14-16,18-20] showed that the mass transfer rates in OBCs is several orders of magnitude higher than in other contactors. The dissolution of oxygen in water in a 26 mm internal diameter OBC was 6-fold faster compared to a bubble column (BC) [18]. The ozone-water mass transfer rate in a semi-batch 25 mm internal diameter OBC equipped with single-orifice baffles was 3-fold higher than in a

BC [20]. Reis et al. described a 1 to 2-fold increase in oxygen mass transfer rate using an oscillating mesotube comparatively to a 50 mm internal diameter oscillatory flow reactor [15,21]. However, an investigation on the effectiveness of ozonation of wastewater in a pilot-scale OBCs, which may lead to full industrial scale reactors, has not been reported.

The novel ozonation reactor concept herein presented is based on a multi-orifice oscillatory baffled column (MOBC) [22] scaled up from single-orifice OBC technology, allowing mixing and hydrodynamic flow conditions produced at laboratory scale to be easily mimicked at pilot and industrial-scale [16]. This study provides proof-of-concept data related to the performance of a continuous 10-L scale MOBC ozonation reactor in the presence of small fluid oscillations. The increased performance of the MOBC ozonation reactor was benchmarked against a BC design, by monitoring the degradation of *p*-hydroxybenzoic acid (*p*-HBA) model water contaminant and total organic carbon (TOC) conversion and their removal efficiencies. The MOBC provides a novel approach in the intensification of water and wastewater treatment with ozone and other gaseous oxidants.

## **2. Material and Methods**

### **2.1. Reagents**

Chemicals were used as received without further purification. *p*-HBA (> 99%) was from Sigma-Aldrich (UK). NaOH and H<sub>2</sub>SO<sub>4</sub> for pH adjustment were Fluka (UK). For HPLC analysis, methanol (HPLC gradient grade) was sourced from Fisher Scientific (UK). Ozone was generated from high-purity oxygen (99.999%, BOC gases, UK) in an OZOMAX, 8Vtt (OZOMAX, Canada), which delivered ozone doses of up to 23 g O<sub>3</sub>/m<sup>3</sup>. The ultra-high purity water used in all experiments and

analytical analysis was sourced from a Millipore Elix 35 water purification system (i.e. type 2 analytical-grade water).

## 2.2. Experimental setup

Continuous flow ozonation experiments were performed in a vertical glass column (diameter 0.150 m, height 0.540 m) having a maximum operating volume of 9.6 L. In the MOBC configuration (Figure 1) the column was equipped with a set of acrylic baffles, based on a previous optimisation study with CO<sub>2</sub> dissolution [16]. Table 1 summarises the design of the baffles.

### Figure 1

### Table 1

Experiments were carried out by continuously injecting ozone, at a concentration of 23 g O<sub>3</sub>/m<sup>3</sup> and at different gas flow rates (2.1 – 4.7 L/min), to a stream of alkaline water (pH 10 ± 0.1, unbuffered) to decompose O<sub>3</sub> to •OH favouring a gas-liquid reaction in the fast kinetic regime (Hatta number higher 0.3), spiked with 50 mg/L of *p*-HBA flowing at a constant flow rate of 4.0 L/min (mean hydraulic time 2.25 min). The pH was monitored throughout the experiments using a Thermo Orion 420 pH meter. The operating volume of the MOBC remained constant at 9.0 L. The concentrations of *p*-HBA and total organic carbon (TOC) were followed by sampling the effluent from the MOBC. Sinusoidal fluid pulsations in the MOBC were imposed using a 125 mm diameter piston attached to the bottom of the column driven by a servo-hydraulic system [16], capable of delivering fluid oscillations of 0–10 Hz frequency ( $f$ ) and 0–10 mm centre-to-peak amplitude ( $x_0$ ). The results presented are for  $f = 2$  Hz and  $x_0 = 10$  mm, corresponding to a modified oscillatory Reynolds

number of  $Re'_o = 20220$  and modified Strouhal number of  $St' = 0.2$  as shown elsewhere [16].

The liquid reactor outlet at the bottom of the column and the reactor inlet at the top section of the column, were driven by two peristaltic pumps (Masterflex 77200-62, Cole-Parmer, USA) that maintained constant flow through the reactor. The gas inlet stream enriched with ozone entered via a perforated tube sparger placed at the column bottom, and the outlet gas stream was directly connected from the top of the column to a thermal ozone destroyer and an ozone analyser. The ozone concentration, at the inlet and outlet gas streams, was continuously monitored with an Ozomat GM-BWA analyser (Anseros, Germany). For ozone measurements in the water phase, AccuVac<sup>®</sup> Ampules (Hach, ozone LR 0.004–0.25 mg/L; ozone MR, 0.005–0.75 mg/L) were immersed in an aqueous sample and analyzed with a Hach Odyssey at 600 nm. Analysis is based on the indigo method [23,24]. All experiments were carried out at atmospheric pressure and room temperature (20°C). Experiments were repeated multiple times at the same conditions to assure results reproducibility. In the BC reactor configuration the same experiments were carried out in the absence of both baffles and fluid oscillations. The ozone consumption was calculated by the difference of the molar flow rate of ozone at the reactors inlet and outlet.

### **2.3. Analytical methods**

Samples collected at appropriate time intervals were filtered through a 0.22  $\mu\text{m}$  cut-off, 33 mm diameter sterile Millex<sup>®</sup> syringe driven filter (Millipore) and injected into a high performance liquid chromatograph (HPLC, Agilent 1100 series) comprising a diode array detector, a mobile phase degassing unit and an autosampler. The optimized operating conditions were: Gemini C18 (250  $\times$  4.6 mm, 5  $\mu\text{m}$ ) column

supplied by Phenomenex, mobile phase constituted by a mixture water/methanol/acetic acid (88/10/2), flow rate 1 mL/min, column temperature 20°C and UV detector  $\lambda = 254$  nm [25,26]. Calibration of the HPLC chromatographic peaks against standards of *p*-HBA resulted in a linear response in the range of concentrations investigated. The organic matter in the samples was characterised in a TOC (Shimadzu TOC-VCPH).

### 3. Results and discussion

Figure 2 shows the influence of ozone flowrate on the degradation and mineralization of *p*-HBA under “continuous flow” operation for both BC and MOBC. The oxidation of *p*-HBA in both MOBC and BC increased with an increase in the ozone-rich gas flow rate from 2.1 to 4.7 L/min, suggesting that the oxidation of *p*-HBA was controlled by the rate of ozone supplied to the reactors. Under this operational regime, the reaction between ozone and *p*-HBA occurred at the gas-liquid interface, under the fast or moderately fast gas-liquid kinetic regime (Hatta number higher 0.3) [25]. The superior performance of the MOBC in comparison to the BC in the oxidation of *p*-HBA, results from using small fluid pulsations and orifice baffles, which gives a significant increase in gas-liquid mass transfer rates [14-16,18-20]. More significantly, the analysis of the composition of inlet and outlet gas streams revealed full, 100% utilization of ozone in the MOBC and only to 60-65% maximum ozone utilization in the BC, revealing that the MOBC was significantly more efficient in utilising the ozone injected in the reactor.

Many ozonation systems are wasteful of either oxidant or space and are inherently inefficient. The oxidation of water contaminants usually benefits from higher rates of ozone supply, however, in practice this is not always possible in conventional, industrial scale, ozonation contactors (e.g., bubble columns), since an

increasing fraction of the ozone supplied usually escapes unreacted at higher gas flow rates. Other ozonation systems utilizing ozone injection through a Venturi tube, a static mixer or other injector systems have very short contact times between the gas and the contaminants, and in consequence they require very bulky downstream holding tanks in which the reaction between the dissolved ozone and the water contaminants is carried out in the slow kinetic regime over a long residence time.

The MOBC presented here is uniquely capable to fully utilise the ozone supplied to the system, as demonstrated from the undetectable ozone concentration in the outlet gas stream (Figure 2b), and this mainly results from two physical phenomena. Firstly, the gas phase in the MOBC is efficiently dispersed into a large number of very small bubbles (in the range of few hundreds of microns, measured by a high speed CCD camera) by the shear produced by the fluid oscillations and orifice baffles, which provided extremely large interfacial area resulting in enhanced mass transfer rates [16]. The visual appearance of the gas-liquid system in the reactor resembled a whitish milky solution, which became fully transparent in the absence of oscillations. Secondly, the strong eddy vortices formed between the baffles in the MOBC are capable of trapping the microbubbles within the liquid (see Graphical Abstract), meaning extended gas-liquid contacting times in the column (large gas hold-up, ~ 20% at  $f = 2$  Hz and  $x_0 = 10$  mm) therefore an increased rate of ozone utilisation [27].

## **Figure 2**

The *p*-HBA mineralisation (i.e. reduction in TOC), this was 3 to 8-fold higher in the MOBC compared to a BC, with a maximum of 18% TOC removal in the MOBC at the highest gas flow rate tested (Figure 2). TOC reduction remained undetectable in



the BC at all gas flow rates tested. Typically, mineralisation of contaminants with ozone is a slow process due to the multistep reaction sequence, however the results in Figure 2 demonstrate that the MOBC is capable of not just oxidising *p*-HBA but also its intermediates (e.g. quinones and carboxylic acids [28]), yielding significantly higher levels of carbon mineralization in comparison to a BC. It can be hypothesised that full mineralisation is possible for extended contacting time in the MOBC.

Figure 3 shows the rates of degradation and mineralization of *p*-HBA in both BC and MOBC, as a function of O<sub>3</sub> supplied (molar flowrate of O<sub>3</sub> fed to the reactor) or consumed (molar flowrate of O<sub>3</sub> fed to the reactor minus the molar flowrate of O<sub>3</sub> at reactor gas outlet) after reaching steady-state conditions in the reactors (note that both reactors were operated in continuous flow). Figure 3a shows that the rate of degradation of *p*-HBA per mole of ozone consumed is equal in both BC and MOBC reactors. This suggests that the reaction stoichiometry is independent of reactor type and that within the observed degree of conversion of *p*-HBA, the ozone demand remains the same in the two reactors. The slope of the best fitting line drawn through the data points yields the inverse of the stoichiometric ratio of the reaction of *p*-HBA with ozone. The stoichiometric ratio determined from our continuous flow experiments in the MOBC and BC at pH 10 was 2.44, which approached the value of 2.0 (pH invariant in the range 2 to 9) reported in a batch reactor [25]. The discrepancy due to the start-up delay in the batch reactor to reach a constant gas hold-up.

In terms of the ozone supplied to each reactor, the MOBC was on average 20% more efficient than the BC (Figure 3c) in the rate of *p*-HBA degradation, but more significant differences between the two reactors were observed in the rate of *p*-HBA mineralization. Figure 3b shows that the rate of TOC removal per mole of ozone

consumed in the MOBC was 75% higher than in the BC, which was evaluated by the ratio of the slopes of the fitting lines. Limitations of the rate of ozone consumption by the reaction in the BC reactor were quickly reached due to gas passing, large bubble size and much smaller gas holdup (approximately 3% [19,22]). In contrast the MOBC reached significantly higher rates of TOC removal. More significantly, Figure 3d shows that at equal rate of ozone supplied to each reactor, the rate of TOC removal in the MOBC was 3.2 fold higher than in the BC. The ozone supplied to MOBC is effectively used in the oxidation of the contaminants and intermediates with zero or very little waste at very high flow rate (Fig. 3) while the BC was ineffective in utilizing all the ozone supplied.

### **Figure 3**

The results herein presented suggest the MOBC is uniquely capable of keeping consistent gas utilization efficiencies independent of the rate of ozone injected into the column reducing the need of further holding gas-liquid equipment. This represents a significant step forward in the intensification of ozonation processes, particularly in water treatment. Conventionally, ozone utilisation efficiencies sharply reduce as the ozone dosage or rate of ozone injection is increased [2,13,29]. Therefore ozonation systems, which are primarily based on adaptation of reactors for ozone disinfection, are normally operated at reduced ozone dosages, which naturally leads to large equipment volume, high consumption rate of gas (air or oxygen) and extended contacting times for removal of higher concentrations of water contaminants.

The MOBC technology approach presented here disrupts conventional ozonation design systems for water treatment and allows significantly higher rates of ozone utilization and process intensification, in a highly compact device. Overall, it is estimated the MOBC achieves a reduction of 5-fold to one-order of magnitude in reactor volume to achieve the same rate of ozone mass transfer obtained in conventional ozonation reactors. Ozonation and ozone-driven treatment processes can be applied to wastewater treatment in the MOBC with lower costs, reduced hydraulic time and less ozone consumption, which can be translated to a smaller carbon footprint. The supplementary energy required for providing the oscillation of the fluid can be evaluated from established OBCs correlations in literature [19] and amounts to less than 1 kWh/m<sup>3</sup> water treated in the MOBC, which would represent an insignificant cost compared to the significant savings due to more effective use of the ozone supplied.

#### **4. Conclusions**

This study presents a novel approach for the intensification of ozonation of water and wastewater using a highly efficient and compact Multi-Orifice Oscillatory Baffled Column ozonation contactor. The MOBC technology approach disrupts conventional ozonation design systems for water treatment and allows significantly higher rates of ozone utilization and process intensification, in a highly compact device. MOBC achieves a reduction of 5-fold to one-order of magnitude in reactor volume to achieve the same rate of ozone mass transfer obtained in conventional ozonation reactors. The increased performance of the MOBC ozonation reactor was benchmarked against a bubble column design and resulted in 20% increase on the rate of p-HBA degradation, 75% increase in the rate of mineralization of p-HBA per mole of ozone

consumed, and 3.2-fold increase in the rate of mineralization of p-HBA per mole of ozone supplied. Ozonation and ozone-driven treatment processes can be applied to wastewater treatment in the MOBC with lower costs, reduced hydraulic time and less ozone consumption, which can be translated to a smaller carbon footprint.

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### **References**

- [1] T.P. Bonacquisti, A drinking water utility's perspective on bromide, bromate, and ozonation, *Toxicology* 221 (2006) 145-148.
- [2] B.Y. Lan, R. Nigmatullin, G. Li Puma, Ozonation kinetics of cork-processing water in a bubble column reactor, *Water Res.* 42 (2008) 2473-2482.
- [3] B. De Witte, H. Van Langenhove, K. Demeestere, K. Saerens, P. De Wispelaere, Ciprofloxacin ozonation in hospital wastewater treatment plant effluent: effect of pH and H<sub>2</sub>O<sub>2</sub>, *Chemosphere* 78 (2010) 1142-1147.
- [4] R. Andreozzi, M. Canterino, R. Marotta, N. Paxeus, Antibiotic removal from wastewaters: The ozonation of amoxicillin, *J. Hazard. Mater.* 122 (2005) 243-250.
- [5] D. Gerrity, S. Gamage, J.C. Holady, D.B. Mawhinney, O. Quiñones, R.A. Trenholm, S.A. Snyder, Pilot-scale evaluation of ozone and biological activated carbon for trace organic contaminant mitigation and disinfection, *Water Res.* 45 (2011) 2155-2165.
- [6] A. Justo, O. González, J. Aceña, S. Pérez, D. Barceló, C. Sans, S. Esplugas, Pharmaceuticals and organic pollution mitigation in reclamation osmosis brines by UV/H<sub>2</sub>O<sub>2</sub> and ozone, *J. Hazard. Mater.* 263 (2013) 268-274.

- [7] R.F. Dantas, M. Canterino, R. Marotta, C. Sanz, S. Esplugas, R. Andreozzi, Bezafibrate removal by means of ozonation: Primary intermediates, kinetics, and toxicity assessment, *Water Res.* 41 (2007) 2525-2532.
- [8] H.M. Letzel, J.C. Schouten, R. Krishna, C.M. van den Bleek, Gas holdup and mass transfer in bubble column reactors operated at elevated pressure, *Chem. Eng. Sci.* 54 (1999) 2237-2246.
- [9] A.K. Bin, B. Duczmal, P. Machniewski, Hydrodynamics and ozone mass transfer in a tall bubble column, *Chem. Eng. Sci.* 56 (2001) 6233-6240.
- [10] Y.Q. Qiu, C.H. Kuo, M.E. Zappi, Performance and simulation of ozone absorption and reactions in a stirred-tank reactor, *Environ. Sci. Technol.* 35 (2001) 209-215.
- [11] V. Farines, S. Baig, J. Albet, J. Molinier, C. Legay, Ozone transfer from gas to water in a co-current upflow packed bed reactor containing silica gel, *Chem. Eng. J.* 91 (2003) 67-73.
- [12] G. Tiwari, P. Bose, Determination of ozone mass transfer coefficient in a tall continuous flow counter-current bubble contactor, *Chem. Eng. J.* 132 (2007) 215-225.
- [13] M.S. Lucas, J.A. Peres, B.Y. Lan, G. Li Puma, Ozonation kinetics of winery wastewater in a pilot-scale bubble column reactor. *Water Res.* 43 (2009) 1523-1532.
- [14] X. Ni, S. Gao, R.H. Cumming, D.W. Pritchard, A comparative-study of mass-transfer in yeast for a batch pulsed baffled bioreactor and a stirred-tank fermenter, *Chem. Eng. Sci.* 50 (1995) 2127-2136.
- [15] N. Reis, R.N. Pereira, A.A. Vicente, J.A. Teixeira, Enhanced gas-liquid mass transfer of an oscillatory constricted-tubular reactor, *Ind. Eng. Chem. Res.* 47, (2008) 7190-7201.
- [16] F.M. Pereira, D.Z. Sousa, M.M. Alves, M. Mackley, N.M. Reis, CO<sub>2</sub> dissolution and design aspects of a multi-orifice oscillatory baffled column, *Ind. Eng. Chem. Res.* 53 (2014) 17303-17316.
- [17] M.R. Mackley, X. Ni, Mixing and dispersion in a baffled tube for steady laminar and pulsatile flow, *Chem. Eng. Sci.* 46 (1991) 3139-3151.

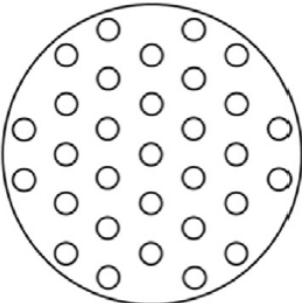
- [18] M.R. Hewgill, M.R. Mackley, A.B. Pandit, S.S. Pannu, Enhancement of gas-liquid mass-transfer using oscillatory flow in a baffled tube, *Chem. Eng. Sci.* 48 (1993) 799-809.
- [19] M.S.N. Oliveira, X. Ni, Gas hold-up and bubble diameters in a gassed oscillatory baffled column, *Chem. Eng. Sci.* 56 (2001) 6143-6148.
- [20] A. Al-Abduly, P. Christensen, A. Harvey, K. Zahng, Characterization and optimization of an oscillatory baffled reactor (OBR) for ozone mass-transfer, *Chem. Eng. Process.* 84 (2014) 82-89.
- [21] N. Reis, P.C. Mena, A.A. Vicente, J.A. Teixeira, F.A. Rocha, The intensification of gas-liquid flows with a periodic, constricted oscillatory-meso tube, *Chem. Eng. Sci.* 62 (2007) 7454-7462.
- [22] K.B. Smith, M.R. Mackley, An experimental investigation into the scale-up of oscillatory flow mixing in baffled tubes, *Chem. Eng. Res. Des.* 84 (2006) 1001-1011.
- [23] H. Bader, J. Hoigné, Determination of ozone in water by the indigo method, *Water Res.* 15 (1981) 449-456.
- [24] J.C. Charpentier, Mass transfer rates in gas-liquid absorbers and reactors, in: T. B. Drew (Ed.), *Advances in Chemical Engineering*, Elsevier, New York, 1981, pp. 2-133.
- [25] F.J. Benitez, J. Beltran-Heredia, J.A. Peres, J.R. Dominguez, Kinetics of *p*-hydroxybenzoic acid photodecomposition and ozonation in a batch reactor, *J. Hazard. Mat.* 73 (2000) 161-178.
- [26] J. Beltran-Heredia, J. Torregrosa, J.R. Dominguez, J.A. Peres, Advanced oxidation processes for the degradation of *p*-hydroxybenzoic acid: 1. photo-assisted ozonation, *J. Chem. Techn. Biotech.* 76 (2001) 1235-1242.
- [27] K. Muroyama, K. Imai, Y. Oka, J. Hayashi, Mass transfer properties in a bubble column associated with micro-bubble dispersions, *Chem. Eng. Sci.* 100 (2013) 464-473.
- [28] N. Al-Hayek, J.P. Eymery, M. Doré, Catalytic oxidation of phenols with hydrogen peroxide, *Water Res.* 19 (1985) 666-675.

[29] M.S. Lucas, J.A. Peres, G. Li Puma, Treatment of winery wastewater by ozone-based advanced oxidation processes ( $O_3$ ,  $O_3/UV$  and  $O_3/UV/H_2O_2$ ) in a pilot-scale bubble column reactor and process economics. *Sep. Purif. Technol.* 72 (2010) 235-241.

As accepted

## Tables

**Table 1** – Geometry of baffles used in the MOBC

		Baffle design
Average number of orifices per baffle	31	
Orifice diameter, mm	10.5	
Equivalent hydraulic diameter,* mm	26.9	
Baffle open area, %	15	
Baffle spacing, mm	40	
Construction material	Acrylic	

\* The definition of the equivalent hydraulic diameter is reported in [16].

AS accepted



## Figure Captions

**Figure 1.** Configuration of the pilot-plant Multi-Orifice Oscillatory Baffled Column used in the ozonation experiments.

**Figure 2.** Influence of ozone flowrate on the degradation and mineralization of *p*-HBA under continuous flow operation (a) BC and (b) MOBC. In the MOBC the fluid was oscillated with  $f = 2$  Hz and  $x_0 = 10$  mm.

**Figure 3.** Rates of degradation and mineralization of *p*-HBA as a function of  $O_3$  supplied or utilised after reaching steady state conditions. The continuous line in (a) and (c) corresponds to cumulative need of 1.0 mole  $O_3$  per mole of *p*-HBA oxidised; whereas dashed line represents cumulative need of 2.0 mole  $O_3$  per mole of *p*-HBA oxidised. (●) MOBC setup; (○) BC setup.

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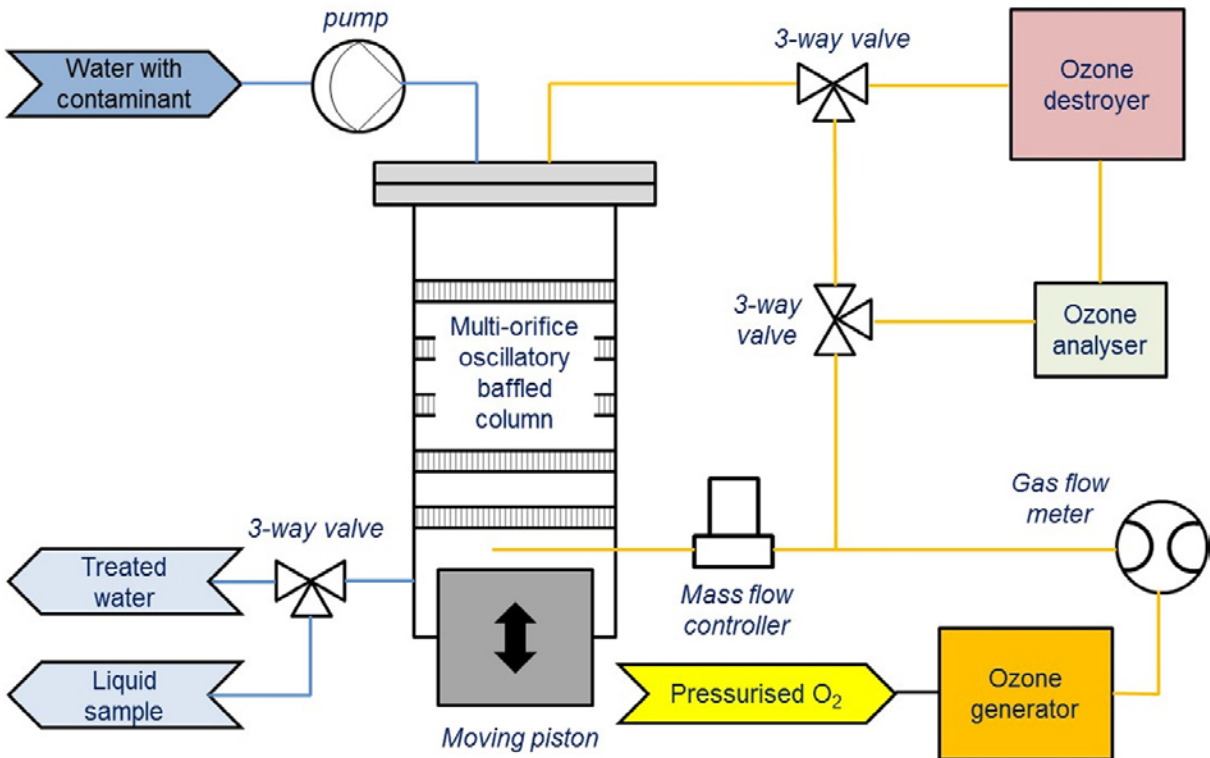


Figure 1

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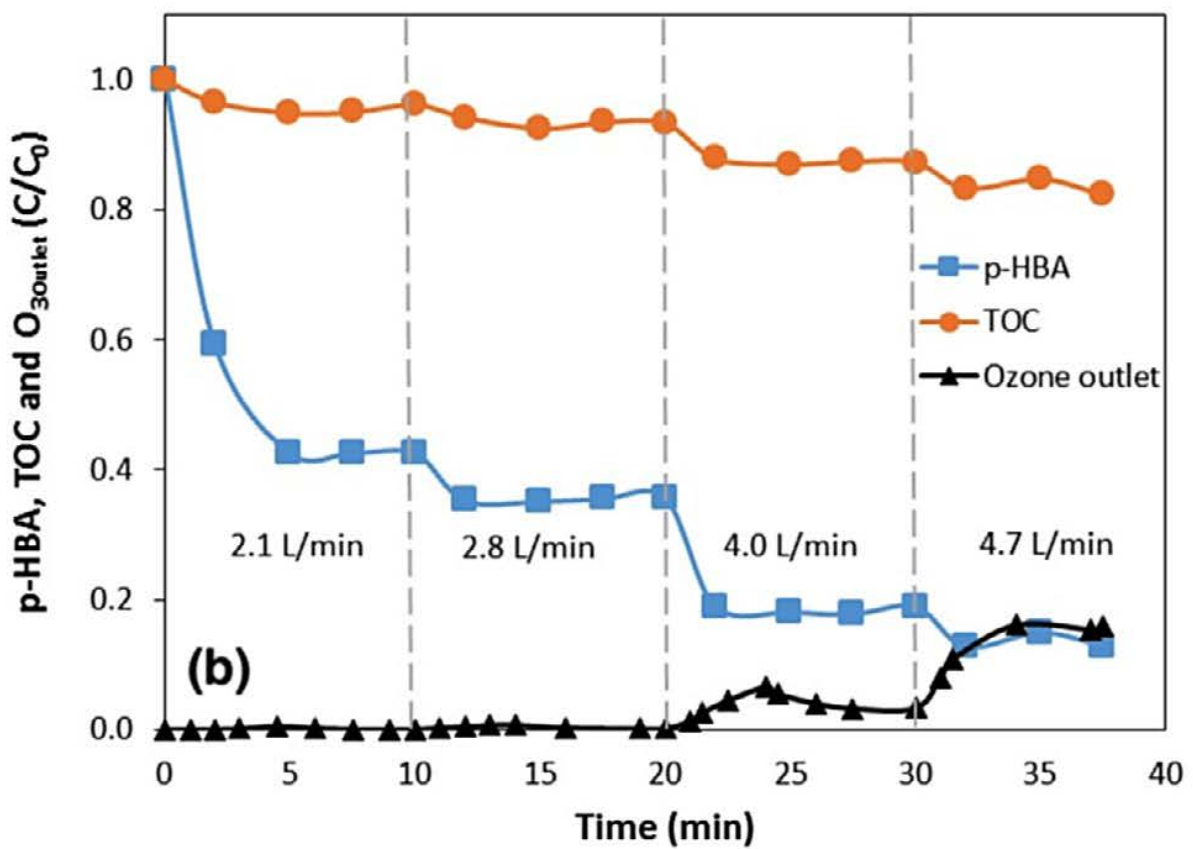
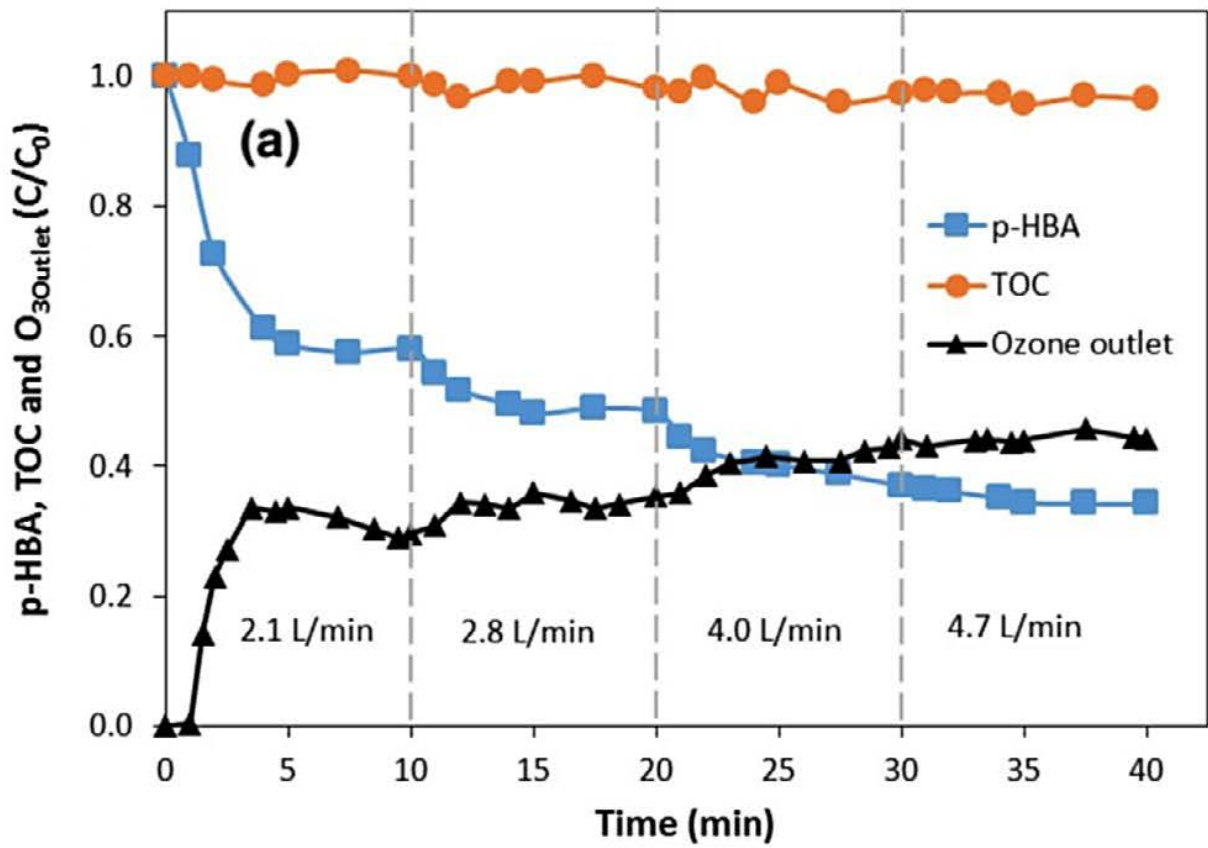


Figure 2

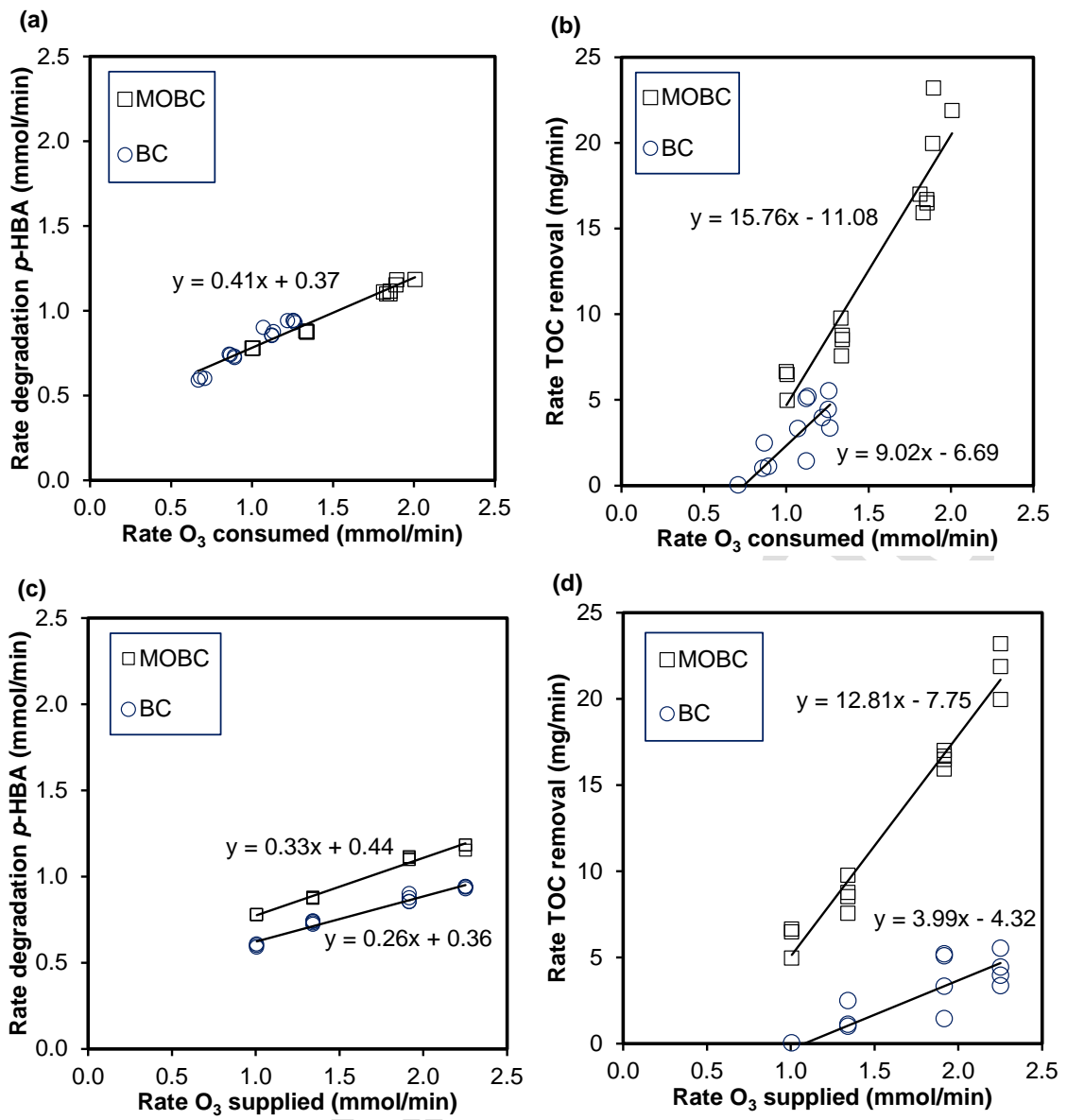


Figure 3