Changes Highlighted

Removal of Radio *N*-Nitrosodimethylamine (NDMA) From Drinking Water by Coagulation and Powdered Activated Carbon (PAC) Adsorption

J. Chung[1], Y. Yoon[2], M. Kim[3,*], S.-B. Lee[3], H.-J. Kim[3], and C.-K. Choi[3]

[1]R&D Center, Samsung Engineering Co. Ltd., 415-10 Woncheon-Dong, Youngtong-Gu, Suwon, Gyeonggi-Do, Korea 443-823

[2]Department of Civil and Environmental Engineering, University of South Carolina,Columbia, SC 29208, USA

[3]Department of Civil and Environmental System Engineering, Hanyang University, 1271 Sa-1 Dong, Ansan, Gyeonggi-Do, Korea 425-791

*Corresponding author: Moonil Kim, Ph.D.

Tel: 82-31-400-5142

Fax: 82-31-408-5140

E-mail: moonilkim@hanyang.ac.kr

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Abstract

The presence of *N*-nitrosodimethylamine (NDMA) in drinking water supplies has raised concern over its removal by common drinking water treatment processes. However, only limited studies have been examined to evaluate the potential removal of NDMA by numerous water treatment technologies within a realistic range (i.e., sub μg/L) of NDMA levels in natural water due to analytical availability. In this study, a simple detection method based on scintillation spectroscopy has been used to quantify the concentration of ^{14}C-labeled NDMA at various ratios of sample to scintillation liquid. Without sample pretreatment, the method detection limits are 0.91, 0.98, 1.23, and 1.45 ng/L of NDMA at scintillation intensity ratios of 10:10, 5:15, 15:5, and 2.5:17.5 (sample : scintillation liquid), respectively. The scintillation intensity in all cases is linear (R^2 > 0.99) and is in the range of 0 to 100 ng/L of NDMA. In addition, because scintillation intensity is independent of solution pH, conductivity, and background electrolyte ion types, a separate calibration curve is unnecessary for NDMA samples at different solution conditions. Bench-scale experiments were performed to simulate individual treatment processes, which include coagulation and adsorption by powdered activated carbon (PAC), as used in a drinking water treatment plant, and biosorption, a technique used in biological treatment of waste water. The results show that coagulation and biosorption may not be appropriate mechanisms to remove NDMA (i.e., hydrophilic based on its low octanol-water partitioning coefficient, Log K_{ow} = 0.57). However, relatively high removal of NDMA (approximately 50%) was obtained by PAC at high PAC dosages and longer contact times.

1 Introduction

N-Nitrosodimethylamine (NDMA) is a toxic and carcinogenic yellow liquid that has been identified as a contaminant in drinking water, ground water, and a variety of other matrices (NRC, 1981; Leoppky and Micheljda, 1994; Mitch and Sedlak, 2004). One factor for the seemingly increasing levels of NDMA is directly related to the increasing sophistication of NDMA analysis. Since the late 1990s, a large number of studies have developed and improved NDMA analysis. Currently, several jurisdictions have implemented regulations that require widespread measurement of NDMA in raw and drinking water—such regulations led to the identification of a higher occurrence of NDMA in water than was expected. Although NDMA is listed as a priority pollutant in the United States (CFR, 2001), a federal maximum contaminant level has not been established for drinking water. However, the Ontario Ministry of the Environment and Energy established an Interim Maximum Acceptable Concentration of 9 ng/L for NDMA in drinking water (MOE, 2000). In addition, after noticing the prevalence of NDMA, the California Department of Health Services has established an interim action level of 20 ng/L, which was later reduced to 10 ng/L (DHS, 2002).

Despite the increasing concern about the adverse effect of NDMA, conventional water treatment technologies may not effectively remove NDMA. NDMA is a semi-volatile, polar organic chemical, and a highly water-soluble compound (Chemfinder, 2003). Separate studies have shown that high concentration NDMA sorbs poorly to granular activated carbon (GAC) or soil at low GAC dosages (Kaplan and Kaplan, 1985; Gumnison et al., 2000). In addition, although there is little information regarding the potential for biological removal of NDMA, NDMA is somewhat resistant to biodegradation and is difficult to remove by air stripping and ozonation (Holgnè and Bader, 1983; Siddiqui and Atasi, 2001). Iron oxides can be used to reduce

NDMA to dimethylamine (DMA) and ammonia; however, due to the slow kinetics of the reaction, this treatment method is not cost effective. Although several previous studies investigated NDMA removal using drinking water treatment technologies (Kaplan and Kaplan, 1985; Gumnison et al., 2000; Holgnè and Bader, 1983; Siddiqui and Atasi, 2001), only high initial concentrations (> 1,000 ng/L) were tested. However, since NDMA is frequently found at extremely low concentrations (parts per trillion, ppt; ng/L) in water supplies and wastewater effluents, there remain issues regarding NDMA removal at low initial concentrations from natural waters containing natural organic matter (NOM) and ions..

In order to evaluate the potential removal of NDMA by numerous water treatment technologies within a realistic range of NDMA levels in natural water, an important goal now is to be able to detect NDMA at ultra-trace levels. Analytical determination of NDMA content from surface water and wastewater commonly involves the use of gas chromatography-mass spectrometry (GC-MS or MS/MS) with chemical ionization, or traditional electron impact with continuous liquid-liquid extraction, solid phase extraction, or solid phase microextraction for ppt level NDMA analysis (Yoo et al., 2000; Mitch et al., 2003; Eaton and Briggs, 2000). However, these analytical techniques have issues regarding pretreatment requirements, compound recoveries, and detection limits. In addition, these techniques are expensive, time consuming, and require a high degree of analytical knowledge.

Less complex analytical approaches may be more suitable for lab process studies—one approach is using a liquid scintillation counter for measuring radio-labeled NDMA. Previous studies used ^{14}C-labeled NDMA for NDMA removal studies from water and soil (Fleming et al., 1996; Gumnison et al., 2000). In these studies, raw water and soil-water mixtures were spiked to initial NDMA concentrations of 1,000 ng/L and 5,000 ng/L, respectively. In addition, a liquid

scintillation counter was used to determine relatively high concentrations of ^{14}C-labeled NDMA (> 100 ng/L). However, it was still unclear how the method detection limits (MDL) were determined in terms of the ratio of sample to scintillation liquid and counting time.

To our knowledge, only limited research on NDMA removal using conventional and/or advanced water treatment technologies has been conducted at low NDMA initial concentrations. Therefore, in this study, several water treatment technologies including coagulation and powdered activated carbon (PAC) adsorption were tested to evaluate the potential of ^{14}C-labeled NDMA removal at extremely low NDMA initial concentrations (C_o = 100 ng/L). In addition, biosorption experiments were conducted using conventional activated sludge to investigate NDMA removal. For this study, a simple analytical technique for rapid determination of a ^{14}C-labeled NDMA was developed using a liquid scintillation counter. We have also demonstrated role of scintillation counting for process studies with emerging contaminant available from pharmacological studies.

2 Materials and methods

2.1 Water sources

Two water sources, Ultrapure (natural organic matter (NOM)–free) water prepared from water purification system (Direct-Q 3 system, Millipore, Korea) and raw drinking water (RDW) collected from a local water treatment plant (WTP), were selected for this study. The characteristics of the water sources used in this study are described in Table 1. An NDMA solution (C_o = 100 ng/L) was added to Ultrapure water in the presence of NaCl, Na_2SO_4, or $CaCl_2$ to generate various conductivity values (30, 60, and 120 mS/m); the pH was adjusted to

4.5, 7, or 9 (± 0.1) using HCl and/or NaOH solutions in the presence of NaCl. The NDMA sample was buffered by adding a 1 M phosphate buffer solution to the sample to create a 1 mM buffer concentration. In separate experiments, NDMA was added to RDW at a concentration of 100 ng/L.

RDW, collected from a local WTP, was used for the evaluation of NDMA removal from water. NDMA was spiked into raw water without filtration for aluminum sulfate (alum), ferric chloride, and ferric sulfate. The source water was filtered using a 1.2 μm (47 mm GF/C) glass-fiber filter (Whatman® International Ltd., Maidstone, England) to remove particulate matter prior to spiking in NDMA for PAC experiments. Control samples containing NDMA spikes in both filtered and nonfiltered water were prepared in at least triplicate.

2.2 NDMA determination by scintillation counter

^{14}C-labeled NDMA was purchased from American Radiolabeled Chemicals (St. Louis, MO, USA) with an activity of 16.8 millicuries (mCi)/mmol in deionized water. This concentration corresponds to 134.7 mg/L of NDMA. Stock solutions were initially prepared in Ultrapure water at 10,000 ng/L of NDMA and were subsequently diluted with Ultrapure water to different concentrations of 1, 5, 10, 25, 50, 75, and 100 ng/L for the calibration run. NDMA radioactivity was determined using a liquid scintillation counter (GMI, Beckman LS6500, Albertville, MN, USA). For scintillation measurements, various ratios of sample to scintillation liquid (ScintiSafe Plus 50%, Fisher Scientific, USA) were tested at 2.5:17.5, 5:15, 10:10, and 15:5. Scintillation liquid was pipetted into a standard glass vial and the sample was added to a total target volume of 20 mL. NDMA quantifications were conducted at various counting times of 1, 5, and 10 minutes (min). The scintillation liquid with Ultrapure water or RDW alone was

used to measure background radiation. The effect of inorganic salts and NOM in RDW on NDMA radioactivity was also determined.

Dissolved organic carbon (DOC) was measured using a combustion/non-dispersive infrared gas analysis method (Shimadzu Model TOC-5050A). The natural water sourcewas prefiltered using a 1.2-μm (47 mm GF/C) ashed glass-fiber filter (Whatman® International Ltd.) prior to use.

2.3 Coagulation and PAC adsorption experiments

Coagulation and PAC adsorption experiments were conducted as jar tests using a six-place gang stirrer (Phipps and Bird, Richmond, VA, USA); jars were 2L glass beakers filled with 1.5 L of NDMA-spiked water. Chemicals (see below) were added via pipette during a rapid mixing step. Mixing conditions were 2 min of rapid mixing at 100 rpm, 20min flocculation at 30 rpm, and 60 min of settling time (no mixing).

Three coagulants, aluminum sulfate, ferric chloride, and ferric sulfate (Fisher Scientific, USA), were used for jar tests to simulate coagulation, flocculation, and settling. Doses for each coagulant (3, 6, and 12 mg/L) were selected based upon an initial estimate of total organic carbon (TOC) concentration. All chemical dosages for individual experiments are summarized in Table 2.

One PAC, 6H (Coal-based, Junsei Chemical Co., Ltd., Saitama, Japan), was used to stimulate adsorption. PAC was hydrated in stock solutions (10 or 1000 mg/L) for 24 hours (hrs) in Ultrapure water prior to use and added as a slurry to the samples. Applied PAC doses ranged from 1 to 50 mg/L. NDMA was contacted with PAC at an initial concentration of 100 ng/L. Table 2 summarizes PAC doses and contact times. Additional experiments were also conducted

135 with RDW with PAC at initial concentrations ranging from 50 mg/L to 300 mg/L for contact
136 times of 5, 20, and 60 hrs. RDW was spiked with an NDMA concentration of 100 ng/L.

137

138 *2.4 Biosorption experiments*

139 The biosorption experiment was performed using activated sludge taken from a
140 municipal wastewater plant (Ansan, Korea). The biosorption experiment was conducted in an
141 Erlenmeyer flask (500 mL) containing a 100 mL NDMA solution at 20 ºC. The 1 M phosphate
142 buffered NDMA solution was adjusted to a pH of 7.0 with H_2SO_4 and NaOH solutions, and a
143 small quantity (0.05 g/L) of glucose was added to the flask for maintenance of the cells. A
144 weighed amount of the resting cells (4.5 g/L, based on dry weight) was added to the flask
145 containing the NDMA solution of known NDMA concentration. The flask was incubated in a
146 shaker at 150 rpm for 24 hrs. Periodically, samples were withdrawn and centrifuged at 5000 rpm
147 for 5 min. The supernatant was analyzed for residual NDMA. All biosorption experiments were
148 conducted in a similar manner to study the effect of biomass concentration (0.5-10.0 g/L) on
149 changes in NDMA concentrations (10-1000 ng/L). All experiments were performed in triplicate.

150

151 **3 Results and discussion**

152 *3.1 NDMA determination by scintillation counter*

153 The scintillation intensities of ^{14}C-labeled NDMA was measured by a scintillation
154 counter at pH 4.5 and at a conductivity of < 0.5 mS/m; these intensity values are presented in Fig.
155 1. All measurements were performed at a counting time of 10 min and at room temperature (20 ±
156 1 ^{o}C). Counts per minute (CPM) is commonly used to describe radiation intensity. Due to

potential instrument-to-instrument variations, the intensities are normalized by the reference intensity, CPM_0 (39 ± 1.9), for a zero concentration of NDMA.

Scintillation intensity varies depending on the ratio of sample to scintillation liquid. The order of scintillation intensity is 10:10 > 5:15 > 15:5 > 2.5:17.5 for NDMA. The scintillation intensity in all cases is clearly linear (R^2 > 0.99) in the range of 0 to 100 ng/L of NDMA. The insert in Fig. 1 shows that the relationship is linear even for very low concentrations with a 10:10 ratio, indicating the robustness of the technique. Furthermore, because the scintillation intensity at 100 ng/L NDMA is nearly 9 (2.5:17.5), 13 (15:5), 16 (5:15), and 30 (10:10) times the intensity for zero concentration, this technique demonstrates a wide dynamic range. MDLs were determined using a 5 ng/L NDMA solution of all ratios of sample to scintillation liquid.

The experiments were repeated eight times and the MDL was calculated based upon the standard deviation (c.v. ≤ 3%) of the replicate measurements. This calculation follows the United States Environmental Protection Agency MDL method (Revision 1.1) (Behymer et al., 1993). MDLs are 0.91, 0.98, 1.23, and 1.45 ng/L of NDMA for 10:10, 5:15, 15:5, and 2.5:17.5 ratios, respectively, without any pretreatment or preconcentration. In some cases, error bars based on standard deviation calculated from triplicates of scintillation intensity measurements are smaller than the graph points in the figure.

The scintillation intensities for ^{14}C-labeled NDMA measured at various counting times (CTs = 1, 5, and 10 min) are presented in Fig. 2 at various concentrations (0 to 100 ng/L). The differences between the scintillation intensity are negligible for CTs of 1 min and 5 min. However, increasing the CT from 5 min to 10 min results in a slight increase (CPM/CPM_0 = 27.9 for 5 min and CPM/CPM_0 = 30.1 for 10 min) in the scintillation intensity. Although it is unclear

how the optimal CT was determined, a previous study also recommended a 10 min CT for ^{3}H-labeled 17-β estradiol determination (Fuerhacker et al., 2001).

The dependence of scintillation intensities of the ^{14}C-labeled NDMA on various conductivities (30, 60, and 120 mS/m with NaCl at pH 7), pH (4.5, 7, and 9 at a conductivity of 60 mS/m with NaCl), and electrolyte salt type (NaCl, Na_2SO_4, and $CaCl_2$ at pH 7 and at a conductivity of 60 mS/m) is shown in Fig. 3 for a single NDMA concentration of 100 ng/L. Although the phosphate buffer is commonly effective in a pH range of 5 to 8 (Perrin and Dempsey, 1974), all pH values remained consistent over the entire range. The scintillation intensity of NDMA remained constant regardless of conductivity and pH levels, and electrolyte salt types. In addition, although the RDW contained DOC (2.4 mg/L) and various anions and cations (with a conductivity of 13.7 mS/m), the differences between the scintillation intensities are negligible. These results are consistent with previous studies that show that scintillation intensity is independent of ion concentration, pH, and ion type (Black et al., 1966; Touiton and Rubinstein, 1986).

3.2 NDMA removal by coagulation precipitation

At the dosages employed, alum and ferric coagulants neutralized particulate surface charge and precipitate metal (hydr)oxide solids. Dissolved organic compounds can precipitate with or adsorb onto the solids. Fig. 4 presents data from RDW in a control sample (no coagulant added) and after three different coagulation treatments. Error bars for the control sample indicate high reproducibility in quantifying NDMA concentrations; similar levels of reproducibility were observed throughout this study. The discussion throughout the remainder of this paper will compare percentage removal ($[1-C/C_o]\times100\%$) of NDMA to simplify the comparison between

coagulants; C_o and C are NDMA concentrations in the control sample and after experiment treatment, respectively.

Three dosages (3, 6, and 12 mg/L) of each coagulant were added to RDW with NDMA at an initial concentration of 100 ng/L. This initial concentration was selected such that 99% removal (log 2) of the compound could be quantified above the MDLs. In a previous study, hydrophobic micropollutants were successfully removed by the coagulants (Westerhoff et al., 2005). NDMA removal increases slightly with increasing coagulant dose for all the coagulants, although the removal was somewhat constant among those coagulants. (Fig. 4). However, chemical precipitation for all coagulants achieves minimal removal (< 7%) of NDMA from the RDW, indicating that coagulation may not be a good removal mechanism for this hydrophilic compound due to the presence of polar functional groups. This is presumably because NDMA's partition onto the particulate matter is minimal due to its decreased hydrophobicity as measured by octanol-water partitioning coefficient (Log K_{ow} = 0.57).

3.3 NDMA removal by PAC adsorption

Kinetic experiments were performed by collecting samples after 1, 4, and 24 h of contact in the presence of PAC at concentrations ranging from 1 to 50 mg/L. Representative PAC dose-response data for NDMA is shown in Fig. 5. At a 1 mg/L PAC dose, less than 6% of NDMA was removed after contact times of 1, 4, and 24 hr. Increasing PAC dose slightly improved NDMA removal (13% to 17%), but the effect of contact time was insignificant. Previous studies showed that longer contact times and higher PAC dose lead to higher removal of hydrophobic micropollutants (Log K_{ow} > 3) (Westerhoff et al., 2005; Yoon et al., 2005). However, NDMA

224 removal was low (less than 20%) in this study, presumably because of the lower levels of
225 hydrophobic interaction (i.e., adsorption) between the PAC and NDMA.

226 To obtain higher NDMA removal by PAC, additional experiments were conducted at
227 relatively high PAC dosages (50 – 300 mg/L) and longer contact times (5, 20, and 60 hr) with
228 RDW. Fig. 6 shows the relationship between PAC dose and NDMA response. At a dosage of
229 300 mg/L PAC, 45% of NDMA was removed after contact times of 5, 20, and 60 h, respectively.
230 Increasing PAC dosage improved NDMA removal, while the effect of contact time became
231 insignificant. These results indicate that hydrophilic NDMA can still be removed at high PAC
232 dosage in the presence of natural organic matter.

233

234 *3.4 NDMA removal by biosorption*

235 Experiments evaluating the influence of initial NDMA concentration on the removal of
236 NDMA by biosorption at fixed values of pH and biomass concentration (7.0 and 5.0 g/L) were
237 carried out. The objective of these experiments was to observe the effect of different parameters
238 on the rate of biosorption (Figure 4a). As shown in Fig. 7a, an increase of initial NDMA
239 concentration increased the biosorption efficiency up to 20%, implying that even concentrated
240 biomass could not absorb the NDMA significantly.

241 Experiments evaluating the influence of biomass concentration on the process of NDMA
242 removal by biosorption were also carried out. The objective of these experimentswas to observe
243 the effect of biomass concentration on the rate of biosorption. The results obtained are shown in
244 Fig. 7b. With an initial concentration of 10 ng/L of NDMA, little change was observed in the
245 biosorption efficiency of 0.5 to 10 g/L of biomass, implying that biomass concentration does not
246 significantly affect biosorption efficiency.

4 Conclusions

- Scintillation spectroscopy proved to be a simple and useful tool for quantifying radio-labeled ^{14}C-labeled *N*-nitrosodimethylamine at low levels of concentration. MDLs were 0.91, 0.98, 1.23, and 1.45 ng/L of NDMA with sample to scintillation liquid ratios of 10:10, 5:15, 15:5, and 2.5:17.5 respectively, without preconcentration. These low detection limits properly define a concentration range for lab process experiments because they represent a realistic range of NDMA concentrations in contaminated natural water. Scintillation intensity increases with increasing sample to scintillation liquid ratio (i.e., sample concentration) ranging from 2.5:17.5 to 10:10, while scintillation intensity decreases significantly at a higher ratio (i.e., 15:5).
- Alum, ferric chloride, and ferric sulfate coagulants removed less than 10% of NDMA. This is presumably because NDMA's partition onto the particulate matter is minimal. Addition of 10 mg/L of PAC with a 4 h contact time removed less than 5%. This was also because low levels of hydrophobic interaction between PAC and NDMA occurred. However, higher PAC dosages improved NDMA removal.
- In addition, it was observed that the removal of NDMA by biosorption was insignificant at the limited conditions, indicating that biosorption may be an ineffective mechanism for removing hydrophilic NDMA even at an extremely low initial concentration (100 ng/L).

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Table 1. Characteristics of source waters

Source water	DOC (mg/L)	Conductivity (mS/m)	pH	NDMA (ng/L)
Ultrapure water	< 0.2	< 0.5, 30, 60, and 120[a]	4.5, 7.0, and 9.0	100
WTP raw water	2.4	13.7	7.1	100

[a]Conductivity was adjusted by adding NaCl, Na_2SO_4, and/or $CaCl_2$ solution after pH was buffered by a phosphoric solution.

Table 2. Water quality and dosages of the experiments

Parameter	Value
Coagulation experiment	
Initial pH	7.1
Dosages [mg/L]	3, 6, and 12
Adsorption experiment	
PAC brand	6H
PAC dosages [mg/L]	1, 2, 4, 6, 8, 10, 25, and 50 mg/L for 1, 4, and 24 hr 50, 100, 200, 250, and 300 mg/L for 1 hr
Biosorption experiment	
MLSS concentrations	0.5 – 10 g/L

Figure captions

Fig. 1. Dependence of scintillation intensity on the ^{14}C-labeled NDMA concentration at pH 4.5, conductivity ≤ 0.5 mS/m, and CT = 10 min. ● sample/scintillation liquid ratio = 2.5:17.5; ◊ sample/scintillation liquid ratio = 5:15; ○ sample/scintillation liquid ratio = 10:10; Δ sample/scintillation liquid ratio = 15:5. *CPM* is the measured scintillation intensity at [NDMA], and CPM_0 (39 ± 1.9) is the scintillation intensity at the reference condition (zero NDMA). The insert shows the detail at low concentrations for NDMA.

Fig. 2. Dependence of scintillation intensity on the ^{14}C-labeled NDMA concentration at various counting times. pH 4.5; conductivity < 0.5 mS/m; sample/scintillation liquid ratio = 10:10. Δ CT = 1 min; ♦ CT = 5 min; ○ CT = 10 min.

Fig. 3. Dependence of scintillation intensity of the ^{14}C-labeled NDMA (100 ng/L) on the (a) conductivity (30, 60, and 120 mS/m with NaCl at pH 7), (b) pH (4.5, 7, and 9 at conductivity 60 mS/m with NaCl), and (c) electrolyte salt type (NaCl, Na_2SO_4, and $CaCl_2$ at pH 7 and conductivity 60 mS/m).

Fig. 4. Effect of coagulant types on NDMA removal from RDW.

Fig. 5. Effect of contact time (1, 4, and 24 h) on NDMA (C_o = 100 ng/L) removal from RDW at relatively low PAC dosages (1 – 50 mg/L).

Fig. 6. Effect of contact time (5, 20, and 60 h) on NDMA (C_o = 100 ng/L) removal from RDW at relatively high PAC dosages (50 –300 mg/L).

Fig. 7. Effect of initial NDMA concentration (10-1000 ng/L) and biomass concentration (0.5-10.0 g/L) in biosorption experiment. (◆: NDMA final concentration (ng/L); △: biosorption efficiency (%)).