



Occurrence and profiling of multiple nitrosamines in source water and drinking water of China



Wanfeng Wang^{a,b}, Jianwei Yu^{a,*}, Wei An^a, Min Yang^a

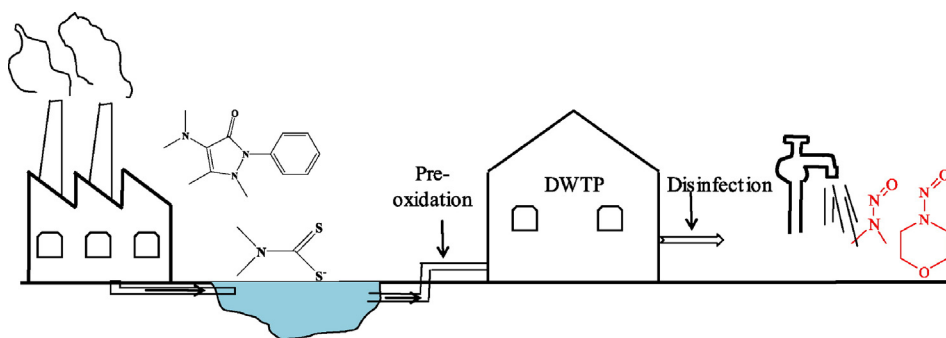
^a Key Laboratory of Drinking Water Science and Technology, State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

^b Key Laboratory for Yellow River and Huai River Water Environmental and Pollution Control, Ministry of Education, Henan Key Laboratory for Environmental Pollution Control, School of Environment, Henan Normal University, Xinxiang 453007, China

HIGHLIGHTS

- Studied the occurrence of multiple nitrosamines in 54 DWTPs across major cities of China.
- NDMA, NDEA and NDBA were the most abundant in DWTPs, and the detection frequencies in source waters were even higher than those in finished waters.
- The three main nitrosamines posed a cancer risk of 2.99×10^{-5} to the local populations due to their occurrence in drinking water.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 20 November 2015
 Received in revised form 26 January 2016
 Accepted 26 January 2016
 Available online xxxx

Editor: Adrian Covaci

Keywords:

Nitrosamines
 Drinking water
 Chlorination
 Chloramination
 Formation potential
 Cancer risk

ABSTRACT

The occurrence of multiple nitrosamines was investigated in 54 drinking water treatment plants (DWTPs) from 30 cities across major watersheds of China, and the formation potential (FP) and cancer risk of the dominant nitrosamines were studied for profiling purposes. The results showed that N-nitrosodimethylamine (NDMA), N-nitrosodiethylamine (NDEA) and N-nitrosodi-n-butylamine (NDBA) were the most abundant in DWTPs, and the concentrations in source water and finished water samples were not detected (ND) – 53.6 ng/L (NDMA), ND – 68.5 ng/L (NDEA), ND – 48.2 ng/L (NDBA). The frequencies of detection in source waters were 64.8%, 61.1% and 51.8%, and 57.4%, 53.7%, and 37% for finished waters, respectively. Further study indicated that the FPs of the three main nitrosamines during chloramination were higher than those during chlorination and in drinking water. The results of Principal Components Analysis (PCA) showed that ammonia was the most closely associated factor in nitrosamine formation in the investigated source water; however, there was no significant correlation between nitrosamine-FPs and the values of dominant water-quality parameters. The advanced treatment units (i.e., ozonation and biological activated carbon) used in DWTPs were able to control the nitrosamine-FPs effectively after disinfection. The target pollutants posed median and maximum cancer risks of 2.99×10^{-5} and 35.5×10^{-5} to the local populations due to their occurrence in drinking water.

© 2016 Elsevier B.V. All rights reserved.

1. Introduction

Considerable attention had been focused on the occurrence of nitrosamines, a class of widespread disinfection by-products (DBPs)

* Corresponding author.
 E-mail address: jwyu@rcees.ac.cn (J. Yu).

and industrial by-products, because of their suspected carcinogenicity and mutagenicity, as well as the risk of stomach, esophageal, and nasopharyngeal cancers (Eichholzer and Gutzwiller, 1998). In particular, N-nitrosodimethylamine (NDMA) has been frequently detected in water resources in many countries, which could be mainly attributed to the use of chlorine, chloramine or ozone for the disinfection of drinking water or wastewater (Mitch et al., 2003; Sedlak et al., 2005; Pehlivanoglu-Mantas et al., 2006; Andrzejewski et al., 2008). It is estimated that concentrations of NDMA as low as 0.7 ng/L in drinking water would be associated with a 10^{-6} lifetime cancer risk; the corresponding concentration is 0.2 ng/L for N-nitrosodiethylamine (NDEA) (US EPA, 2007). The International Agency for Research on Cancer (IARC) has classified NDMA and NDEA as probable carcinogens to humans (Group 2A), and six other nitrosamines including N-nitrosomethylethylamine (NMEA), N-nitrosopyrrolidine (NPyr), N-nitrosomorpholine (NMor), N-nitrosopiperidine (NPip), N-nitrosodipropylamine (NDPA), and N-nitrosodi-n-butylamine (NDBA) are classified as possible carcinogens to humans (Group 2B) (IARC, 1978).

As a group of emerging DBPs, nitrosamines have been detected in water treatment processes in many countries. NDMA has been widely investigated in drinking water in Canada (Zhao et al., 2008), the United States (Mitch et al., 2003), and Japan (Asami et al., 2009). The concentrations of NDEA have been reported in only several countries and regions, in drinking water (3.7–12.9 ng/L) (Jurado-Sanchez et al., 2010; Planas et al., 2008) and domestic wastewater (0.132 µg/L) (Hartmetz and Slemrova, 1980). Although other nitrosamines including NMEA, NDPA, NMor, NPip, NPyr, NDBA, and N-nitrosodiphenylamine (NDPhA) have been investigated in several drinking water treatment plants (DWTPs) (Wang et al., 2011) and a digestive system cancer region (Ma et al., 2012), due to the lack of a detailed national survey, information regarding the occurrence and concentration levels of multiple nitrosamines in different treatment types of DWTPs across major watersheds in China is still very limited. On the other hand, several studies have investigated the effects of different disinfection processes on nitrosamine formation. However, DBP formation is a complicated process that is influenced by many variables including the pH, temperature, turbidity, natural organic matter (NOM) content and concentration in source water, the different types of pre-oxidation and filtrations, as well as the type and concentration of the disinfectants used and disinfection contact time. Some of the previous studies have evaluated the formation of NDMA during ozonation and pre-oxidation (Andrzejewski et al., 2008; Schmidt and Brauch, 2008). Pre-oxidation processes such as pre-chlorination, pre-chloramination and pre-ozonation are widely used in drinking water treatment (Susan et al., 2003; Rodgers, 1997; Camel and Bermond, 1998), leading to the formation of nitrosamines before filtration and disinfection processes. However, few previous reports have shown the relationship between the migration, transformation and control techniques of the multiple nitrosamines and the different types of treatment processes in DWTPs.

With the development of the economy and society, the water environment is becoming more and more complicated in China. The occurrence and fate of toxic compounds in China's DWTPs is necessary for study of their corresponding control techniques. Therefore, in the current study, a total of 54 source water and finished water samples from 30 cities across China were collected for a survey of multiple nitrosamines, and the dominant nitrosamine-FPs were analyzed during chlorination and chloramination. In addition, we made an attempt to estimate the cancer risk of the dominant nitrosamines in potable water for the local populations.

2. Materials and methods

2.1. Sample collection

The study areas and sampling sites are shown in Fig. 1. Source water and finished water samples were collected from 54 full-scale DWTPs of

30 cities in China between March and May 2010 (Spring), including the Songhua River basin, the Liao River basin, the Yellow River basin, the Yangtze River basin, and the Pearl River basin (Fig. 1). There were 24 source waters coming from rivers, 23 source waters were from reservoirs, and 7 source waters derived from groundwater. There was no rainfall during the sampling periods. The samples were collected in amber glass bottles and sodium thiosulfate (20 mg/L) was added to quench any chlorine residue. Then the samples were delivered under refrigerated conditions (4 °C in cooling boxes) within 48 h to the lab. Once they arrived in the lab, the samples were stored in the dark at 4 °C until they were analyzed. Information regarding the source water characteristics, disinfectants, and water treatment processes is shown in Supplementary Table A.

2.2. Sample preparation for analyzing nitrosamines

After filtration with a glass microfiber filter (GF/C, 1.2 µm; Whatman, Maidstone, UK), 0.5 L of water was adjusted to pH 8.0 with about 1.0 g of solid sodium bicarbonate and spiked with 20 ng/L internal standard (NDMA-d₆). The samples were extracted using Resprep EPA 521 cartridges (2 g/6 mL, Restek, Bellefonte, PA, USA) which had been preconditioned with 10 mL of hexane, 20 mL of dichloromethane, 20 mL of methanol, and 20 mL of water at a flow rate of 3–5 mL/min. Next, the cartridges were dried under a flow of nitrogen gas. After 20 mL of dichloromethane was used to elute each cartridge, 400 µL of a water/methanol solution (95:5, v/v) was added to the elutions, and the dichloromethane was completely removed using a rotary evaporator (LabTech, USA). Then the sample volume was gravimetrically adjusted to 0.5 mL using ultrapure water. To remove possible solid particles, all samples were filtered through syringe filters (GHP Acrodisc 13 mm, 0.2 µm, PALL) prior to injection into the UPLC–MS/MS system.

2.3. Nitrosamine formation potential test for source waters

To investigate the potential risks in drinking water, we studied the main nitrosamine-FPs of 41 source water samples in chlorination and chloramination. The Cl₂-demand (Cl₂-D) and NH₂Cl-demand (NH₂Cl-D) were measured according to our previous study (Wang et al., 2011). The samples were chlorinated and chloraminated in 500 mL amberized vials by keeping a free chlorine or chloramine residual of 1.0 mg/L after storage at (25 ± 1) °C for 24 h. Analyses of nitrosamine were then conducted after the residual chlorine and chloramine were quenched. The Cl₂-D and NH₂Cl-D for 41 source waters are listed in Supplementary Table B.

2.4. UPLC–ESI-MS/MS analysis for nitrosamines

Nitrosamine analysis followed our previous method (Wang et al., 2011). Briefly, the LC apparatus was an Acquity Ultra performance LC (Waters, USA). All analytes were separated using a Waters Acquity ultra-performance liquid chromatography (UPLC) BEH C₁₈ column (150 mm × 2.1 mm, 1.7 µm particle size; Waters, USA). Methanol (A) and ultrapure water containing 10 mmol/L ammonium bicarbonate (B) were used as mobile phases. Mass spectrometry was performed using a Premier XE tandem quadrupole mass spectrometer (Waters) equipped with a Z-Spray ionization (ESI) source. ESI-MS/MS detections were performed in the positive ion mode, and quantitative analysis was performed in the multiple reaction monitoring (MRM) mode. Recoveries and method detection limits (MDLs) of nine nitrosamines in various types of aqueous matrices are shown in Supplementary Table C.

2.5. Statistical analysis

Principal component analysis (PCA) between source water quality variables and the sum FPs of the three main nitrosamines were performed. The water quality parameters of 41 source waters included

Download English Version:

<https://daneshyari.com/en/article/6323217>

Download Persian Version:

<https://daneshyari.com/article/6323217>

[Daneshyari.com](https://daneshyari.com)