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# Seasonal variations in the concentration and removal of nonylphenol ethoxylates from the wastewater of a sewage treatment plant

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Introduction

#### ABSTRACT

In this study, we investigated the occurrence and fate of nonylphenol (NP), nonylphenol 16 monoethoxylate (NP1EO) and nonylphenol diethoxylate (NP2EO) in a full scale sewage 17 treatment plant, which applied an Anaerobic/Oxic process. Concentrations of NP, NP1EO 18 and NP2EO in the wastewater were measured during the period spanning a whole year. 19 The results showed remarkable seasonal variation in the concentrations of the compounds. 20 The NPnEO compounds were most abundant in winter, with the total concentrations of 21 influent NP, NP1EO and NP2EO ranging from 3900 to 7000 ng/L, 4000 to 4800 ng/L and 5200 22 to 7200 ng/L, respectively. Regarding the total removal efficiencies of the three types of 23 short-chain NPnEO compounds, different trends were exhibited according to different 24 seasons. The average removal efficiency of NP for the different seasons ranked as follows: 25 winter > summer > autumn > spring; NP2EO concentrations decreased as follows: summer > 26 autumn > winter > spring, while NP1EO concentrations reduced according to: spring > 27 summer > autumn > winter. We also investigated the contribution ratio of individual 28 treatment units in the A/O process, with the findings suggesting that the anaerobic 29 treatment unit plays an important role in the elimination of short-chain NPnEOs from the 30 wastewater.

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Alkylphenol ethoxylates (APnEOs), where n indicates the number of ethoxy units, are a commercially important group of nonionic surfactants which have been widely employed for industrial, agricultural, and domestic applications. Annually, approximately 700,000 tons of APnEOs surfactants are manufactured worldwide (Jonkers et al., 2005). The most extensively used APnEOs products are nonylphenol ethoxylates (NPnEOs), which, account for 80%–85% of the total use (Loyo-Rosales

et al., 2010). The NPnEOs polyethoxylate chains are com- 54 prised of 1 to 20 ethoxy units, which cause the NPnEO com- 55 pounds to be unstable. In sewage treatment plants (STPs), 56 NPnEOs undergo a series of rapid transformations, where 57 the polyethoxylate chains breakup and generate short-chain 58 NPnEOs (n ranging from 0 to 2), which include nonylphenol 59 (NP), nonylphenol monoethoxylate (NP1EO) and nonylphenol 60 diethoxylate (NP2EO) (Loyo-Rosales et al., 2007; Planas et al., 61 2002). These increasing contaminants are discharged into 62 aquatic ecosystems from sewage plants (Stasinakis et al., 63

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2012). The NPnEOs metabolites exhibit higher toxicity than their precursors, and have been identified as xenoestrogens. NPnEOs have been detected at high concentrations in the atmosphere and waterways in many countries around the world (Ahel et al., 1996; Barber et al., 2009; Maruya et al., 2015; Johnson et al., 2005; Pryor et al., 2002). Consequently, the use and production of nonylphenol derivatives have been banned or restricted by many countries. According to the recommended water quality criteria for nonylphenols, reported by the U.S. EPA in 2005, the concentration of NP should be below 6.6 μg/L in freshwater, and below 1.7 μg/L in saltwater (US EPA, 2005). However, the high performance efficiency of nonylphenols makes it difficult to identify alternative substitutes, and many countries, including China, have not taken action to avoid or reduce their use (Cailleaud et al., 2007; Soares et al., 2008). However, increasing attention is being paid to short-chain NPnEOs, because of their pervasive nature and the high concentrations that have been detected in the aquatic environment (Cespedes et al., 2008).

The removal efficiency of NPnEOs from STPs has been widely reported in the literature. In Spain, researchers compared two types of treatment processes for a wastewater treatment plant (WWTP) that treated municipal, industrial and hospital wastewaters, with a capacity of 27,000 m<sup>3</sup>/day. The conventional activated sludge treatment process was found to have an average primary elimination of NP1EO and NP2EO of 50%, whilst NP was more efficiently removed (96%). Alternatively, the membrane bioreactor treatment process demonstrated superior performance in the elimination of short-chain NPnEO compounds; with 90% of NP1EO and NP2EO eliminated, and 96% of NP (Gonzalez et al., 2007). Researchers have also investigated the removal of a comprehensive group of NPnEOs and their metabolites from WWTPs. The results demonstrated efficient elimination percentages, ranging from 75.7% to 90.8%.

The previous studies have indicated that approximately 80% of the compounds sourced from river water samples were present in the dissolved phase (Isobe et al., 2001). Consequently, the removal of NPnEOs is dependent on water temperature and it is widely accepted that NPnEO compounds demonstrate a resistance to biodegradation at low temperatures (Loyo-Rosales et al., 2007). Further studies have indicated that higher temperatures may accelerate the microbial transformation of long-chain NPnEOs, and that the de-ethoxylation rates of long-chain NPnEOs are more temperature sensitive than the short-chain NPnEOs (Lian et al., 2009).

To date, the concentrations and removal efficiencies of NPnEOs in STPs have been substantially investigated. However, their behavior and their fate in relation to seasonal variations, especially in tertiary (advanced) treatment processes, are less well known. The study focuses on the occurrence of short-chain NPnEOs in a STP in Northern China. The total removal efficiencies of NP, NP1EO and NP2EO in the dissolved phase and the contribution ratios of the individual treated units in the A/O process are measured. To evaluate the seasonal variation of short-chain NPnEOs, samples were collected from May, 2009 to April, 2010. The results of this study will contribute to improving the removal of short-chain NPnEOs in cold areas, in order to improve the efficiency of STPs in China.

#### 1. Materials and methods

#### 1.1. Standards and regents

A standard of 4-tert-nonylphenol (purity > 95%) was pur- 127 chased from sigma-Aldrich (Taufkirchen, Germany). Techni- 128 cal nonylphenol mono- and di-ethoxylates were obtained from 129 Dr. Ehrenstorfer (Augsburg, Germany). The standards were 130 further diluted with iso-octane to prepare seven standardized 131 mixtures. High purity pesticide analytical grade solvents, 132 including acetone, methanol, ethyl acetate and iso-octane, 133 were purchased from J.T. Baker Co., USA. Carbon-free deionized 134 water (DI water) was obtained from a NANO pure system 135 (Bamstead International, Dubuque, IA).

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#### 1.2. Sewage treatment plant

The sewage treatment plant which wastewater mainly comes 138 from domestic sewage is located in Harbin, the capital and 139 largest city in the northeast of China. In Harbin, the lowest 140 temperature was between –13.2 to –24.8°C in winter, and the 141 highest temperature was between 20.2 to 28.8°C in summer. 142 The STP configured such that after the primary settling tank 143 wastewater passes through an Anaerobic/Oxic (A/O) process. 144 The A/O process consists of six corridors, two of which are 145 running under anoxic conditions while the other four are under 146 oxic conditions. The hydraulic retention time (HRT) is 10 hr, and 147 the sludge retention time (SRT) is 19 days in bioreactors. The 148 process flow of the A/O system is illustrated in Fig. 1.

#### 1.3. Sampling procedures

Grab wastewater samples were collected from all stages of 151 the treatment process. All samples were collected by a vessel 152 pre-cleaned with acetone, and then immediately stored at 153 4°C. The extraction was carried out within 24 hr once the 154 samples collected. To prevent biological degradation, formal- 155 dehyde (1 vol.%) was added to the samples (Gao et al., 2014). 156

#### 1.4. Extraction and analysis

Caliper solid-phase extraction, coupled with Waters C18 158 cartridges were used to concentrate the water samples. The 159 pretreatment and analysis of samples were conducted ac- 160 cording to the reference (Gao et al., 2014).

#### 1.5. Quality assurance/quality control

Blank samples consisting of ultrapure water, and spiked 163 samples which contained a different sample in each batch 164 were processed along with every seven sample extraction 165 round. Relative recovery values were then calculated from 166 these samples. The recovery of blank samples ranged from 167 65% to 106% (mean  $83\% \pm 10\%$ ). Recovery values for all the 168 NPnEO compounds ranged from 90% to 135%, with a mean 169 value of  $104\% \pm 7\%$ . Blank contamination was also assessed 170 from two field blanks and two laboratory blanks, and all of 171 the NPnEO congeners in the blank samples were below the 172 detection limit (Gao et al., 2014).

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