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New chlorinated amphetamine-type-stimulants disinfection-by-products formed during drinking water treatment

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ABSTRACT

Previous studies have demonstrated high removal rates of amphetamine-type-stimulants (ATs) through conventional drinking water treatments; however the behaviour of these compounds through disinfection steps and their transformation into disinfection-by-products (DBPs) is still unknown. In this work, for the first time, the reactivity of some ATs such as amphetamine, methamphetamine, 3,4-methylenedioxyamphetamine (MDA), 3,4-methylenedioxymethamphetamine (MDMA) and 3,4-methylenedioxyethylamphetamine (MDEA) with chlorine has been investigated under simulated and real drinking water treatment conditions in order to evaluate their ability to give rise to transformation products. Two new DBPs from these illicit drugs have been found. A common chlorinated-by-product (3-chlorobenzo)-1,3-dioxole, was identified for both MDA and MDEA while for MDMA, 3-chlorocatechol was found. The presence of these DBPs in water samples collected through drinking water treatment was studied in order to evaluate their formation under real conditions. Both compounds were generated through treatment from raw river water samples containing ATs at concentration levels ranging from 1 to 15 ng/L for MDA and from 2.3 to 78 ng/L for MDMA. One of them, (3-chlorobenzo)-1,3-dioxole, found after the first chlorination step, was eliminated after ozone and GAC treatment while the MDMA DBP mainly generated after the postchlorination step, showed to be recalcitrant and it was found in final treated waters at concentrations ranging from 0.5 to 5.8 ng/L.

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1. Introduction

The presence of human-use compounds in aquatic environments has been recognized as an important issue in environmental chemistry. In this context, illicit drugs have been identified in the last years as a new group of water contaminants. To date, these compounds have been found in several

water matrices around the world (rev. in: (Zuccato and Castiglioni, 2009; Castiglioni et al., 2011)) and the concern regarding their presence in these resources is growing due to their potent psychoactive properties and their unknown effects to the aquatic environment. These compounds enter directly the water system through sewage waters and their non-quantitative elimination in wastewater treatment plants

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(WWTPs) has a direct effect in surface waters. The presence of illicit drugs in raw waters which can be used for drinking water production may have a negative impact in the quality of the final tap water. Compounds such as cocaine, amphetamine, ecstasy, or cannabinoids have been found at concentrations in the nanogram per litre range in wastewater treatment plant (WWTP) effluents in United States, Italy, Germany, Spain, Ireland, United Kingdom, and Belgium (Jones-Lepp et al., 2004; Castiglioni et al., 2006; Hummel et al., 2006; Huerta-Fontela et al., 2007, 2008a; Bones et al., 2007; Boleda et al., 2007; Kasprzyk-Hordern et al., 2008; Gheorghe et al., 2008; van Nuijs et al., 2008; Zuccato and Castiglioni, 2009; Petrovic et al., 2009; Postigo et al., 2010). Recent published works (Huerta-Fontela et al., 2008b; Boleda et al., 2009) have demonstrated that conventional drinking water treatments are able to remove most of the illicit drugs or metabolites detected in raw waters. For instance, amphetamine-type-stimulants (except ecstasy) were removed during prechlorination steps while granulated activated carbon (GAC) filtration allowed removing cocaine (>99%) and benzoylcegonine (72%) and postchlorination achieved the elimination of ecstasy. Despite the fact that some of them are mainly removed, their presence during these treatment steps can potentially cause the generation of new disinfection-by-products (DBPs).

Chlorine is currently used for the disinfection of drinking water all over the world and despite its benefits one well-known drawback is the formation of DBPs (Rook, 1974; Richardson et al., 2007). Recently the formation of chlorinated DBPs from several emerging contaminants (Richardson, 2007; Huerta-Fontela and Ventura, 2008) such as alkylphenol ethoxylate surfactants (Petrovic et al., 2003), ethinylestradiol (Alum et al., 2004), acetaminophen (Bedner and MacCrehan, 2006), antibacterial agents (Rule et al., 2005), bisphenol A (Wu et al., 2009) or X-ray contrast media (Duirk et al., 2011) among others has been demonstrated.

Due to the specific structures of illicit drugs and their presence in raw waters subjected to chlorination treatments, some of these compounds can be considered as candidates to generate new DBPs. Specifically, amphetamine-type-stimulants (ATs): amphetamine, methamphetamine (METH), 3,4-methylenedioxymphetamine (MDA), 3,4-methylenedioxymethamphetamine (MDMA) and 3,4-methylenedioxymethamphetamine (MDEA), are an interesting group since chlorine reacts easily with primary and secondary amines by electrophilic substitution (Deborde and von Gunten, 2008). As suggested by Abia et al. (1998) the reaction occurs between the amine moiety and the HOCl molecule leading to the formation of chloramines and other small modifications in their structure susceptible to generate toxic haloamine DBPs.

The aim of the present study was to assess the formation of DBPs from ATs during chlorination treatment. The reactivity of these compounds was studied and chlorination products were characterized by using liquid chromatography (LC) coupled to mass spectrometry (MS). The unequivocal structural confirmation of the identified DBPs was performed by using commercial or synthesized standards. Finally, the presence of the new DBPs along the potabilization process in a drinking water treatment plant (DWTP) was investigated.

2. Experimental section

2.1. Materials and methods

Amphetamine, METH, MDA, MDMA, and MDEA, amphetamine-d8, methamphetamine-d9, MDA-d5, MDMA-d5, MDEA-d5 at a concentration of 1 mg/L, (99%) were purchased from Cerilliant (Austin, TX, USA). 3-chlorobenzoic acid and 4-chlorobenzoic acid, (3-chlorobenzo)-1,3-dioxole, (4-chlorobenzo)-1,3-dioxole, 3-chlorocatechol and 3-chloro-2-hydroxybenzaldehyde standards were obtained from Sigma–Aldrich (USA) (>99%). In Table 1 properties of the selected compounds are shown.

Sodium hypochlorite solution (NaOCl with 10–15% (m/V) of active chlorine), sodium hydroxide (98%), hydrogen peroxide solution ($\geq 30\%$) and sodium sulphate ($\geq 99\%$) were purchased from Sigma–Aldrich (USA). Hydrochloric acid ($\geq 99\%$) was obtained from Carlo Erba (Italy) and dichloromethane from Merck (USA).

Acetonitrile LC-MS grade and water obtained from Merck (USA) were used as LC mobile phases and methanol Purge and Trap grade was obtained from Riedel-de-Haën (Germany).

Water used for experiments was purified (18.2 M Ω cm, 25 °C and TOC < 5 μ g/L) in an Elix-Milli-Q system (Millipore Corp., USA)

2.2. Water samples

Water samples were collected from a DWTP located in NE-Spain from April to June 2009. In this facility, chlorination is performed twice throughout the treatment. First, chlorine is added to the raw river water until break-point is achieved in order to completely remove the natural ammonia present in these raw waters. Next, after successive treatments including flocculation, coagulation, ozonization and GAC filtration, postchlorination is performed to maintain a chlorine residual concentration of about 0.8–1.2 mg/L through the distribution system (Fig. 1). The torrential regime of this Mediterranean river leads to significant temporal variation in the quality of raw water at the intake of the DWTP as shown in Table 2.

Raw water and treated water samples were collected after each treatment step by step taking into account the hydraulic retention times (HRT). They were stored in amber glass bottles below 4 °C and extracted within 48 h of collection. Surface raw waters were filtered through glass microfiber GF/A filters (Whatman, UK). Sodium thiosulphate to quench chlorine can convert N-chloro compounds back into their original form (Pinkston and Sedlak, 2004) and therefore ascorbic acid was added to treated waters in order to prevent further degradation.

In order to detect the parent compounds in raw waters, a highly sensitive SPE LC-MS/MS method, described elsewhere (Huerta-Fontela et al., 2007), was used. The DBPs were only traced for those intake samples showing the presence of the related parent compounds. Additionally, samples were collected after each treatment step in order to evaluate the DBPs formation, their evolution through the treatment and the influence of other oxidative and non-oxidative steps.

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