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Highlighted article

Leaching risk of *N*-nitrosodimethylamine (NDMA) in soil receiving reclaimed wastewater

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Abstract

N-Nitrosodimethylamine (NDMA) is a potential carcinogen frequently found in treated wastewater as a byproduct of chlorination. The potential for NDMA to contaminate the groundwater is a significant concern. A solute fate and transport model, Hydrus-1D, was used to evaluate the leaching potential of NDMA under different irrigation practices and soil properties. The results indicate that the risk of NDMA to reach the ground water is slim, when the reclaimed wastewater is applied under the customary conditions for landscape irrigation. The NDMA disappears in the reclaimed wastewater receiving soils rapidly through the microbial degradation and the volatilization processes. The factors that enhance the leaching risk are the soil hydraulic conductivity, the NDMA adsorption constants, and the irrigation intensity. When the hydraulic conductivity of soil is high, the NDMA adsorption constant of soil is low and/or the irrigation intensity is high, the NDMA leaching risk may dramatically increase. To reduce the NDMA leaching risk, it is imperative that the fields be irrigated at the proper volume and frequency and attention be paid to fields with soils having high-hydraulic conductivities and/or low-NDMA adsorption constants.

Keywords: Fate and transport; Model; Landscape irrigation; Degradation; Volatilization

1. Introduction

In California, around $650 \times 10^6\,m^3$ of reclaimed wastewater is beneficially used annually and the uses will continue to increase, according to the California Water Plan 2005 Update (California Water Resources Control Board, 2003). Disinfection is an essential treatment step preparing reclaimed wastewater destined for irrigation. As one of the most common and reliable disinfection method, chlorination is effective at eliminating pathogens in the water. But it produces unintended disinfection byproducts (DBPs). Among these emerging trace compounds, *N*-nitrosodimethylamine (NDMA) is known for its high-cancer potency and is frequently found in treated wastewater (Mitch and Sedlak, 2002; Mitch et al., 2003; Levine and Asano, 2004). Under the most conducive condition, the mean concentration of NDMA was found to be as high as $1000\,ng\,L^{-1}$ (Water Reuse Foundation, 2005; Gan et al., 2006).

There is inadequate technical information to assess the potential adverse impacts of NDMA released during the landscape application of reclaimed wastewater. When they are present in the soil, NDMA are subject to volatilization in the ambient environment and are readily degradable through chemical and biological reactions. They are also expected to be adsorbed by the soil organic matter. As a result, they are not likely to enter the plant tissue through root absorption (Arienzo et al., 2006). However, if releasing to natural water bodies, the potential eco-toxicological consequences cannot be overlooked.

The environmental behavior of NDMA has been extensively studied on degradation in soils (Tate and Alexander, 1975; Oliver et al., 1979; Mallik and Tesfai,

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1981; Kaplan and Kaplan, 1985; Gunnison et al., 2000; Yang et al., 2005; Gan et al., 2006; Arienzo et al., 2006), volatilization rates (Oliver, 1979), Henry's law constants (Mirvish et al., 1976), and soil adsorption coefficients (Dean-Raymond and Alexander, 1976; Oliver et al., 1979; Kaplan and Kaplan, 1985; Gunnison et al., 2000; Yang et al., 2005). These processes occur simultaneously and *in situ*. Arienzo et al. (2006) followed its short-term dynamic behavior in turf grass fields for 14 d using ^{14}C labeled NDMA. Gan et al. (2006) investigated the leaching and degradation of NDMA under typical conditions of turf grass soil irrigated with wastewater effluent. However, the results are often limited by the detection limits of the equipment and methods available for analysis. In addition, it is hard to tell the environmental risk of NDMA under different scenarios based on the experimental data itself. For useful environmental risk assessments, it is important to combine the field-based experiments with mathematical modeling. Computation model allows simultaneous simulations of the interactive processes governing the fate and transport of NDMA in soil–water–plant system. The outcomes are useful at answering "what if" types of questions. Starting with a reference scenario, the worst or best possible cases may be illustrated.

The purpose of this research was to evaluate the leaching risks of NDMA under fields irrigated with reclaimed wastewater, based on simulation outcomes from model of Hydrus-1D. The key factors that affected the fate and transport of NDMA in soils were investigated.

2. Model approach

The fate and transport of NDMA was simulated by Hydrus-1D (version 2.0), which is a one-dimensional finite element model simulating the movement of water, heat, and multiple solutes in variably saturated heterogeneous or layered soils subject to a variety of atmospheric and other boundary conditions (Šimůnek et al., 1998). In Hydrus-1D, the water flow is modeled by the Richards' equation:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x}\left[K(h)\left(\frac{\partial h}{\partial z} + 1\right)\right] - S, \quad (1)$$

where h is the water pressure head (cm); θ is the volumetric water content ($cm^3\,cm^{-3}$); $K(h)$ is the unsaturated hydraulic conductivity function ($cm\,h^{-1}$); and S is the root water uptake term ($cm^3\,cm^{-3}\,h^{-1}$). In model simulation, the van Genuchten (1980) equation was used to describe the relationship among θ, h, and $K(h)$.

The convective-dispersive equation describing NDMA transport in the soil profile is as follows:

$$\frac{\partial \theta c}{\partial t} + \frac{\partial \rho s}{\partial t} + \frac{\partial a_v g}{\partial t} = \frac{\partial}{\partial x}\left(\theta D_w \frac{\partial g}{\partial x}\right) + \frac{\partial}{\partial x}\left(a_v D_g \frac{\partial g}{\partial x}\right) - \frac{\partial qc}{\partial x} - \mu_w \theta c - \mu_s \rho s - \gamma_w \theta - \gamma_s \rho, \quad (2)$$

where the notations c, s, and g denote NDMA concentrations in the water ($ng\,cm^{-3}$), solid ($ng\,g^{-1}$), and gaseous ($ng\,cm^{-3}$) phases, respectively; ρ is the soil bulk density ($g\,cm^{-3}$); a_v is the volumetric air content ($cm^3\,cm^{-3}$); D_w is a molecular diffusion coefficient in water phase ($cm^2\,d^{-1}$), and D_g is a molecular diffusion coefficient in gaseous phase ($cm^2\,d^{-1}$); q is the volumetric water flux ($cm\,d^{-1}$); μ_w and μ_s are the first-order degradation rate constants for NDMA in the water and solid phases (d^{-1}), respectively; γ_w and γ_s are zero-order degradation rate constants for NDMA in the water ($ng\,cm^{-3}\,d^{-1}$) and solid ($ng\,g^{-1}\,d^{-1}$) phases, respectively.

The governing equations for flow and transport were solved numerically using Galerkin-type linear finite element schemes. The NDMA uptake by plants was not considered as Arienzo et al. (2006) showed that the absorption of NDMA by turf grass grown in lysimeters was negligible.

3. Model parameters and validation

The data from Gan et al. (2006) were used to validate the model performance. Information based on the field experiment conducted by Gan et al. (2006) as well as other published literature was extracted to define the parameters needed for the model simulations, summarized as follows.

3.1. Initial conditions

The initial volumetric water contents were assumed as $0.1\,cm^3\,cm^{-3}$ soil at all depths of the 89 cm soil profile (the depth of the lysimeters). Initial NDMA concentrations in the soil were set zero at all depths of soils following the actual condition at the field.

3.2. Boundary conditions

At the soil surface, the boundary was set at the atmospheric conditions. The lower boundary was set as free drainage. The boundary layer at the soil surface where NDMA diffuses from soil to the atmosphere was assumed to be 0.5 cm (Jury and Horton, 2004).

3.3. Irrigation practice

The irrigation frequency was set to three times per week. Each irrigation lasted 8 h from the midnight to the morning. The amount of irrigation varied from 110% to 160% of the cumulative potential evapotranspiration (ET_0) since the previous irrigation. The ET_0 was obtained from the California Irrigation Management Information System (CIMIS) for summer periods at Riverside, California. The total volume of irrigation was 104 cm during a period of 113 d. The reclaimed wastewater contained $930\,ng\,L^{-1}$ of NDMA, resulting a total loading of $967\,mg\,m^{-2}$.