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Can CabECO $[®]$ technology be used for the disinfection of highly faecal-</sup> polluted surface water?

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HIGHLIGHTS

CabECO® cell can be used for direct disinfection of fecal-polluted water.

- The higher the electric charge passed, the higher is the pathogens removal.
- Electrolysis at proper intensities for disinfection is not efficient for TOC removal.
- Ozone, hypochlorite and chloramines are involved in the water disinfection.
- Chlorates and perchlorates formation depends on current density.

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ABSTRACT

In this work, the disinfection of highly faecal-polluted surface water was studied using a new electrochemical cell (CabECO® cell, manufactured by CONDIAS) specifically designed to produce ozone in water with very low conductivity. The disinfection tests were carried out in a discontinuous mode to evaluate the influence of the electrode current charge passed. The effect of the current density was also studied in order to optimize the disinfection conditions and to simultaneously prevent the formation of undesirable by-products (chlorates and perchlorates) during the electrolysis. The results demonstrate that this technology is robust and efficient, and it can suitably disinfect water. During electrolysis, the chloride contained in the water was oxidized to hypochlorite, and this compound was combined with ammonia to form chloramines. Both hypochlorite and chloramines (formed by the well-known break point reaction) promoted persistent disinfection and seemed to be mainly responsible for the disinfection attained during the electrochemical process. Chlorate and perchlorate could also be produced, although the low concentrations of chloride in the tested water made them irrelevant. The removal of the total organic carbon under the applied operating conditions was not very efficient (although it reached 50% in 2 h) and the production of trihalomethanes was very low, below 100 ppb for all tests.

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1. Introduction

The loss of water quality associated with pollution caused by human activities related to the development of modern society (e.g., industries and farms), as well as abruptly changing rain patterns and more extreme drought periods in many regions due to climate change are contributing to water scarcity in developing

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<https://doi.org/10.1016/j.chemosphere.2018.06.106> 0045-6535/© 2018 Elsevier Ltd. All rights reserved. areas and poor countries. Thus, the search for high-quality reservoirs of water and/or the development of technologies that can improve the quality of water is a topic of major interest.

Currently, surface water and groundwater are the two major sources of drinking water, and in some countries, reclaimed water has also become a new source of drinking water. However, in many other countries, reclaimed water is still controversial and is banned by regulations for the production of drinking water, although it is allowed for other applications, such as irrigation ([Rodrigo et al.,](#page--1-0) [2010\)](#page--1-0).

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E-mail address: cristina saez@uclm es (C Sáez) **Expanding from lakes, artificial reservoirs, rivers Surface water, originating from lakes, artificial reservoirs, rivers**

or wetlands, naturally contains sediments and organic species, such as humic and fulvic acids. The concentrations of each of these pollutants strongly depend on the level of the eutrophication of the source and the hydrodynamic characteristics of the water catchment area. Consequently, surface water may be turbid. In addition, in rural areas, farming may cause faecal pollution of the water sources. In several countries farming is seriously affecting the quality of fresh water reservoirs used for human supply, thus necessitating the oversizing of the typical technologies used to obtain the drinking water or even designing new types of treatment technologies capable of addressing faecal pollution.

Generally, the treatment of surface water is targeted to remove turbidity (with all associated pollutants) and kill microorganisms, and it typically consists of the following sequence: coagulation, flocculation, sedimentation, filtration, adsorption, then chemical disinfection. Chemical disinfection technologies, such as chlorination or ozonation, present different drawbacks that limit their application. The formation of disinfection by-products and the loss of disinfection efficiency through the presence of organic matter are crucial when considering chemical disinfection technologies ([Arapoglou et al., 2003;](#page--1-0) [Kinani et al., 2016](#page--1-0); [Pan et al., 2016](#page--1-0); [Li et al.,](#page--1-0) [2017b](#page--1-0)). Physical disinfection processes, such as UV irradiation, membrane separation and thermal disinfection, are associated with high costs and maintenance efforts and do not fulfil the requirements for primary and residual water disinfection. Consequently, disinfection using electrochemical technology appears to be an environmentally friendly, economical and operationally competitive technology for use against a wide range of microbiological contaminations in water ([Kolosov et al., 2001;](#page--1-0) [Oturan et al.,](#page--1-0) [2001;](#page--1-0) [Louhichi et al., 2008](#page--1-0); [Ma et al., 2010;](#page--1-0) [Piuleac et al., 2012\)](#page--1-0). The direct electrolysis of the water to be disinfected without the further addition of chemicals is based on the production of oxidants from the oxidation of anions directly contained in the raw water or wastewater, such as chloride, sulphate, phosphate or carbonate ([Canizares et al., 2009](#page--1-0)). These powerful oxidants can attack microorganisms and help to remove them from a solution [\(Martinez-](#page--1-0)[Huitle and Brillas, 2008](#page--1-0)). A critical factor in these electrochemical processes is the role of the anode material ([Labiadh et al., 2016\)](#page--1-0). Among others, boron doped diamond (BDD) has shown remarkable properties as an anodic material ([Pulgarin and Kiwi, 1996](#page--1-0); [Vlyssides](#page--1-0) [et al., 2004;](#page--1-0) [Ribeiro et al., 2005;](#page--1-0) [Yuan et al., 2006](#page--1-0); [Skoumal et al.,](#page--1-0) [2008\)](#page--1-0). In addition to the anodic production of the oxidants from the anions contained in the water ([Velazquez-Pena et al., 2013](#page--1-0)), the cathodic production of hydrogen peroxide by the reduction of the oxygen produced anodically (which typically saturates water during electrolysis) ([Valero et al., 2017\)](#page--1-0) and the formation of other oxidants, such as ozone [\(Heim et al., 2015](#page--1-0); [Rajab et al., 2015](#page--1-0)), must be taken into account because they can contribute to the disinfection activity. Unfortunately, electrochemical disinfection using BDD electrodes may lead to the formation of undesirable disinfection by-products, such as the highly oxidized chloro-species chlorate and perchlorate ([Bergmann et al., 2009](#page--1-0); [Schaefer et al., 2015\)](#page--1-0), depending on the sp^3/sp^2 ratio of the BDD electrode [\(Brito et al.,](#page--1-0) [2015\)](#page--1-0). The presence of these chloro-species should be prevented, as they have a possible carcinogenic effect on human health. Moreover, the reaction of halogen species with organic molecules found in water will promote the formation of harmful adsorbable organically bound halogens (AOX) and trihalomethanes (THMs). There are many electrochemical technologies, and one of the most promising to avoid problems with by-products is the CabECO[®] process (manufactured by CONDIAS) [\(Fryda et al., 2016\)](#page--1-0). The CabECO® technology consists of a special cell design, made of diamond anodes, which has been optimized for the production of ozone in water with low conductivity. Because of this special feature, this technology has been proven to be efficient in indirect

disinfection by dosing ozone to water.

The goal of this work was to establish the efficiency of the CabECO® technology equipped with diamond electrodes in the direct disinfection of surface water containing faecal pollution. The disinfection of the surface water sample tested in this work was evaluated by the quantification of two indicators of pathogenicity: total coliforms and Pseudomonas aeruginosa. The indicators were selected taking into account that: 1) they are the most common microbial contaminants in natural waters and wastewaters ([Li et al.,](#page--1-0) [2017a\)](#page--1-0), and 2) the versatile metabolism of Pseudomonas aeruginosa and its increasing resistance to antibiotics ([Bruguera-Casamada](#page--1-0) [et al., 2017\)](#page--1-0). In addition, total aerobic microorganisms were also monitored in order to determine the effect of the technology not only on disinfection but also on sterilization. The effects of the current density and the operation mode on both the disinfection efficiency and the formation of undesirable by-products were also evaluated.

2. Materials and methods

2.1. Chemicals and raw water

To simulate surface water with faecal pollution, water collected at the inlet of the municipal Water Treatment Plant (WTP) of Ciudad Real (which received surface water pumped directly from the Gasset Reservoir) was merged with the effluent of the secondary clarifier of the municipal Wastewater Treatment Plant (WWTP). The volumetric ratio used for the mixture was 95/5 (volume surface water/volume secondary treated wastewater).

2.2. Analytical techniques

A complete physico-chemical characterization was carried out. The concentration of the ions was measured by ion chromatography using a Metrohm 930 Compact IC Flex coupled to a conductivity detector. A Metrosep A Supp 7 column was used to determine the anions using a mobile phase consisting of 85:15 v/v 3.6 mM Na₂CO₃/acetone at a flow rate of 0.8 cm³ min $^{-1}$. In addition, a Metrosep A Supp 4 column was used to analyse cations, with a mobile phase consisting of 1.7 mM HNO₃ and 1.7 mM 2,6pyridinedicarboxylic acid at a flow rate of $0.9 \text{ cm}^3 \text{ min}^{-1}$. The temperature of the oven was 45 and 30 $\,^{\circ}$ C for the determination of anions and cations, respectively, and the injection volume was 20μ L. The Total Organic Carbon (TOC) concentration was monitored using a Multi N/C 3100 Analytik Jena analyser. Hypochlorite was analysed by titration with 0.001 M $As₂O₃$ in 2 M NaOH. Oxidants were determined iodometrically [\(Canizares et al., 2009\)](#page--1-0). Inorganic chloramines were determined following the DPD standard method described in the literature [\(APHA-AWWA-WPCF,](#page--1-0) [1998\)](#page--1-0). The pH and conductivity were measured using a CRISON $pH25$ + and CRISON CM35+, respectively. The organo-chlorinated intermediates were analysed by GC-MS using a Thermo Scientific DSQ II Series Single Quadrupole GC-MS with a NIST05-MS library. The column was a polar TR-WAXMS (30 m \times 0.25 mm x 0.25 µm). The temperature profile was 70 °C for 1 min, with a ramp of 30 °C min^{-1} to 300 °C and a hold time of 5 min. The inlet, source and transfer line temperatures were 250, 200 and 300 \degree C, respectively.

For the microbiological characterization, standard microbiological tests conforming to the following methods were used: UNE-EN ISO 6222:1999 for the quantification of total aerobic microorganisms, UNE-EN ISO 11133:2014 for the quantification of total coliforms, and UNE-EN ISO 16266:2008 for the quantification of Pseudomonas aeruginosa. Further information is given in the supplemental material section.

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