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Natural attenuation of NDMA precursors in an urban, wastewater-dominated wash

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ABSTRACT

N-Nitrosodimethylamine (NDMA) is a disinfection by-product (DBP) that is potentially carcinogenic and has been found to occur in drinking water treatment systems impacted with treated wastewater. A major gap in NDMA research is an understanding of the persistence of wastewater-derived precursors within the natural environment. This research sought to fill this knowledge gap by surveying NDMA precursors across the length of a wastewater effluent-dominated wash. Significant precursor reduction (17%) was found to occur from introduction into the wash to a point 9 h downstream. This reduction translates into a half-life of roughly 32 h for bulk NDMA precursors. Further laboratory experiments examining rates of photolysis, biodegradation and loss to sediments, illustrated that both photolytic and biological degradation were effective removal mechanisms for NDMA precursors. Loss to sediments that were acquired from the wash did not appear to reduce NDMA precursors in the water column, although a control conducted with DI water provided evidence that significant NDMA precursors could be released from autoclaved sediments (suggesting that sorption does occur). Microbial experiments revealed that microbes associated with sediments were much more effective at degrading precursors than microbes within the water column. Overall, this study demonstrated that natural processes are capable of attenuating NDMA precursors relatively quickly within the environment, and that utilities might benefit from maximizing source water residency time in the environment, prior to introduction into treatment plants.

1. Introduction

N-Nitrosodimethylamine (NDMA) is a disinfection by-product (DBP) that forms readily in drinking water treatment systems that utilize chloramines for disinfection. Within US drinking water systems, NDMA has been found above detection limits in ~50% of samples in systems using chloramines for primary disinfectant (Woods and Dickenson, 2015; USEPA, 2012). This DBP is a potential human carcinogen that is believed to pose a greater health risk than currently regulated DBPs (e.g., trihalomethanes and haloacetic acids). California's Office of Environmental Health Hazard Assessment has a health goal of 3 ng/L (CDPH, 2013), while Ontario (Canada) has a guideline of 9 ng/L, and the World Health Organization has a guideline of 100 ng/L (WHO, 2008). The USEPA required monitoring of NDMA as part of the second Unregulated Contaminant Monitoring Rule (UCMR2; USEPA, 2012) and a future regulation on NDMA is currently under consideration. Numerous treatment strategies have been examined for the removal of NDMA; UV treatment and biofiltration, for example, are treatment strategies that have found some successful applications at removal (Mitch et al., 2003b). This contaminant, however, is water soluble, relatively non-reactive and consequently fairly stable in the aqueous environment (Wilczak et al., 2003; Krasner et al., 2013). Once formed, NDMA is difficult to remove from treated waters and research efforts have consequently also focused on identifying methods to destroy and/or remove NDMA precursors. Such strategies have included precursor oxidation (Chen and Valentine, 2008; Lee et al., 2008; Shah and Mitch, 2012), activated carbon applications (Sacher et al., 2008; Farré et al., 2011; Hanigan et al., 2012), and riverbank filtration (Sacher et al., 2008; Krasner et al., 2012).

In addition to treatment strategies for the removal of NDMA and NDMA precursors, ongoing research has sought to elucidate the

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major sources of NDMA precursors so as to better eliminate NDMA formation. Natural sources of NDMA precursors within the environment have been examined (e.g., natural organic matter and algae), but precursors derived from human activities appear to have much higher NDMA yields (Dotson et al., 2009; Gerecke and Sedlak, 2003; Krasner et al., 2013). Materials from water treatment have been shown to contribute some degree of NDMA precursors, including anion exchange resins and organic polymer aids (such as the widely-used polyDADMAC) (Wilczak et al., 2003; Park et al., 2009; Nawrocki and Andrzejewski, 2011; Flowers and Singer, 2013). Rubber gaskets and seals originating from distribution systems have further been found to contribute NDMA precursors (Morran et al., 2011; Teefy et al., 2011; Woods et al., 2015). On the whole, however, treated wastewater has been identified as an important and major source of NDMA precursors to drinking water systems (Krasner et al., 2009; Nawrocki and Andrzejewski, 2011; Russell et al., 2012). Though much research has focused on compounds likely to be prominent NDMA precursors (e.g., numerous pharmaceuticals), research is largely lacking as to the fate of NDMA precursors in the actual environment. After precursors are discharged from wastewater treatment plants, they are transported some distance through the environment until entry into intake pipes at drinking water treatment plants (the exception to this being direct potable reuse, which to-date is not a common practice).

The fate of precursors during the transition from wastewater to drinking water systems is currently not well understood. Though studies have examined the potential fate of NDMA in natural systems (e.g., photolysis and biodegradation have been found to effectively remove NDMA) (Williams et al., 2011; Plumlee and Reinhard, 2007), few studies have examined the attenuation of NDMA precursors in the natural environment. Previous research conducted by Schreiber and Mitch (2006) utilized boron as a conservative tracer of wastewater effluent, and followed the removal of NDMA precursors within a river in Connecticut and an engineered treatment wetland in Northern California. River samples taken between treatment plants in the upper portion of the river (~8 km) had precursor removal rates of ~10% in spring and ~30% in summer. The engineered wetland was found to remove ~50% of precursors over 7 days. Further research conducted by Pehlivanoglu-Mantas and Sedlak (2006), coupled incubation studies of wastewater effluents, inoculated with activated sludge mixed liquor, and measurements from a wastewater effluent-impacted river in Southern California. Wastewater effluents incubated for 20–30 days were found to average a decrease of 24% in NDMA-FP values, but one site was found to decrease by 80% over 30 days. The effluent-impacted river had a NDMA precursor reduction of ~30% over the 40 km sampling transect.

The research presented here expanded on these previous studies in that sampling of a wastewater effluent-dominated wash was coupled with multiple bench-scale experiments to help elucidate potential removal mechanisms. This study was somewhat exploratory in that specific mechanisms of NDMA precursor removal are largely not understood. Potential losses due to sediment sorption, photolysis and/or biological degradation were investigated. This research sought to fill the knowledge gaps of precursor fate in the environment, identify potential removal mechanisms, and provide a knowledge base from which subsequent studies can be designed to more precisely define and quantify removal mechanisms. Such information will aid researchers in understanding the fate of NDMA precursors after discharge from wastewater plants and ultimately in curbing NDMA formation in drinking water. The results demonstrate that NDMA precursors can be significantly attenuated in the environment within a relatively short timeframe.

2. Materials and methods

2.1. Site description

The Las Vegas Wash is a hydrographic basin that drains the Las Vegas Valley (3998 km^2) in a 20 km channel that feeds into Lake Mead (Gautam et al., 2014). Originally an ephemeral channel, the wash was created by modern settlement that has generated perennial flow from urban runoff, stormwater drainage, shallow groundwater, and treated wastewater. Since the 1950s, hundreds of hectares of wetlands have been created in the wash, improving water quality and wildlife diversity. Lake Mead is the largest reservoir in the United States, fed primarily by the Colorado River (~95%), and a very important source of drinking water for Southern Nevada, Arizona and Southern California (Holdren and Turner, 2010; Gautam et al., 2014). For the Las Vegas Valley, drinking water intakes are downstream of where the wash enters the lake, and consequently water quality in the wash is of significance. Approximately 900 million gallons are withdrawn daily from Lake Mead for use in southern Nevada, with about half of this returned via the Las Vegas Wash as treated wastewater (LaBounty and Horn, 1997).

The studied portion of the wash is illustrated in Fig. 1 and extends approximately 18 km, from just below a major input of treated wastewater until about a km before entry into Lake Mead. Weather conditions were not unusual for that time of year. The daily temperature averaged 31 °C (with an historic average of 32 °C), and month-to-date precipitation was 0.35 mm (with an historic average of 0.28 mm). Over the duration of sampling, the flow averaged ~7.0 cubic meters per second throughout the wash (USGS, 2015), with an estimated 8.8 h of travel time between the site furthest upstream (LW9) and one of the sites furthest downstream (LW1). This estimate was derived based upon previous tracer studies conducted by Southern Nevada Water Authority (SNWA) personnel (Leising, 2003). The hydraulic pulse was determined based upon the daily peak flow on August 26, 2014, as measured by 4 USGS gages in the main wash (sites LW9, LW7, LW3, LW1; Fig. S1). The previously determined solute:hydraulic pulse (Leising, 2003) was then applied to determine an average solute travel time of 8.8 h from site LW9 to LW1. A conservative estimate of uncertainty for this average may be as high as 50% (based upon personnel communication with Joe Leising, formerly with SNWA).

Width and depth vary along the wash and have been estimated to range from ~22 to 85 m and ~1.2 to 1.8 m, respectively (Buckingham and Whitney, 2007). Water temperature averaged 28 °C, pH 8.0, conductivity 2500 μS/cm and dissolved oxygen (DO) averaged 8.6 mg/L (data provided in Supplementary Material, Tables S1 and S2). Average ammonia (as N), nitrate/nitrite (as N), total organic carbon (TOC) and specific UV absorbance at 254 nm (SUVA) were 0.084 mg/L, 11 mg/L, 4.9 mg/L and 1.7 (L/mg-M), respectively (data provided in Supplementary Material, Tables S3 & S4). The riverbed contains fine materials and mud, but courser sediments and gravel can be found at points downstream. The wash contains several grade flow control structures and sections of wetlands. Vegetation varies along the wash from sparsely vegetated cover to herbaceous grasses to cottonwood and willow forested regions, with shrubland regions occurring as the most dominant land covering (Shanahan et al., 2007). Daily discharge from the main wastewater plant (CCWRD) is fairly consistent. Fig. S2 illustrates the USGS gage readings (USGS, 2015) at site LW9 over the week during which the main sampling event occurred. At this point in the wash, there was a daily high around noon of ~9 cubic meters per second. Flow continued at ~7.5 cubic meters per second for the remainder of the day, dropping after midnight to a low of ~4 cubic meters per second by 6 am.