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# Micropollutant degradation, bacterial inactivation and regrowth risk in wastewater effluents: Influence of the secondary (pre)treatment on the efficiency of Advanced Oxidation Processes

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## ABSTRACT

In this work, disinfection by 5 Advanced Oxidation Processes was preceded by 3 different secondary treatment systems present in the wastewater treatment plant of Vidy, Lausanne (Switzerland). 5 AOPs after two biological treatment methods (conventional activated sludge and moving bed bioreactor) and a physiochemical process (coagulation-flocculation) were tested in laboratory scale. The dependence among AOPs efficiency and secondary (pre)treatment was estimated by following the bacterial concentration i) before secondary treatment, ii) after the different secondary treatment methods and iii) after the various AOPs. Disinfection and post-treatment bacterial regrowth were the evaluation indicators. The order of efficiency was Moving Bed Bioreactor > Activated Sludge > Coagulation-Flocculation > Primary Treatment. As far as the different AOPs are concerned, the disinfection kinetics were: UVC/$H_2O_2$ > UVC and solar photo-Fenton > Fenton or solar light. The contextualization and parallel study of microorganisms with the micropollutants of the effluents revealed that higher exposure times were necessary for complete degradation compared to microorganisms for the UV-based processes and inversed for the Fenton-related ones. Nevertheless, in the Fenton-related systems, the nominal 80% removal of micropollutants deriving from the Swiss legislation, often took place before the elimination of bacterial regrowth risk.

## 1. Introduction

Throughout the years, urban wastewater treatment plants (WWTPs) have implemented strategies to eliminate the organic load, chronologically followed by the inorganic one (phosphorus and nitrogen), and presently, the current average treatment stops at the disinfection level. Chlorination was until some time ago the most common disinfection method. However, its use has been connected with trihalomethane (THM) production, a harmful disinfection by-product (DBP) of the reaction with organic matter (Krasner et al., 2009). Hence, treatment at disinfection and decontamination level was turned towards safer "greener" techniques (Michael et al., 2012).

Lately, ozone and ultraviolet light have been widely employed to tackle the issue of microorganism elimination in wastewater (Drinan and Spellman, 2012). UVC alone, combination with $H_2O_2$ and/or $O_3$ are some of the most studied and well-understood Advanced Oxidation Processes (AOPs) for this purpose. The UVC-based processes have a well-established disinfection efficiency when applied in secondary wastewater effluents (Rodríguez-Chueca et al., 2015), but the main concern of bacterial regrowth is yet to be resolved. In overall, the AOPs gained supporters during the last two decades, mainly for their non-selective character against organic matter and microorganisms (Moncayo-Lasso et al., 2012). However, despite the interest gain, the limited number of full-scale applications engulfs the danger of over- or mal-dimensioning of such units, since the pre-treatment process differs from plant to plant.

On the other hand, in less wealthy countries of the developing

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**Abbreviations list**

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| AOP | Advanced Oxidation Process |
| AS | Activated Sludge |
| CF | Coagulation-Flocculation |
| CFU | Colony Forming Units, |
| COD | Chemical Oxygen Demand |
| CPD | Cyclobutane-pyrimidine dimer |
| DBP | Disinfection By-Product |
| DOM | Dissolved Organic Matter |
| EfOM | Effluent Organic Matter |
| Fe/S | Iron-Sulfur |
| HRT | Hydraulic Retention Time |
| MBBR | Moving Bed BioReactor |
| MO | Microorganism |
| MP | Micropollutant |
| MW | Molecular Weight |
| OxOM | Oxidizable Organic Matter |
| PhOM | Photosensitizable Organic Matter |
| POM | Particulate Organic Matter |
| PP | PhotoProduct |
| T | Transmittance |
| ROS | Reactive Oxygen Species |
| S | Synergy |
| SODIS | Solar Disinfection |
| SRT | Sludge Retention time |
| SS | Suspended Solids |
| SUVA | Specific UV Absorbance |
| TOC | Total Organic Carbon |
| TSS | Total Suspended Solids |
| UV | Ultraviolet |
| WW | Wastewater |
| WWTP | Wastewater Treatment Plant |

world, ozone and UV-based techniques are far from applicable. Instead, the use of solar ponds has been widely applied (Von Sperling, 2005), as a simple, and quite efficient method of treating wastewater effluents. As this process has been successfully applied, enhancing its performance with the photo-Fenton reagents could significantly increase the removal of microbial and organic loads (Moncayo-Lasso et al., 2012). Iron and $H_2O_2$ are abundant and environmentally safe, respectively, and the inactivation potential of photo-Fenton can improve the effluent quality while reducing the residence times in such configurations (Von Sperling, 2005) or in Raceway Pond Reactors (RPRs) (Rivas et al., 2015).

In Switzerland, although special effort has been made to effectively remove micropollutants (Giannakis et al., 2015a; Margot et al., 2013, Margot et al. 2011), the recommended strategies have not included the microorganism risk in the design, neither in the legislation. The upgrade of wastewater treatment plants (WWTPs) affects the >10.000 inhabitant equivalent, thus leaving a large number of WWTPs without disinfection units. For instance, the WWTP of Vidy (Lausanne, Switzerland) in its current reconstruction planning, which focuses on the micropollutant removal after the secondary treatment of wastewater (WW), involves the use of activated carbon, ozonation, followed by UVC light, but mostly for degrading the by-products of the previous two installations.

In this work, we take advantage of the simultaneous presence of 3 parallel secondary treatment systems of wastewater treatment in the plant of Vidy, in order to study the effect of secondary pretreatment on the efficiency of AOPs. More specifically, wastewater from Activated Sludge, Moving Bed Bioreactors and Coagulation-Flocculation units (with primary wastewater effluent as control) has been subjected to various oxidation methods. The UVC alone, UVC/$H_2O_2$, Fenton, solar light and (solar) photo-Fenton processes (namely UV-based and Fenton-related processes) were tested on the (immediate) bactericidal removal efficiency, as well as the post-treatment regrowth. Finally, to put things into the real wastewater context, the evolution of 8 micropollutants were monitored, and insights were given on the comparative order of removal of micropollutants (MPs) and microorganism (MO) regrowth risks.

## 2. Materials and methods

### 2.1. Collection of wastewater samples and treatment plant specifications

For the needs of the microbial testing, 6 sampling campaigns were performed. During each visit, wastewater from the following points was collected: i) before secondary treatment (after primary decantation) (PT), ii) after secondary treatment by activated sludge and secondary clarification (AS), iii) after secondary treatment by moving bed bioreactors (MBBR) and iv) after physicochemical treatment by coagulation-flocculation (CF). The aforementioned points can be found in Fig. 1. Each time, a 5-L grab sample was collected and transported immediately to the laboratory for treatment. For the micropollutants, the strategy has been analyzed in a previous work (Giannakis et al., 2015a).

The AS unit has a hydraulic retention time (HRT) of 4 h and sludge retention time (SRT) of 2 days, approximately, without nitrification. The MBBR capacity is only 5% compared to the AS unit, but has a longer retention time and includes a nitrification step. Finally the CF unit is based on chemical coagulation and flocculation process by $FeCl_3$ as coagulant. More details on the different units can be found in Margot et al. (2013).

### 2.2. Employed chemicals and reagents

For the experiments of bacterial inactivation $H_2O_2$ 30%, $FeSO_4 \cdot 7H_2O$ and Titanium (IV) oxysulfate was acquired from Sigma-Aldrich (Switzerland) and $NaHSO_3$ for $H_2O_2$ elimination from Sigma-Aldrich and Acros Organics, respectively. Finally, the plate count agar (PCA) was purchased from Sigma-Aldrich (Switzerland).

### 2.3. Experimental set-up: reactors and apparatus

The experiments are divided in two main groups, namely the UV-based and the Fenton-related ones. For the UV-based experiments, two double-wall, water-jacketed merry-go-round reactors were used in parallel, for the UVC and UVC/$H_2O_2$ experiments, respectively. The water recirculating in the glass reactors was controlled at 22 °C (for protection of the UVC equipment). UVC light was provided by 35-W low pressure UVC lamps (Model: UVI 40 4C P 15/300), with an emission of 350 $\mu W/cm^2$, acquired from UV-Technik Speziallampen (see Supplementary Fig. S1 for the reactor scheme). Among the Fenton-related experiments, the solar only and solar-assisted photo-Fenton process were performed in 100-mL Pyrex glass reactors, placed on magnetic stirrers and constantly agitated by magnetic bars (300 rpm). The Fenton experiment took place in shaded reactors, in the dark, whereas the solar and the photo-Fenton experiments took place in a solar simulator (Atlas, Suntest CPS+). This artificial solar light source was