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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 ecotoxicological 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:

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

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:

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

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 $967\,mg\,m^{-2}$.

3.4. Soil hydraulic properties

The leaching potential of NDMA under two types of soil, the loamy sand soil and the sandy loam soil was evaluated. In the simulations, the hydraulic parameters for the loamy sand soil were selected from the default data file of the Hydrus-1D (Table 1). For the sandy loam soil, the data from Saito et al. (2006) was used who determined the hydraulic parameters of the sandy loam soil in the turf grass covered lysimeters adjacent to the filed experiment plots (Table 1).

3.5. NDMA distribution

The liquid and gas phase concentrations are related via the dimensionless Henry's law constant K_H and the liquid and solid phase concentrations via the linear distribution coefficient K_d ($cm^3\,g^{-1}$). The dimensionless Henry's law constant was set as 1.0×10^{-4}, based on the average of reported values by Mirvish et al. (1976) and by IPCS (http://www.inchem.org/documents/cicads/cicads/cicad38.htm). Yang et al. (2005) measured the K_d for the sandy loam lysimeters adjacent to the field experiment plots, which was $0.56\,cm^3\,g^{-1}$. The K_d of NDMA in difference soils was small (Gunnison et al., 2000). For the convenience, the linear adsorption coefficient (K_d) was set as $0.56\,cm^3\,g^{-1}$ for both type of soil.

3.6. Biodegradation rates

The degradation of NDMA in the soils may follow either a first-order reaction model (Gan et al., 2006) or zero-order reaction model (Arienzo et al., 2006). Since it is one of the most important factors controlling the fate and transport of NDMA in soils, the leaching potentials under both types of reaction were evaluated. Table 2 summarizes the degradation rate constants used in the numerical simulation, based on the experimental data of Gan et al. (2006) and Arienzo et al. (2006).

Table 1
Soil hydraulic parameters used in numerical simulations[a]

Parameters	θ_r	θ_s	α (cm^{-1})	n	K_s ($cm\,h^{-1}$)
Loamy sand	0.057	0.41	0.124	2.28	14.6
Sandy loam	0.012	0.44	0.028	1.38	1.43

[a] θ_r and θ_s are the residual and saturated volumetric water content, respectively. α and n are fitting parameters between the water suction in soils and the volumetric water content in the van Genuchten model. K_s is the saturated hydraulic conductivity.

Table 2
Degradation constants of NDMA for the loamy sand soil and sandy loam soil, data based on Gan et al. (2006) for the first-order kinetics and Arienzo et al. (2006) for the zero-order kinetics

Depth (cm)	First-order (day^{-1})		Zero-order ($ng\,cm^{-3}\,d^{-1}$)	
	Loamy sand	Sandy loam	Loamy sand	Sandy loam
0–10	0.170	0.620	169.9	255
10–25	0.124	0.113	124.0	18.6
25–50	0.012	0.052	12.0	1.8
50–89	0.012	0.052	12.0	1.8

3.7. Diffusion coefficients

The molecular diffusion coefficients of NDMA in water (D_w in Eq. (2)) and in air (D_g in Eq. (2)) were set to 0.84 and $11.578\,cm^2\,d^{-1}$, respectively, based on the database from GSI Environmental Inc. (formerly Groundwater Service Inc.) (http://www.gsi-net.com/UsefulTools/ChemPropDatabaseHome.asp).

The simulation outcomes based on the previously defined parameters represent the typical conditions of turf grass irrigation with wastewater effluent. Since the concentrations of NDMA in the leachates are very low and the measurements are often limited by the detection limit of the instrument, while the model simulation does not have a low limit, thus direct comparison of the mass flow between measurement and simulation is difficult. The model simulation results were in agreement with the field observations by Gan et al. (2006) that the concentrations of NDMA in the leachates were consistently below $2\,ng\,L^{-1}$ for the 113-d duration, indicating the model predictions might be well reveal the fates of NDMA in soils through irrigation of reclaimed wastewater.

4. Simulation scenarios

Under the typical conditions defined in the previous section, it is unlikely that NDMA will contaminate the groundwater. The case can be used as a reference scenario (default case). The leaching potential of NDMA may be elevated through higher NDMA loading, favored soil characteristics and irrigation practices. Simulations were conducted to evaluate the leaching potential of NDMA under the following scenarios.

4.1. NDMA input

The leaching potential of NDMA in soils is in proportion to its inputs. Under the default conditions, the reclaimed wastewater contained $930\,ng\,L^{-1}$ of NDMA. In worse cases, the NDMA in the applied wastewater might be two or three time higher than that of the default case.

4.2. Soil characteristics

Hydraulic properties are important factors controlling the water transport in soils, and hence the leaching of NDMA. Two types of soil were used in the simulation. The loamy sand soil ($K_s = 14.6\,cm\,h^{-1}$) represents soils with high-hydraulic conductivity, while sandy loam soil ($K_s = 1.43\,cm\,h^{-1}$) represents soils with low-hydraulic conductivity. In addition to the soil hydraulic properties, the liquid and solid phase distribution coefficient (K_d) has a direct impact on NDMA leaching. Studies showed that NDMA has negligible affinity for soils (Gunnison et al., 2000; Yang et al., 2005). Under the default case, the K_d was set to 0.56 $cm^3\,g^{-1}$ for two type of soils. The simulation scenarios at K_d of 0.28, 0.14, and 0 $cm^3\,g^{-1}$ (worst case) was included in the simulation scenarios to demonstrate the NDMA leaching potential under less affinity for soils.

4.3. Irrigation practices

The leaching potential of NDMA in soils may be significant affected by the irrigation management. Over-irrigation will accelerate the leaching of NDMA. For comparison, the scenarios at the irrigation volume of $1 \times ET_0$, $2 \times ET_0$, and $3 \times ET_0$ were conducted. In addition to the amount of irrigation, the leaching potential may be affect by the irrigation frequency. Under the default scenario, the irrigation was applied three times per week with a period of 8 h. In cases the irrigation is applied daily or once per week with the same irrigation period, the irrigation rate has to be reduced or increased to meet the same amount of irrigation. Consequentially, the NDMA leaching potential will be different.

The above-described scenarios were conducted by varying the mentioned parameters one at a time while holding the values of the other parameters at the default levels.

5. Results

5.1. NDMA leaching under first-order degradation

The simulated outcome of default conditions is represented by the three times per week irrigation frequency, one time NDMA concentration in the irrigation water, and K_d of 0.56 $cm^3\,g^{-1}$ in Fig. 1. Under this condition, a negligible amount of NDMA, approximately 0.001% of the chemical added through irrigations, may be leached below the 89 cm loamy sand soil profile in 113 d.

The simulation outcomes show that the leaching loss was not significantly affected by NDMA inputs up to three times of the default concentration. The other factors have significant effects to the leaching loss. For examples, the NDMA leaching would rise from the default level to >1% when irrigation volume increases to $3 \times ET_0$ or drop from the default level to <0.000001% when the irrigation volume is held at $1 \times ET_0$. The initial NDMA break through of the 89 cm soil profile took place at >100, 60, and 40 d when the irrigation volume increased from 1×,

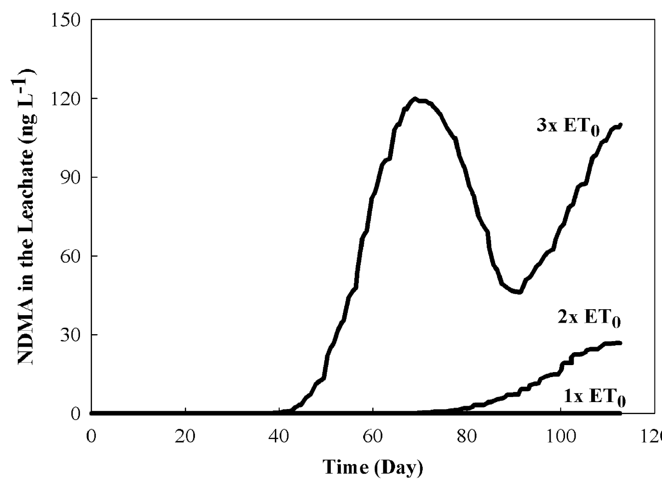

Fig. 2. Temporal changes of NDMA concentrations in the bottom flux of the loamy sand soil at different irrigation intensity under first-order degradation kinetics.

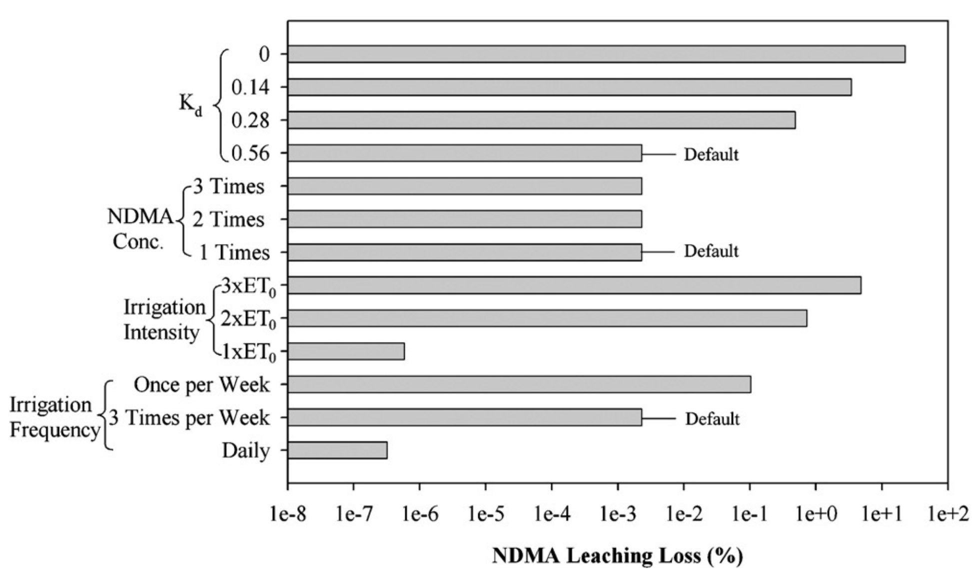

Fig. 1. NDMA leaching rates at various simulation scenarios under the first-order degradation kinetics in loamy sand soil. The default simulation is represented by the three times per week irrigation frequency with a 8 h period, one time NDMA concentration in the irrigation water (930 $ng\,L^{-1}$), and K_d of 0.56 $cm^3\,g^{-1}$.

$2\times$, to $3\times ET_0$, respectively (Fig. 2). When the irrigation frequency is changed from the default level of three times per week to daily or once per week, the NDMA leaching would drop from the default level to <0.000001% or rise from the default level to 0.1% of the added NDMA when the irrigation frequency changed from the default level to daily irrigation. When all parameters are held constant, the NDMA leaching loss increased with reduction in K_d. The initial break through time rose from 25, 45, 70, to >100 d as the K_d for NDMA was changed from 0, 0.14, 0.28, to $0.56\,cm^3\,g^{-1}$, respectively (Fig. 3). When the irrigation intensity exceeds $2\times ET_0$ and the $K_d<0.28\,cm^3\,g^{-1}$, the NDMA leaching were >0.1% of the inputs and the NDMA concentrations in leacheates may greatly exceed $10\,ng\,L^{-1}$ (Figs. 2 and 3), the California Department of Health Services notification level for NDMA in drinking water. Under no other conditions did the NDMA concentrations in the leachates exceed the $2\,ng\,L^{-1}$ LOD.

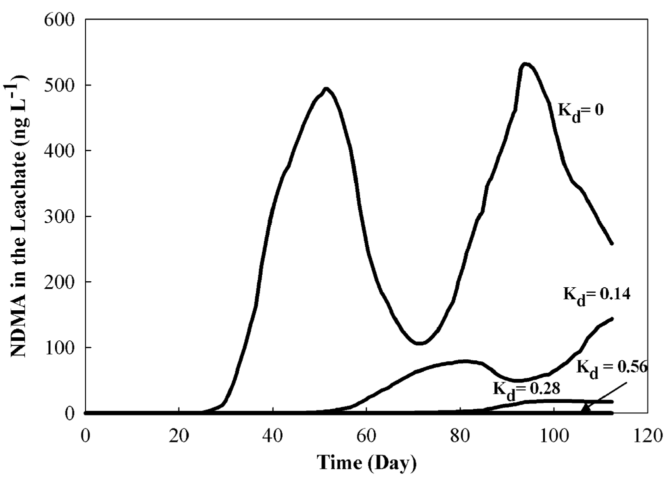

Fig. 3. Temporal changes of NDMA concentrations in the bottom flux of the loamy sand soil at different distribution coefficient (K_d) under the first-order degradation kinetics.

In model simulation, the soils were continuously irrigated with the reclaimed wastewater (NDMA concentration of $930\,ng\,L^{-1}$). Therefore, more peaks would be observed if the simulation lasted longer. Since the amount of irrigation was adjusted weekly according to the ET_0, which varies significantly from summer to winter, the leaching flux would change according. There, the peaks might become smaller during the winter period (less irrigation) or higher for hotter weather (more irrigation).

The patterns of NDMA leaching in the sandy loam soils were similar to those of the loamy sand soils (data not shown), namely, the relatively higher leaching rates were associated with greater intensity of irrigation, higher irrigation frequency, and smaller value of K_d. However, the actual NDMA leaching rates in sandy loam soils were two orders of magnitude lower than those in the loamy sand soils.

5.2. NDMA leaching under zero-order degradation

Based on the zero-order NDMA degradation kinetics (Table 2), the leaching rates in the soils were considerably lower than when the first-order degradation kinetics was imposed (Fig. 4). The patterns, however, were similar and irrigation frequency, irrigation intensity and K_d were the most significant factors in determining the leaching potentials. At the default level, the NDMA leaching under the loamy sand soil was an insignificant amount of 10^{-21}% of the total inputs from irrigation over the 113-d simulation period. As the irrigation frequency shifted from three times per week to daily or to once per week, the NDMA leaching dropped to 10^{-28}% or rose to 10^{-18}%, respectively. When the irrigation intensity rose from $1\times$, $2\times$, to $3\times ET_0$, the NDMA leaching over the simulation period leapfrogged

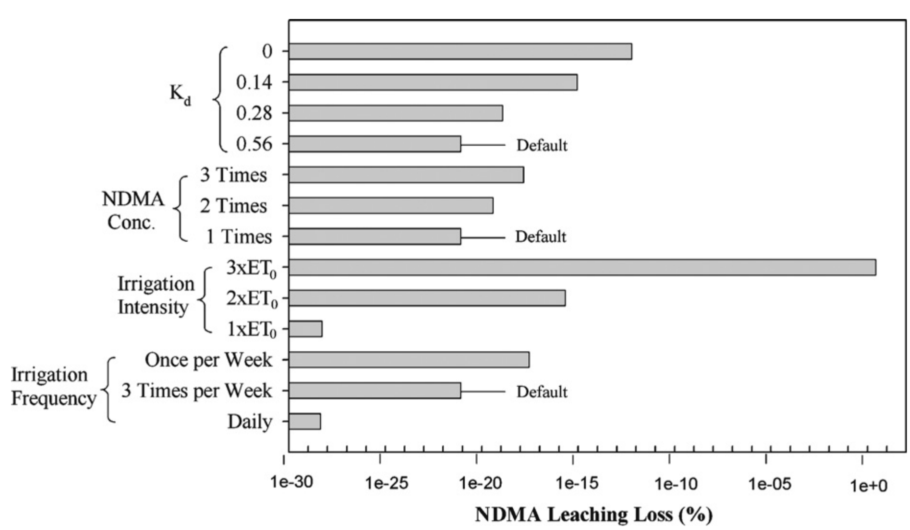

Fig. 4. NDMA leaching rates at various simulation scenarios under the zero-order degradation kinetics in loamy sand soil. The default simulation is represented by the three times per week irrigation frequency with a 8 h period, one time NDMA concentration in the irrigation water ($930\,ng\,L^{-1}$), and K_d of $0.56\,cm^3\,g^{-1}$.

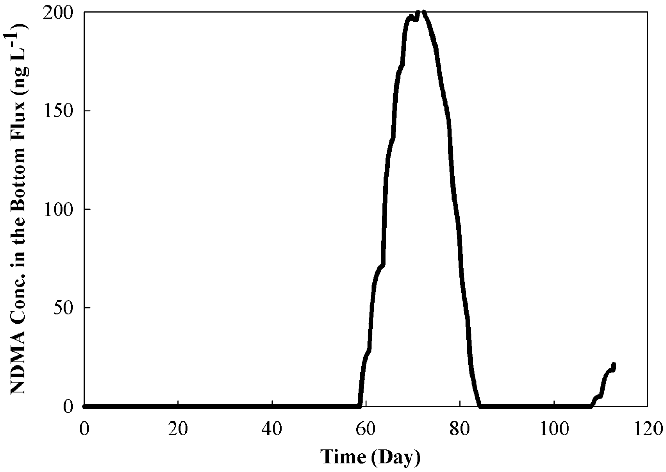

Fig. 5. Temporal changes of NDMA concentrations in the bottom flux of the loamy sand soil in case of $3 \times ET_0$ irrigation intensity and zero-order degradation.

from 10^{-28}%, 10^{-20}%, to 10^{-0}% of the total inputs, respectively. As K_d decreased from 0.56 to $0\,cm^3\,g^{-1}$, the NDMA leaching losses rose from 10^{-20}% to 10^{-12}% of the respective inputs. The actual leaching losses were minuscule and the changes were insignificant. When the irrigation intensity was $3 \times ET_0$, however, the NDMA leaching loss over the simulation period reached 1% of the total inputs. Under this condition, the NDMA started to break through the 89 cm soil profile approximately 60 d of simulated water applications and the concentration in the leachates reached $200\,ng\,L^{-1}$ and then gradually subsided over the next 25 d (Fig. 5). Under no other conditions, the NDMA concentrations in the leachates reach the $10\,ng\,L^{-1}$, the California Department of Health Services notification level for NDMA in drinking water.

The zero-order NDMA degradation kinetic more accurately reflected with the actual high-rate removal of ^{14}C-NDMA in lysimeters (Arienzo et al., 2006) than that based on the first-order degradation rate constants. The likelihood of NDMA leaching during the landscaping irrigation thus appeared slim. Unless the irrigation volume is extremely high ($>2 \times ET_0$), K_d for NDMA is exceptionally low (i.e., K_d approaches $0\,cm^3\,g^{-1}$) and/or the soil is extremely sandy (i.e., a loamy sand or lighter texture), the NDMA leaching potential in the reclaimed wastewater irrigated turf fields will not be significant.

5.3. Degradation and volatilization

The NDMA added through the reclaimed wastewater irrigation were essentially all lost through microbial degradation in the soil profile and volatilization at the soil atmosphere interface. The percentage of NDMA retained in the soil profile was under 5.6% in the all simulation scenarios. The relative proportion of the degradation and the volatilization loses were functions of the soil chemical properties and the hydraulic loading. In loamy sand soil, they varied from 36.9% to 75.4% and 23.7% to 61.9% for the degradation and volatilization, respectively. For the sandy loam soil, the rates of the degradation and volatilization accounted for 35.3–81.1% and 17.4–64.7%, respectively. Both of the biodegradation and the volatilization have significant roles in the NDMA disappearance in the reclaimed wastewater irrigated turf grass covered soils. Fig. 6 shows the relations between the degradation rates and the volatilization rates in the all simulation results. The contributions of microbial degradation and volatilization to the NDMA losses were linearly correlated ($r = -0.92$). The simulations demonstrated the interactive nature of the processes that the degradation and the volatilization complemented one another to sustain the high rates of NDMA removal in turf grass soils.

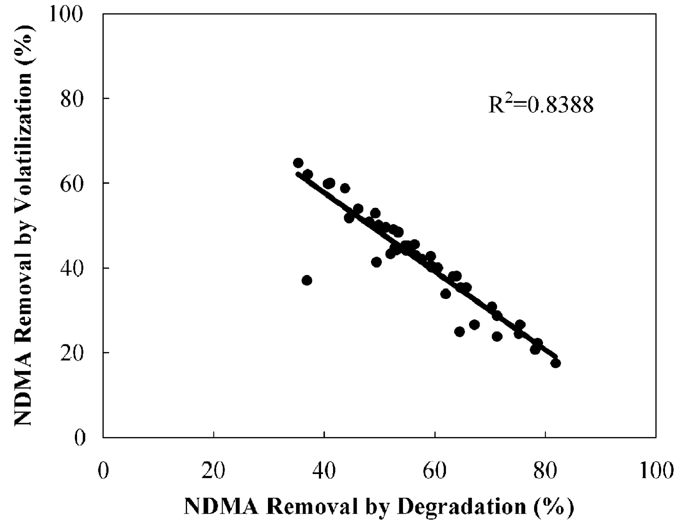

Fig. 6. Relationship between the removal by degradation and volatilization under different conditions in the numerical simulations conducted in this study.

6. Discussion

NDMA is miscible with water and is negligibly adsorbed to soil, which suggests that NDMA have a high-leaching potential. The weak adsorption of NDMA was confirmed by many researchers. For instance, Gunnison et al. (2000) observed that the K_d for NDMA in subsurface soil samples (sand, sandy loam, and loamy sand) ranged from 0.4 to $1.2\,L\,kg^{-1}$, and Yang et al. found the measured K_d for NDMA was only $0.45–0.64\,L\,kg^{-1}$ in landscape soils planted with turfgrass, groundcover, or trees. The study by Dean-Raymond and Alexander (1976) indicated that NDMA moved as fast as chloride when NDMA and chloride were leached through saturated soil columns. In the field situation, the soil remained unsaturated for most time. Therefore, the NDMA mobility may be reduced significantly. More important, the leaching loss of NDMA may be offset by other loss pathways such as degradation, volatilization at the soil surface, plant uptake of NDMA from soil.

The information regarding the plant uptake of NDMA from soil is limited. Dean-Raymond and Alexander (1976) found plants were able to take up approximately 5% of the applied NDMA, while the study by Arienzo et al. (2006) showed that the absorption of NDMA by turf grass was negligible. In model simulation, the plant uptake of NMDA was not considered. The contribution of plant uptake to the removal of NDMA should be further characterized.

Given its relatively low-boiling point (152 °C) and high-vapor pressure (2.7 mm Hg at 20 °C), volatilization could serve as an important loss pathway that helped to negate NDMA leaching in soils. NDMA has moderate to long persistence in soil, with half-lives in excess of 5 d (Tate and Alexander, 1975; Oliver et al., 1979; Kaplan and Kaplan, 1985; Gunnison et al., 2000; Yang et al., 2005). Customarily, these two processes were evaluated separately, resulting in different conclusions. For instance, Gunnison et al. (2000) and Yang et al. (2005) suggested that the microbial degradation was a significant factor for the disappearance of NDMA, while Oliver (1979), Arienzo et al. (2006), and Gan et al. (2006) reported that volatilization was the primary pathway for NDMA disappearance in the reclaimed wastewater irrigated fields. Our model simulation showed the microbial degradation and volatilization were negatively correlated (Fig. 6). Both of them can be dominant loss pathways of NDMA from soils.

The processes controlling the fate and transport of NDMA in the field conditions occur simultaneously and *in situ*. Thus, it is hard to tell the environmental risk of NDMA based on the experimental data on individual processes. By combing the experimental data with mathematical modeling, we successfully evaluated the leaching risks of NDMA under different field conditions. The information is useful to regulate the risk associated with NDMA in irrigation water. The method can be applied to other toxic organic compounds.

7. Conclusions

When reclaimed wastewater is used for landscaping irrigation, the risk is relatively low for NDMA to leach out of the surface soil profile and reach the ground water if the irrigation properly managed. The small amounts of NDMA added through the water applications will be lost through the microbial degradation in soils and the volatilization at the soil and atmosphere interface. However, the NDMA leaching risk may be dramatically increased when the hydraulic conductivity of the soil is high, the adsorption constant is low, and/or the irrigation intensity is high. To reduce the NDMA leaching risk, it is imperative that the fields be irrigated with the proper volume of water and attentions be paid to the soils having high-hydraulic conductivity and low-NDMA adsorption constants. In addition, the model simulations illustrated the kinetic of biodegradation have significant effect on the leaching potential of NDMA. The simulated leaching risk under first-order kinetics is much greater that that of zero order. Future research into the biodegradation kinetics of NDMA will be necessary to confirm the low levels of risk that are indicated in this research.

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