



Occurrence of nitrosamines and their precursors in drinking water systems around mainland China



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ABSTRACT

N-Nitrosamines (NAs) in drinking water have attracted considerable attention in recent years due to their high carcinogenicity, frequent occurrence, and their potential regulation. During the past three years, we have collected about 164 water samples of finished water, tap water, and source water from 23 provinces, 44 cities from large cities to small towns, and 155 sampling points all over China. The occurrence of NAs in the finished and tap water was much higher in China than that in the U.S. Nine NAs were measured and NDMA had the highest concentration. The occurrence of NDMA was in 33% of the finished waters of water treatment plants and in 41% of the tap waters. The average NDMA concentration in finished and tap waters was 11 and 13 ng/L, respectively. Formation potentials (FPs) of source waters were examined with an average NDMA FP of 66 ng/L. Large variations in NA occurrence were observed geographically in China and temporally in different seasons. The Yangtze River Delta area, one sub-area in East China, had the highest concentrations of NAs, where the average NDMA concentrations in the finished and tap water were 27 and 28.5 ng/L, respectively, and the average NDMA FP in the source water was 204 ng/L. NA control may be achieved by applying breakpoint free chlorination and/or advanced treatment of ozone – granular activated carbon process to remove the NA precursors before disinfection.

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1. Introduction

N-Nitrosamines (NAs) have been given much attention in the water industry in recent years in many industrialized nations due to their high carcinogenicity, frequent occurrence in disinfected drinking waters and wastewaters, and their potential regulation (Krasner et al., 2013). Nine alkyl NAs, i.e., *N*-nitrosodimethylamine (NDMA), *N*-nitrosomethylethylamine (NMEA), *N*-nitrosodiethylamine (NDEA), *N*-nitrosodi-*n*-propylamine (NDPA), *N*-nitrosodi-*n*-butylamine (NDBA), *N*-nitrosodiphenylamine (NDPhA), *N*-nitrosopyrrolidine (NPYR), *N*-nitrosopiperidine (NPIP), and *N*-nitrosomorpholine (NMOR), have been studied (Krasner et al., 2013). Some have been identified as disinfection by-products (DBPs) during chloramination (Choi and Valentine, 2002; Schreiber and Mitch, 2006) or the reaction of ozone with dimethylamine or certain anthropogenic chemicals (Andrzejewski et al., 2008;

Schmidt and Brauch, 2008; Marti et al., 2015).

The alkyl NAs, primarily NDMA, have been frequently detected in chloraminated drinking water systems in many countries (Krasner et al., 2013). NDMA was first identified as a DBP in drinking water in Ontario, Canada in 1989 (Jobb et al., 1994). In the U.S., NDMA was first discovered as a groundwater contaminant from rocket fuel in 1998, but was subsequently shown to be a DBP in chloraminated waters (Najm and Trussell, 2001; CDHS, 2012). Recently, NDMA was shown to be a DBP all over the U.S. as part of the Unregulated Contaminant Monitoring Rule 2 (UCMR2, Russell et al., 2012). Researchers from other developed countries, including Canada, Australia, the U.K., Germany, and Japan, have also conducted NA surveys (Charrois et al., 2007; Templeton and Chen, 2010; Sacher et al., 2008; Asami et al., 2009; Krasner et al., 2013). NAs in the U.K., Germany, and Japan were typically low in occurrence and concentration. Alternatively, in Australia, NDMA was relatively high in frequency of occurrence and concentration due to the high prevalence of chloramination, wastewater recycling, and effective chloramination resulting from high source water

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ammonia concentrations.

NA regulations and guidelines in drinking water throughout different parts of the world have been moved forward by their toxicology and their occurrence at levels of health concern (Table S1). In North American studies, most have found that NDMA formation is more associated with chloramination than with chlorination (Krasner et al., 2013). In UCMR2, systems with high plant effluent NDMA (>50 ng/L) typically used chloramines as the primary rather than secondary disinfectant (Russell et al., 2012), reflecting the potential for precursor deactivation by strong pre-oxidants. NDMA formed as a result of chlorination may be attributable to high source water ammonia concentrations, as chlorination of ammonia-containing waters may result in chloramine formation (Krasner et al., 2013).

In China, there were sporadic detections of NAs in certain locations (Liang et al., 2009; Wang et al., 2010, 2011, 2012, 2015; Luo et al., 2012). An NDMA concentration of 79 ng/L was reported (Liang et al., 2009) in the distribution system of Shanghai. Other detections of NDMA were a couple of dozen ng/L. Very recently, Wang et al. (2016) reported on a national NA survey with samples collected between March and May 2010 from 54 full-scale drinking water treatment plants (WTPs) from 30 cities in China. However, the WTPs selected for the above studies were mainly large WTPs with fairly protected water sources. The small cities and rural areas with waste-impacted water sources and not well operated WTPs were not under surveillance. Nonetheless, in some recent surveys of Chinese drinking waters, NAs occurred frequently, often due to contamination from industrial and domestic wastewaters (discussed in Krasner et al., 2013; Wang et al., 2016). Moreover, compared to other countries, nitrosamines other than NDMA (e.g., NPYR, NMOR, NPIP) have been more frequently detected in Chinese treated waters. A study on Chinese source waters found detectable levels of seven nitrosamines in certain surface waters, with concentrations >5 ng/L on average for NDMA, NDEA, NMOR, NPIP, and NDBA (Ma et al., 2012).

Epidemiology studies have shown that NAs are closely associated with digestive system cancers in some regions of China with heavy NA drinking water contamination from industrial waste (Lu et al., 1986; Chen et al., 1989; Xu et al., 2008; Fan et al., 2013). One recent publication showed that there was severe NA contamination, especially NPIP and NMOR concentration in the hundreds of ng/L, in the source waters of several cities in Jiangsu province, East China (Li et al., 2015). The 10^{-6} lifetime excess cancer risk for NDMA has been reported at 0.7 ng/L (EPA IRIS, 1993). The U.S. Environmental Protection Agency tries to regulate carcinogenic compounds in their 10^{-6} – 10^{-4} risk range.

During the past three years, we collected 164 water samples from 23 provinces, in 44 cities throughout mainland China. In this paper, the occurrence results are presented and the NA distribution in the regions of mainland China is discussed. Compared with the recent publication from Wang et al. (2016), large differences in the sampling profile, NA precursor detection method, and data mining were present. First, we collected tap waters as well as the source water and finished water, whereas Wang et al. did not sample tap water. Second, we collected samples from a more diverse range of WTPs, from large cities, small cities and towns with different sizes, processes, and water sources, whereas Wang et al. collected water samples from only the major cities in China. Third, we conducted formation potential (FP) tests to determine NA precursor levels with a 7-day reaction and a high monochloramine dose, whereas Wang et al. used a short contact time (1 day) that will not allow a full determination of the level of NA precursors. Fourth, the two papers had different approaches for data mining. Wang et al. (2016) focused on correlation analyses of nitrosamines and basic water quality parameters and risk assessments, whereas our paper gives

more details on geographical occurrence, the performance of treatment processes, and a comparison between China and the U.S. However, each paper presents some information about the NA formation and occurrence. The results from each paper are very valuable and complement each other. The results from these studies will be very useful for water professionals to understand NA occurrence in mainland China, which has some of the most diverse occurrence of NAs in the world. This occurrence information is one step in solving the public health concern over NAs in drinking water.

2. Materials and methods

2.1. Collection of water samples

The NA sampling campaign was conducted continuously since 2012. We collected a total of 164 water samples of finished water, tap water, and source water, which came from 44 cities or towns, in 23 provinces throughout China. Among these samples, 43 finished water samples and 74 tap water samples were analyzed for NAs and other water quality parameters; 47 source water samples were analyzed for NA FP and other water quality parameters (Included in the Supplementary Information [Tables S8–S11]). These water samples represented seven major areas (East, South, North, Northeast, Central, Northwest, Southwest) of mainland China (Fig. S1). The source water samples included river water (Fig. S2), lake or reservoir water, and groundwater.

Briefly, the treated water samples for NAs were collected in amber glass bottles and dechlorinated with sodium thiosulfate immediately. The source water samples for NA FP and other bulk water quality parameters were first collected without addition of sodium thiosulfate (raw water samples should not have a disinfectant residual). All samples were kept in an ice box during shipment, and held in a refrigerator below 6 °C until analysis. Analysis of the water samples were accomplished within 14 days.

2.2. N-nitrosamine and FP analysis

Nine N-nitrosamines were analyzed using a modified solid-phase extraction (SPE)-LC/MS/MS procedure (Zhao et al., 2006; Wang et al., 2012). An SPE instrument (Auto Trace 280, Dionex Corp., U.S.A.) and pre-packed SPE cartridges with coconut charcoal for EPA 521 method (CNW Technologies GmbH, Shanghai, China) were used to isolate the NAs from water samples. The water sample (1 L) was concentrated down to 1 mL for LC/MS/MS analysis. A check standard was run after every 10 samples. The ultra-performance (UP) LC and MS/MS parameters are included in the Supplementary Information (Table S3). The reagents used are also included in the Supplementary Information.

The procedure for the NA FP test was similar to that used in other studies (Mitch et al., 2003; Liao et al., 2014, 2015a). An excess dosage of monochloramine (20 mg/L as Cl_2) was added into an amber bottle stored in the dark at 25 °C. The pH was adjusted to 8 with a 0.02 mol/L phosphate buffer solution. After 7 days, the reaction was quenched by addition of an excess amount of sodium thiosulfate. Then the samples were processed for N-nitrosamines analysis.

2.3. Quality assurance and quality control of N-nitrosamine analysis

A series of NA solutions were prepared with concentrations of 0, 0.5, 1, 2, 5, 10, 20, 50, and 100 ng/L each. Additionally, NDMA-d6 (20 ng/L) was added to each sample before SPE to check the recovery performance, and NDPA-d14 (20 ng/L) was added to each

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