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Abstract

Water is essential for life in all the levels: humans, animals and plants depend on it for their existence. The unsustainable growth, the changes in the consumption pattern and the climate change have positioned water resources under pressure. In this scenario where the water quality and quantity are a worldwide concern, research and development have analyzed and generated various emerging technologies that can promote the use of alternative sources of water.

In the problematic water pollution scenario mentioned before, advanced oxidation processes (AOPs) emerge as a possible alternative to treat the biologically persistent wastewater improving water quality and therefore restoring the aquatic environment. These processes degrade organic pollutants by forming hydroxyl radicals (OH·) which are highly reactive and non-selective. Nowadays, AOPs include also the processes that involve other radicals as sulfate radicals ($SO_4^{2-}\cdot$).

Ozone application is used in wastewater, disinfection and air treatment to minimize the pollution. This process has two main strengths: on one hand, the strong oxidant potential and secondly, the lack of residues after its application. Ozone can react directly, via molecular pathway or indirectly, via hydroxyl radical. Different criteria have been observed when ozonation is considered or not an AOP, depending on the reacting via.

In this work, ozone has been applied to different non-conventional points of the treatment line, to check if its action could promote the enhancement of the whole treatment. Thus, it has been applied at the outlet of the primary effluent leading us to an improvement in the water quality parameters and in the removal of micropollutants. Moreover, other significant parameters for ozone application as the ozone demand and mass transfer have been studied. Applying ozone doses below 30 mg/L, which corresponds to the first stage of ozone reactions, ensures a high enhancement of water quality. For these low doses, an important and fast oxidation of the OM was achieved. Regarding micropollutants, good removals have been achieved at low ozone transferred doses. Moreover, a strong relationship between the molecular structure of each compound and the second order rate constant k_{O3} has been established, except for Ibuprofen, Ketoprofen and Alachlor where the reaction may undergo via radical pathway.

Afterwards, the study was focus in the application of ozone on the activated sludge matrix. Activated Sludge is the main process and globally used in the WWTPs. However, focusing on the conventional activated sludge (CAS) stage, sludge excess disposal can generate pollution regarding the release to the environment of the pollutants (heavy metals, pathogens, persistent organic pollutants) that are absorbed in the sludge. In addition, the excess of sludge generated during the wastewater treatment highly increase the treatment's cost accounting for the 25-60% of the total wastewater treatment cost. As a consequence, alternative plans for sludge management have been promoted.

In this case, ozone application on sludge showed good performance too, improving the settleability, increasing the solubility of sludge and eliminating micropollutants in both phases (sludge and supernatant). A correlation was observed between the sludge solubilization and IOD

stage completion, concerning COD, TN, pH and $UV_{254.}$ Moreover, even at low doses, this solubilization takes place and it is related with an improvement of the sludge settleability.

On the other hand, the presented results show that ozone might quickly and selectively react with the studied pharmaceuticals present in solid and/or liquid phases despite the fact that the working matrix is a complex sludge matrix presenting a high ozone demand. Good removals have also been achieved regarding PAHs and PBDEs, even at low ozone doses. Regarding surfactants, good removals have been achieved for 4-nonylphenol, non-ionic and anionic surfactants. For all the surfactants eliminations higher than 50% have been reached for the highest TODs 18 to 25 mg/ gSS.

Finally, the combination of ozone application with biological treatments was tested to check if it promotes a synergetic effect. Thus, ozone was applied to the primary effluent which was lately treated by an aerobic biological treatment. To do so, different groups of micropollutants were analyzed: pharmaceuticals, surfactants, pesticides, PBDEs and PAHs. The applied TODs were found below 15 mg/L and the SIR for VSS was 0.4. In this case, good performances were observed at the level of micropollutants. A possible inhibitory effect was detected due to the lack of biomass acclimation. However, in the case of some compounds poorly biodegradable as sulfamethoxazole, a synergetic effect has been observed even though the overall removal was still low, 23%. Lately, ozone was applied to the conventional activated sludge matrix which was subsequently introduced in an anaerobic process to check the enhancement of biogas production. In this case, only two ozone doses below 60 mg O_3/g SS showed better performances than the initial sludge without ozone pretreatment. Since VFA are the precursors of $CH_{4,}$ when the VFA are considered, the highest TOD applied seems to be the more promising. Biodegradability and acute toxicity was studied for the primary effluent before and after ozonation, showing an improvement when the transfer ozone dose increased.