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Application of chemical oxidation for pharmaceuticals removal in wastewater effluents

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Executive Summary

This study was conducted to evaluate the potential of the chemical oxidation processes chlorine dioxide (ClO₂) and ozone (O₃) as tertiary treatment step to remove trace active pharmaceutical ingredients (APIs) in Swedish municipal wastewater treatment plants (WWTPs). Wastewater effluents of varying organic load (COD ~ 30-90 mg/L) were collected from different WWTPs in Sweden to represent different types of biological treatment. Batch experiments were carried out employing ClO₂ (0-20 mg/L) and O₃ (0-12 mg/L) to treat biologically treated wastewater spiked with approx. 1 µg/L mixed APIs. Additionally, treatment with perozone (O₃/H₂O₂) is carried out to enhance API oxidation rate by non-selective hydroxyl radicals. Some of the APIs investigated are shown in Fig. 1 and 2. From the ClO₂ treatment, API removal varied from no significant removal at the highest ClO₂ dose to more than 90% removal with 0.5 mg/L of the oxidant. The low COD effluent exhibited most of the APIs removed at 5 mg/L ClO₂ dose while a significant increase in API removal from the high COD effluent after treatment with 8 mg/L ClO₂. Shown in Fig. 1, treatment with ClO₂ of low COD effluent removes diclofenac by >90% at low oxidant dose of 1.25 mg/L while in high COD effluent around 3 mg/L ClO₂ is needed to reach 90% removal. Repaglinide is also removed at low ClO₂ dose. In comparison to ozonation, the same degree of removal of these APIs is reached but with much higher ozone dose. On the other hand, citalopram and trimetoprim in low COD effluent (Fig. 2) can be removed by ozonation at lower dose than ClO₂. Ozonation significantly enhanced the removal of many APIs including carbamazepine, metoprolol, flutamid, bupropion and beclomethasone (Fig. 2). In addition, ozonation allows removal of ibuprofen in low COD wastewater but at higher oxidant dose.

This study illustrates that treatment of wastewater containing trace pharmaceuticals is possible with either chlorine dioxide or ozone as additional treatment step depending on the target pollutant and taking into consideration the economic aspect of the process. For small-scale WWTPs, ClO₂ treatment could be an option when ozonation is too expensive and complicated to operate.

Fig. 1. Comparison of API removal by ClO₂ in low (left bar graph) and high COD (right bar graph) effluents.

Fig. 2. Comparison of API removal by O₃ in low (left bar graph) and high COD (right bar graph) effluents.