ELSEVIER

Contents lists available at SciVerse ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Chemical Engineering Journal

Ozonation-based advanced oxidation for pre-treatment of water with residuals of anti-inflammatory medication



Asu Ziylan, Nilsun H. Ince*

Bogaziçi University, Institute of Environmental Sciences, Bebek 34342, Istanbul, Turkey

HIGHLIGHTS

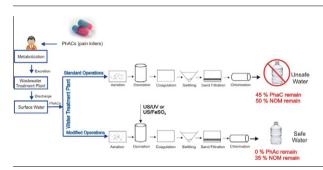
- ► Enhancement of DCF oxidation by synergy of O₃, US, UV and Fe salts.
- ▶ Pretreatment of water by O₃/US, O₃/ UV, O₃/US/UV or O₃/US/Fe²⁺ provides DCF-free water.
- ➤ Coagulation of pretreated water provides 65% less DOC.
- ► Integration of WTP-O₃ unit with US/ Fe eliminates chemical addition into coagulation tank.

ARTICLE INFO

Article history: Received 21 September 2012 Received in revised form 18 December 2012 Accepted 26 December 2012 Available online 11 January 2013

Keywords:
Pharmaceuticals
Diclofenac-Na
AOPs
WTP
NOM
Ozonation
Sonication
Hydroxyl radicals
Coagulation

G R A P H I C A L A B S T R A C T



ABSTRACT

The study is about pre-treatment of water by O₃-based AOPs as O₃/UV, O₃/ultrasound (O₃/US), O₃/H₂O₂, O₃/UV/US and O₃/US/FeSO₄ to remove residuals of anti-inflammatory pharmaceuticals and to propose a simple modification to an existing drinking water treatment plant (WTP) containing a pre-ozonation unit. Experiments were run in ultrapure and raw water (collected from the aeration tank of the WTP) spiked with 30 µM Diclofenac-Na (DCF)-the model compound to represent anti-inflammatory medication. The results showed that the most effective test processes were O₃/US/UV and O₃/US/Fe²⁺ (at 2-8 mg L⁻¹ O₃, 861 kHz US, 254 nm UV irradiation) that were significantly more effective than ozonation alone or other combinations. The outcome was attributed to the synergy of excess mass transfer and OH radical formation rates, and the presence of additional reaction routes and species. As such, 10-min pre-treatment of pure water by O₃/UV/US and O₃/US/Fe²⁺ provided nearly 90% DCF conversion; 26% and 46% mineralization, respectively. The efficiency of the processes for conversion in raw water was slightly lower, but that of mineralization was appreciably higher (55% and 58%, respectively) to be attributed to the synergy of combinations causing the interaction of excited NOM fragments with the intermediate products of DCF, followed by oxidative degradation of all to yield CO₂. Coagulation/flocculation of the pretreated streams (of raw water) with alum and without chemicals respectively provided DCF-free water and about 65% DOC mineralization. Hence, integration of the existing water treatment facility with either of the above processes is an excellent option to destroy anti-inflammatory pharmaceutical residues such as DCF and to provide appreciable DOC elimination, thus reducing the likelihood of THM formation in the distribution system. An additional benefit offered by the second process using a ferrous salt was that it allowed for a chemical-free coagulation basin.

© 2013 Elsevier B.V. All rights reserved.

1. Introduction

The presence of pharmaceutically active compounds (PhACs) in surface water is due to rapid excretion of the parent compounds and metabolites with urine (to domestic sewage), their low degree

^{*} Corresponding author. Tel.: +90 212 3597038; fax: +90 212 2575033. E-mail address: ince@boun.edu.tr (N.H. Ince).