

Numerical Simulation and Sensitivity Analysis for Nitrogen Dynamics Under Sewage Water Irrigation with Organic Carbon

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Abstract This study is focused on investigating the impacts of organic carbon on the denitrification process of nitrogen transformation and transport. A numerical model, Nitrogen-2D, is modified by considering the impact of organic carbon in the denitrification equation. The modified model is used to simulate the soil nitrogen (including nitrate and ammonium) dynamics under the primary and secondary sewage water irrigation with different organic carbon concentrations. The simulated results of accumulated drainage water amount, soil nitrogen concentration, and nitrogen concentration in the drainage water show that the simulations and measurements are consistent. The comparison of results from the original and improved models shows the necessity to consider the impact of organic carbon. The nitrogen mass balance is calculated to analyze the nitrogen transformation processes quantitatively under different input organic carbon sources. Furthermore, the effect of different input organic carbon sources on the soil nitrogen dynamics is investigated by using the modified

Nitrogen-2D model with the calibrated parameters. The input organic carbon source helps to speed up the mineralization and denitrification, which contributes to the slight increase of ammonium concentration and the decrease of nitrate concentration in the shallow soil. Since a large number of soil water and nitrogen transformation and transport parameters are needed when setting up the model, a local sensitivity method is conducted to evaluate the input parameters by the sewage water irrigation case. The results show that the drainage water amount is very sensitive to the exponent n and the coefficient α of the soil water retention function and that the ammonium concentration is very sensitive to the first-order nitrification rate constant, the decomposition rate coefficient in humus pool, and the soil ammonium adsorption coefficient. The nitrate concentration is sensitive to more parameters, especially to the exponent n and the coefficient α in the soil water retention function and to the denitrification rate constant.

Keywords Nitrogen-2D · Organic carbon · Sewage water irrigation · Sensitivity analysis

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1 Introduction

With the threat of water shortage becoming more severe, conventional resources of potable water cannot meet irrigation demands in many countries, and treated or untreated sewage water has been increasingly used as an alternative irrigation water source (Bixio et al. 2006; Mackie et al. 2009). Sewage water supply is stable over

time, abundant in its amount, and rich of nutrient, such as nitrogen (N), phosphorus (P), and potassium (K) included. Therefore, irrigation with treated sewage water not only can relieve the water scarcity issue but also can increase agricultural productivity (Stevens et al. 2003). However, sewage water irrigation may cause negative environmental impacts, including potential nitrogen contamination to surface water and groundwater, salt accumulation in soils, and potential degradation of soil quality (Bedessem et al. 2005; Rocca et al. 2005). For example, it has been reported that dissolved nitrate in sewage water is highly mobile for transporting to groundwater and/or surface water, resulting in nitrate pollution (Jalali et al. 2008; Muyen et al. 2011). Therefore, understanding the transport and transformation processes of wastewater nutrients (mainly as nitrogen) in soils is essential for sustainable use of sewage water irrigation in agricultural areas. The understanding is also essential for estimating the influence of sewage water irrigation on crop production and soil quality. A significant amount of efforts has been carried out to better understand the complex processes of nitrogen transformation and transport in the unsaturated and saturated zones by conducting field investigation and numerical modeling to investigate the impacts of certain environmental factors and farming practices on nitrogen losses to the environment (Sophocleous et al. 2009).

In subsurface environmental systems, multiple nitrogen species exist and experience complicated transformation processes, including mineralization, immobilization, nitrification, denitrification, volatilization, and root uptake (Miller and Donahue 1995; Shrestha and Ladha 2002). Since direct measurements of these processes are difficult in the field conditions of sewage water irrigation, mathematical modeling has been used as an effective tool for managing irrigation and nitrogen application management especially during the crop growth period. A number of models have been developed to simulate water flow in unsaturated and saturated soils, nitrogen transport with complicated biological transformations, and crop uptake of water and nitrogen (Rahil and Antonopoulos 2007). Many models (e.g., Hydrus1D, SWAP, WAVE, SOILN, and WANISIM) consider the one-dimensional (1-D) vertical flow and solute transport in soil and focus on fertilizer application and nitrate leaching to groundwater systems (Rahil and Antonopoulos 2007; Lu et al. 2015; Sophocleous et al. 2009; Yang et al. 2008). Two-dimensional (2-D) models have been developed, such as DRAINMOD-N and

Nitrogen-2D, to simulate more complex hydrological conditions and nitrogen dynamics under irrigation and drainage conditions in agricultural fields (Bechtold et al. 2007; Yang et al. 2007, 2008).

Nitrogen transformation processes are significantly affected by soil physical and chemical conditions, such as soil moisture content, temperature, pH, and organic carbon (Echersten et al. 1996; Hansen et al. 1993). The effects of soil moisture and temperature on nitrogen transformation are usually considered in nitrogen models, but not including the organic carbon (Wu and McGechan 1998). More studies have shown that organic carbon concentration has significant impact on nitrogen mineralization, immobilization, nitrification, and denitrification. Organic carbon is of particular importance to denitrification, because organic carbon acts as an electron donor for denitrification to occur (van Rijn et al. 2006). Since denitrification is the only biogeochemical process that transforms nitrate to nitrogen gas to remove nitrate from the subsurface environmental system (Robertson et al. 2000; Su and Puls 2006), increasing denitrification rate under sewage water irrigation is an effective way to decrease the risk of nitrate contamination. Organic carbon was found to limit the denitrification rate in aquifers (Starr and Gillham 1993). As shown in Dodla et al. (2008), in comparison with soil pH and EC (electrical conductivity), total organic carbon (TOC) was the dominant factor controlling potential denitrification rate. In various field and laboratory studies carried out to investigate the nutrient removal efficiency with different sewage water types, infiltration rates, soil types, and crop cultivation methods (Lance et al. 1980; Marofi et al. 2015), it has been found that, when sufficient nitrate is available to induce denitrification, an increase in soil organic matter (SOM) increases soil denitrification rates (Bijay-Singh et al. 1988; Gale et al. 1993; Garcia-Montiel et al. 2003). Therefore, even using the primary effluent, nitrate concentration can still be reduced significantly if organic carbon concentration is high in the irrigated water (Lance et al. 1980; Cheng and Xu 2012). Thus, it is important to quantitatively estimate the impacts of organic carbon on nitrogen transformation especially on nitrate denitrification. The effect of organic carbon on denitrification can be calculated in CANDY (Franko et al. 1995), CERES (Godwin and Jones 1991), NITWAT (McIssac et al. 1993), SUNDIAL (Bradbury et al. 1993), WASMOD (Reiche 1994), WAVE (Vanclouster et al. 1996), and WHNSIM (Huwe and Totsche 1995) by expressing the denitrification rate

coefficient as a function of carbon degradation (Heinen 2006). However, few studies are carried out to estimate the detailed impact of input organic carbon on soil denitrification and the soil nitrate dynamics.

Since sewage water irrigation is an important organic carbon input sources, it will significantly impact the nitrogen transformation processes, especially denitrification. So, this study focuses on evaluating nitrogen dynamics under the impact of sewage water irrigation with high to relatively low organic carbon concentration. The nitrogen transformation and transport model, Nitrogen-2D (Yang et al. 2008; Sun et al. 2015), is used in the investigation to simulate soil nitrogen dynamics with sewage water irrigation. The model is improved to include a response term to simulate the impact of organic carbon on denitrification. The Nitrogen-2D model is calibrated and validated by the field experiment irrigated with primary and secondary sewage water of high and low organic carbon concentrations. Then four input organic carbon concentration scenarios are implemented to investigate the impacts on soil nitrogen transformation and transport. Since a relatively large number of parameters are involved in the simulation, sensitivity indices of soil hydraulic parameters and nitrogen transformation and transport parameters are calculated by using a local sensitivity analysis method.

2 Nitrogen-2D Model Development

Nitrogen-2D simulates nitrogen transformation and transport processes in the unsaturated-saturated zone (Lu 2004; Wang 2007; Yang et al. 2008). It can handle variable initial and boundary conditions related to irrigation and drainage practices in field conditions. The model is flexible to solve the governing equations of nitrogen transport in one dimension (the vertical direction) and two dimensions (the lateral and vertical directions). The current version of Nitrogen-2D simulates the nitrogen movement in the unsaturated-saturated zone driven by water flow and affected by soil moisture content, temperature, and pH. However, the current version does not consider the impact of organic carbon on nitrogen transport and transformation. To address this problem, in this study, we modified the model by adding an impact term of organic carbon for evaluating the denitrification rate.

The major nitrogen transformation and transport processes are shown in Fig. 1. Two organic matter pools are

considered as the litter pool and the humus pool. Organic nitrogen transport is not simulated in the model. The model evaluates transient organic nitrogen concentration in different nitrogen pools, each of which has its own transformation (e.g., mineralization/immobilization) rate and input nitrogen sources. The transport of two inorganic nitrogen species of nitrate and ammonium is simulated by the advection and dispersion equation (ADE) that includes the processes of nitrification, denitrification, volatilization, and root uptake as source/sink items. The model considers the major factors that affect each nitrogen transformation process. For example, the major factors affecting the denitrification process are soil moisture content (θ), temperature (T), and organic carbon concentration (C_c). All the major factors are listed in Fig. 1.

2.1 Governing Equation of Unsaturated-Saturated Water Flow

The two-dimensional Richards' equation in the x - z plane is used to describe the unsaturated-saturated water flow,

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x_i} \left[K \left(K_{ij}^A \frac{\partial h}{\partial x_j} + K_{iz}^A \right) \right] - S, \quad (1)$$

where θ is the soil moisture content [-]; t is the time [T]; x_i ($i=x$ and z) is the spatial coordinates [L]; h is the soil pressure head [L]; S is the root water uptake or other source/sink item [T^{-1}]; K is the unsaturated hydraulic conductivity [$L T^{-1}$]; and K_{ij}^A ($i, j=x, z$) are the components of the dimensionless anisotropy tensor for the unsaturated hydraulic conductivity [-].

The van Genuchten (VG) model (van Genuchten 1980) is adopted to describe the soil water characteristics and coupled with the unsaturated hydraulic conductivity to simulate soil water flow. The relationship of h - θ and h - K can be written as,

$$\theta(h) = \begin{cases} \theta_a + \frac{\theta_m - \theta_a}{(1 + |\alpha h|^n)^m} & h < h_s, \\ \theta_s & h \geq h_s \end{cases}, \quad (2)$$

$$K(h) = \begin{cases} K_s K_r(h) & h \leq h_k \\ K_k + \frac{(h-h_k)(K_s-K_k)}{h_s-h_k} & h_k < h < h_s, \\ K_s & h \geq h_s \end{cases} \quad (3)$$

where $\theta(h)$ and $K(h)$ are the volumetric content and hydraulic conductivity corresponding to h ; θ_m is the

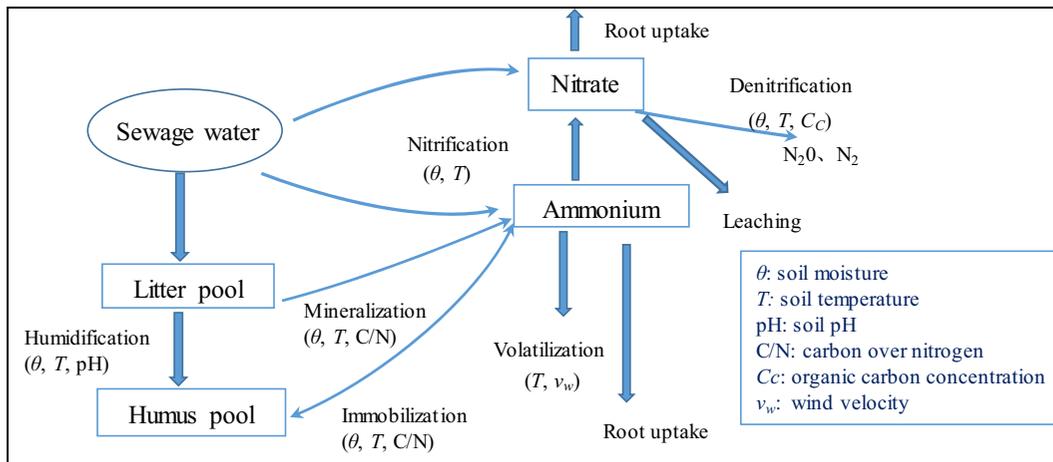


Fig. 1 Nitrogen transformation and transport processes and their specific major impact factors

fictitious saturated water content in soil water retention function [-]; θ_r is the residual water content [-]; θ_a is the parameter in the soil water retention function [-]; θ_s is the saturated water content [-]; K_s is the saturated hydraulic conductivity [$L T^{-1}$]; K_r is the relative hydraulic conductivity [$L T^{-1}$]; K_k is the measured value of the unsaturated hydraulic conductivity corresponding to h_k [$L T^{-1}$]; h_s is the saturated pressure head [L]; h_k is a pressure head less than or equal to h_s [L]; α is the coefficient in the soil water retention function [L^{-1}]; and m and n are the exponents in the soil water retention function [-].

2.2 Governing Equation of Nitrogen Transport

While ammonium can be adsorbed on soil particles but nitrate is easily mobile, the ADEs controlling ammonium and nitrate transport in the soil have different forms as follows,

$$\frac{\partial \theta C_{N,4}}{\partial t} + \frac{\partial \rho S_{N,4}}{\partial t} = \frac{\partial}{\partial x_i} \left(\theta D_{ij} \frac{\partial C_{N,4}}{\partial x_j} \right) - \frac{\partial q_i C_{N,4}}{\partial x_i} + R_4, \quad (4)$$

$$\frac{\partial \theta C_{N,3}}{\partial t} = \frac{\partial}{\partial x_i} \left(\theta D_{ij} \frac{\partial C_{N,3}}{\partial x_j} \right) - \frac{\partial q_i C_{N,3}}{\partial x_i} + R_3, \quad (5)$$

where $C_{N,4}$ and $C_{N,3}$ are the dissolved ammonium and nitrate concentrations [ML^{-3}]; q_i ($i = x, z$) is the Darcy flux [$L T^{-1}$]; ρ is the soil bulk density [ML^{-3}]; $S_{N,4}$ is the adsorbed ammonium concentration on soil particles, which is linearly expressed by the empirical coefficient K_d multiplying with the dissolved ammonium concentration $C_{N,4}$ [$M L^{-3}$]; D_{ij} is the component of the

dispersion coefficient tensor which combined the diffusion of nitrogen in soil and the hydrodynamic dispersion resulting from the variation of pore water velocity [$L^2 T^{-1}$]; and R_3 and R_4 are the source/sink terms related to nitrate and ammonium [$M L^{-3} T^{-1}$]. The source/sink terms can be expressed as,

$$R_3 = R_{m3} + R_n + R_{dn} + R_{p3}, \quad (6)$$

$$R_4 = R_{m4} + R_n + R_v + R_{p4}, \quad (7)$$

where R_{m3} is the net immobilization from nitrate [$M L^{-3} T^{-1}$]; R_n is the nitrification term [$M L^{-3} T^{-1}$]; R_{dn} is the denitrification term [$M L^{-3} T^{-1}$]; R_{p3} and R_{p4} are the root uptake of nitrate and ammonium [$M L^{-3} T^{-1}$]; R_{m4} is the net mineralization/immobilization to/from ammonium [$M L^{-3} T^{-1}$]; and R_v is the ammonium volatilization term [$M L^{-3} T^{-1}$].

2.3 Nitrogen Transformation Processes

2.3.1 Mineralization/Immobilization

The zero-order kinetics is adopted to describe the mineralization/immobilization processes,

$$\frac{\partial C_N}{\partial t} = -K_0 \times (\theta + \rho K_d) \times f, \quad (8)$$

where C_N is the nitrogen concentration of the soil solution [ML^{-3}]; K_0 is the zero-order rate constant [T^{-1}]; K_d is the distribution coefficient for adsorbing solute [$L^3 M^{-1}$], which is zero for nitrate; and f is the comprehensive response function from the different factors

such as soil moisture, temperature, carbon over nitrogen ratio, and soil pH value [-].

2.3.2 Nitrification Process

The first-order kinetics is used to describe the nitrification process,

$$\frac{\partial C_{N,4}}{\partial t} = K_n(\theta + \rho K_d) f_\theta f_T C_{N,4}, \quad (9)$$

where $C_{N,4}$ is the ammonium concentration in the soil solution [$M L^{-3}$]; K_n is the first-order nitrification rate constant [T^{-1}]; and f_θ , and f_T are the response functions of the soil water moisture and soil temperature.

2.3.3 Volatilization Process

An empirical calculation of volatilization rate is used as the upper boundary condition (Zhu et al. 2009),

$$R_v = -K_v(C_{S4} - C_A), \quad (10)$$

where R_v is the volatilization flux from the upper boundary [$M L^{-3} T^{-1}$]; C_{S4} is the soil surface ammonium concentration [$M L^{-3}$]; and C_A is the atmospheric ammonia concentration which is assumed to be zero [$M L^{-3}$]. K_v is the mass transfer coefficient in the liquid [T^{-1}] given by,

$$K_v = 48.4 \times v^{0.8} \times T^{-1.4}, \quad (11)$$

where v is the wind speed [$L T^{-1}$] and T is the air temperature [Θ]. The empirical equation is well-tested to calculate the volatilization under sewage water irrigation condition as shown in Zhu et al. (2009).

2.3.4 Denitrification Process

The first-order kinetics is used to describe the denitrification process by adding the impact of the organic carbon as follows (Heinen 2006),

$$\frac{\partial C_{N,3}}{\partial t} = -K_{dn} \theta f_\theta f_T C_C C_{N,3}, \quad (12)$$

where $C_{N,3}$ is the nitrate concentration in the soil solution [$M L^{-3}$]; K_{dn} is the denitrification coefficient when considering the soil organic carbon concentration [$M^{-1} L^3 T^{-1}$]; f_θ , and f_T are the response functions of the soil water moisture and soil temperature; and C_C is the organic carbon concentration [$M L^{-3}$].

As shown in Eqs. (8), (9), and (12), soil moisture and soil temperature are two major important factors that affect the nitrogen transformation processes. While soil moisture can be calculated by solving Eq. (1), soil temperature needs to be calculated via (Rijtema and Kroes 1991),

$$T(z, t) = T_a + A_0 \exp(-z/D_m) \cos(\omega t + \phi - z/D_m), \quad (13)$$

where $T(z, t)$ is the temperature [Θ]; T_a is the average yearly temperature [Θ]; A_0 is the amplitude of temperature wave [Θ]; D_m is the damping depth [L]; ω is the frequency of temperature wave [T^{-1}]; and ϕ is the phase shift [-].

3 Simulation of Nitrogen Dynamics under Sewage Water Irrigation

3.1 Sewage Water Irrigation Experiment

The experiment was carried out in six vertical columns. Three of them were irrigated with primary sewage water and the other three with secondary sewage water. The primary and secondary sewage waters were obtained from a wastewater treatment plant in Beijing. The columns had the same size of 120 cm in height and 30 cm in diameter, and each had a drainage hole at the bottom to control drainage. The experiment scheme was shown in Fig. 2. The columns were exposed in the natural climate condition, except during the experiment period from May 13, 2008 to October 5, 2008, when the columns were isolated from precipitation by a rain shelter. A total of 17 irrigation events happened in the whole experiment period, with 7–10-day interval between two irrigation events. The irrigation rate, ammonium concentration, nitrate concentration, and total nitrogen concentration in the primary and secondary sewage irrigation water were measured, and the measurements are plotted in Fig. 3. The chemical oxygen demand (COD) and TOC concentration in the secondary sewage water were measured, and the measurements were used to obtain a relation function between the two variables (Fig. 4). The function is used in this study to calculate TOC based on COD for both primary and secondary sewage water.

The soil ammonium, nitrate, and total nitrogen concentrations at the depth of 10, 40, and 70 cm of the three columns were measured after each irrigation event. The

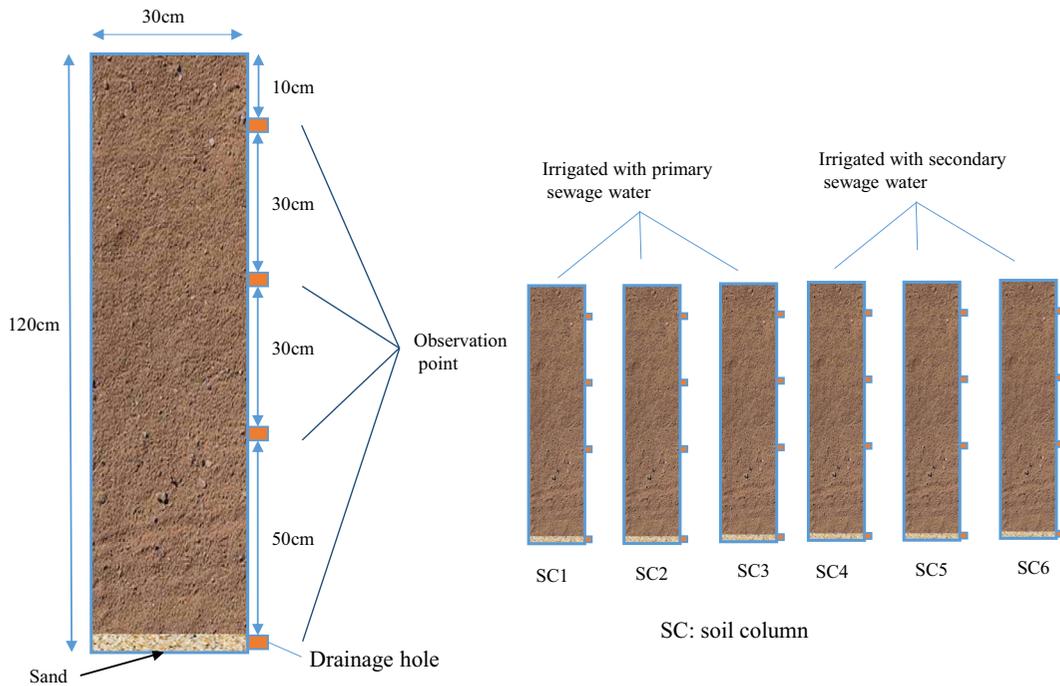


Fig. 2 The experiment scheme

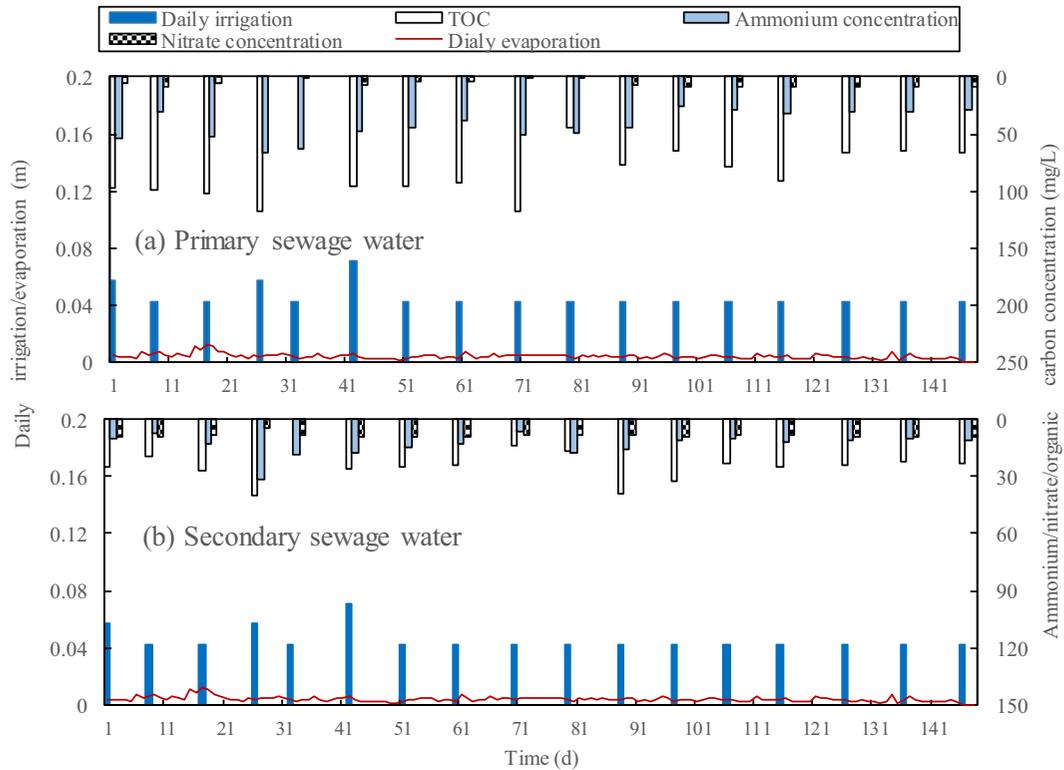


Fig. 3 Irrigation and soil evaporation rates as well as the ammonium, nitrate, and organic carbon concentrations in the primary and secondary sewage water

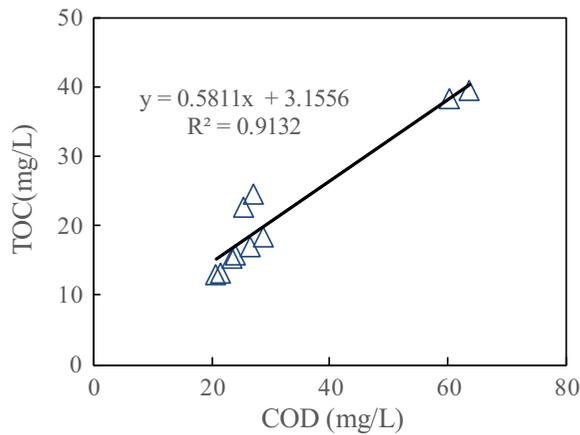


Fig. 4 The relationship between COD and TOC in the secondary sewage water

drainage hole at the bottom of each column was opened after 72 h of irrigation to drain water. The amount of drainage water and the concentrations of nitrate, ammonium, and total nitrogen in the drainage water (at the depth of 120 cm) were measured. The daily average air temperature available in the China Meteorological Data Sharing Service System is used as the temperature at the ground surface ($z = 0$ m) to calibrate the parameters in Eq. (13). The calibrated parameter values are that, $T_a = 14$ °C; $A_o = 15$ °C; $D_m = 1000$ m; $\omega = 0.0172$ day⁻¹; and $\phi = 5.228$.

The experiments with the primary sewage water were used to calibrate the model parameters, and the

experiments with the secondary sewage water were used to validate the model. The soil water characteristic parameters were calibrated by using the amount of average drainage water over the three columns, and the nitrogen transformation and transport parameters were calibrated by using the average soil nitrogen concentration over the three soil columns.

3.2 Model Calibration Results

As shown in Fig. 5, there is a good agreement between the simulated and measured drainage water accumulated over the modeling period, and the simulated accumulation process is highly consistent with the irrigation events. The soil water characteristic parameters are as follows: $\theta_s = 0.42$, $\alpha = 4.0$ m⁻¹, $n = 1.28$, $K_s = 1.3$ m/day, and $\theta_r = 0.02$. Figures 6 and 7 show the simulated and measured ammonium and nitrate concentration profiles at different times. The model simulations are consistent with the measurements (average measurements of three soil columns) in the profiles. The simulated nitrate concentration agrees well with the measurements below the depth of 40 cm with the root mean squared error (*RMSE*) ranging from 0.56 to 0.84 mg/L, while relatively large derivations are found near the ground surface with the *RMSE* of 4.44 to 18.58 mg/L, as listed in Table 1. The *RMSE* of the simulated and measured nitrate concentration for all samples is 9.63 mg/L, and the mean error (*ME*) is 4.59 mg/L. The simulated

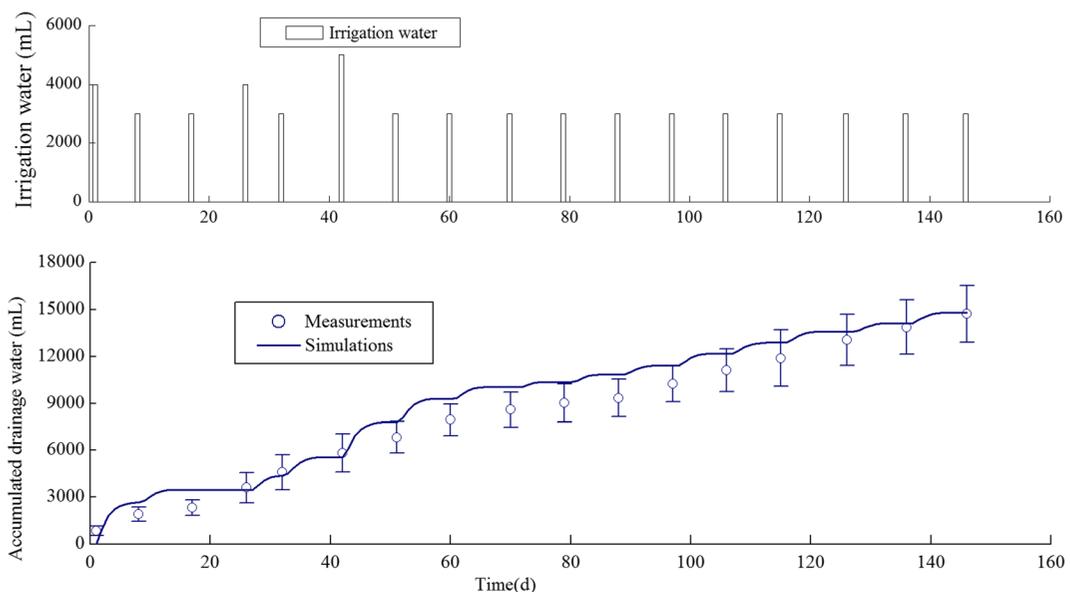


Fig. 5 Simulated and measured drainage water accumulation

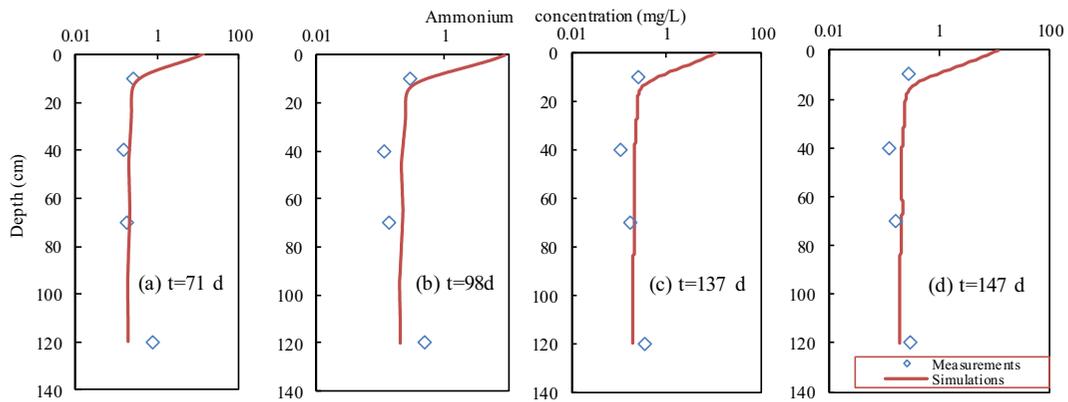


Fig. 6 Simulated and measured ammonium concentration profiles at different times

ammonium concentration agrees well with the measurements with the *RMSE* ranging from 0.078 to 0.48 mg/L, and *ME* ranging from 0.032 to 0.39 mg/L. Relative large derivations are found near the ground surface with the *RMSE* of 0.48 mg/L and at the bottom 0.37 mg/L, as listed in Table 1. The *RMSE* of the simulated and measured ammonium concentration is 0.31 mg/L, and the *ME* is 0.21 mg/L.

Figure 8 plots temporal variation of the simulated and measured ammonium concentration during the experiment season at the depths of 10, 40, 70, and 120 cm. The maximum, minimum, and average measured ammonium concentrations of three soil columns are plotted. At the depths of 40, 70, and 120 cm, while the simulated ammonium concentrations are smoother than the measured concentrations, the simulations are of the same magnitude of the measurements. At the depth of 10 cm,

the temporal variation of simulated ammonium concentration agrees with the change of irrigation events. There is an abrupt increase of the simulations from 26 to 42 days, because of the larger ammonium concentration in the sewage water on day 26, day 34, and day 42 than on other days. The large variation of measured ammonium concentration is observed after the third irrigation event. Figure 9 plots the temporal variation of simulated and measured nitrate concentrations during the experiment season at the depths of 10, 40, 70, and 120 cm. The maximum, minimum, and average measured nitrate concentrations of three soil columns are plotted. The simulations have good agreements with the measurements at the depths of 40, 70, and 120 cm, while the simulations are larger than the measurements in the shallow soil layer at the depth of 10 cm. Similar to the simulated ammonium concentration at the depth of

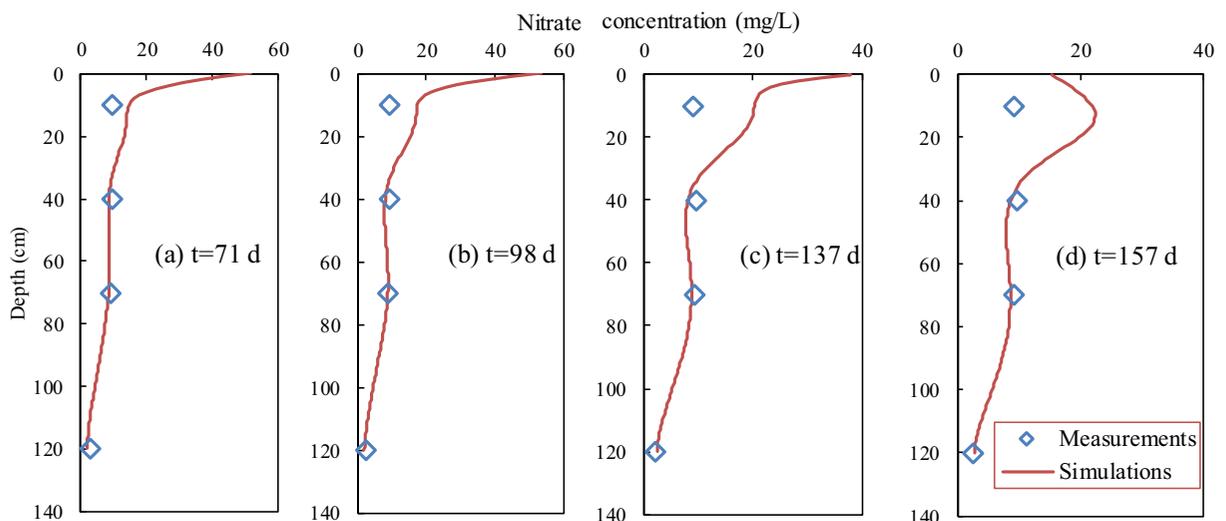


Fig. 7 Simulated and measured nitrate concentration profiles at different times

Table 1 The measured and simulated values, and the *ME* and *RMSE* of the simulated and measured nitrogen concentration in the model calibration

Depth (cm)	Sample	Nitrate (mg/L)				Ammonium (mg/L)			
		Measurement	Simulation	<i>ME</i>	<i>RMSE</i>	Measurement	Simulation	<i>ME</i>	<i>RMSE</i>
10	17	9.15	23.02	13.87	18.58	0.26	0.65	0.39	0.48
40	17	9.32	10.60	1.28	4.44	0.13	0.21	0.085	0.107
70	17	8.86	8.60	-0.26	0.56	0.19	0.22	0.032	0.078
120	16	2.69	2.34	-0.35	0.84	0.50	0.25	-0.25	0.37
Total	67	7.58	11.27	3.70	9.63	0.27	0.34	0.07	0.31

Note: *ME* means the mean error, $ME = \frac{1}{n} \sum_{i=1}^n (Y_{i\text{mod}} - Y_{i\text{obs}})$; *RMSE* is the root mean square error, $RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (Y_{i\text{mod}} - Y_{i\text{obs}})^2}$; *n* is the sample number; $Y_{i\text{mod}}$ is the simulated value; and $Y_{i\text{obs}}$ is the observed value

10 cm, the simulated nitrate concentration has peaks related to the irrigation events, whereas these peaks are not observed in the measurements. The reason maybe that the measured samples were taken 3 days after the irrigation event, while would result in missing the nitrate concentration peak. Figure 9a shows three sharp

increases in the simulated nitrate concentration from 26 to 42 days. The reason is that the organic carbon in the soil profile has the increasing trend with the input source from the sewage water and then becomes steady. The denitrification is smaller before the organic carbon being steady than after being steady. Also, the

Fig. 8 Simulated and measured ammonium concentration over time at the depths of (a) 10 cm, (b) 40 cm, (c) 70 cm, and (d) 120 cm

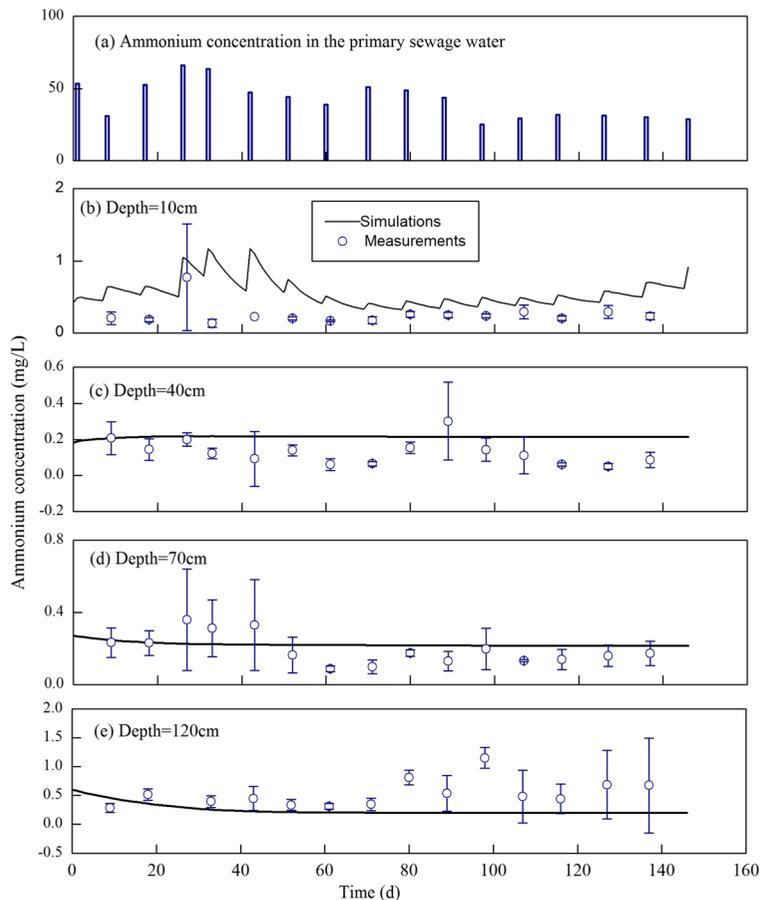
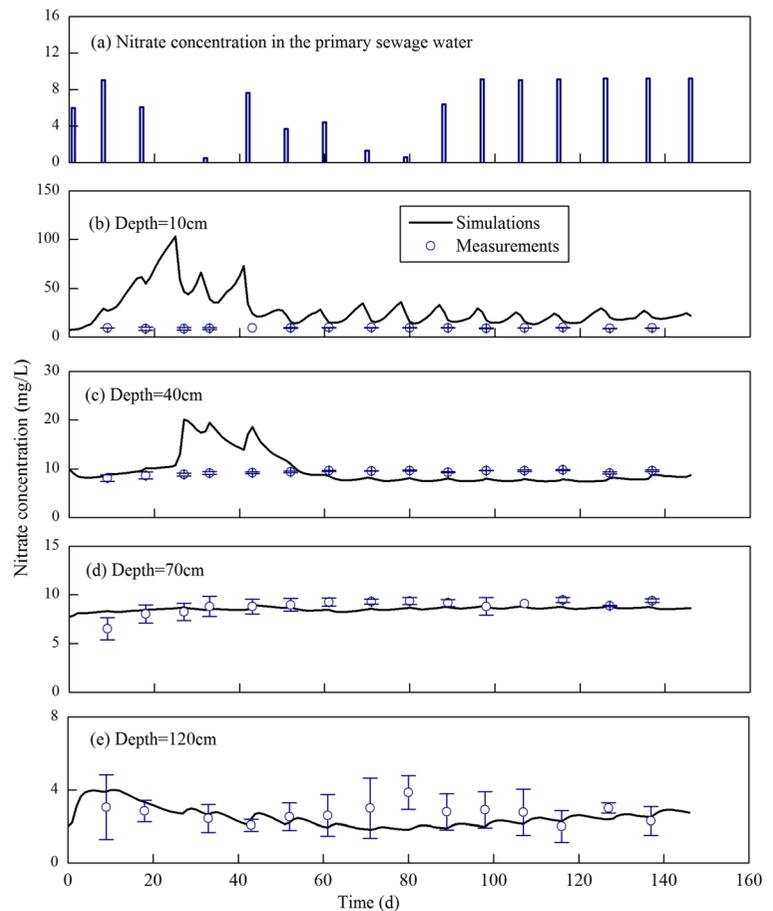


Fig. 9 The simulated and measured nitrate concentration over time at the depths of **a** 10, **b** 40, **c** 70, and **d** 120 cm



denitrification rate is slow and the nitrification rate is fast due to the low soil moisture at the top soil. The increasing trend at the depth of 40 cm is caused by the nitrate movement from the upper soil system. The impact disappears at the deeper depth below 40 cm. In general, the simulations are consistent with the measurements. The nitrogen transformation and transport parameters used herein are shown in Table 2.

The mass balance analysis is shown in Figs. 10a, b for nitrate and ammonium, respectively. The relative errors of the ammonium and nitrate mass balance are 0.21 and 5.15%, respectively, which is satisfactory. The input ammonium mass from the sewage water is the major source for the simulated system. Denitrification and nitrification are the two major transformation processes to control the nitrogen transport in the system. The ammonium and nitrate in the drainage water are very small amount contributing to the nitrogen loss from the system. Only denitrification removes nitrogen in the soil. The amounts of nitrate and ammonium in the soil at

the end of the simulation period are larger than those at the initial time, which indicate the nitrogen contamination risk under this intensive sewage water irrigation.

Table 2 The soil nitrogen transformation and transport parameters

Parameter	Unit	Value
ρ	kg m^{-3}	$1.57 * 10^3$
α_L	m	0.07
α_T	m	0.01
K_d (ammonium)	$\text{kg}^{-1} \text{m}^3$	4.0
Zero-order coefficient of litter pool (K_0) _l	/	0.0045
Zero-order rate coefficient of humus pool (K_0) _h	/	0.00006
C/N ratio in the humus pool	/	10
Denitrification coefficient K_{dn}	$\text{kg}^{-1} \text{m}^3 \text{day}^{-1}$	0.004
First-order nitrification rate K_n	day^{-1}	0.06

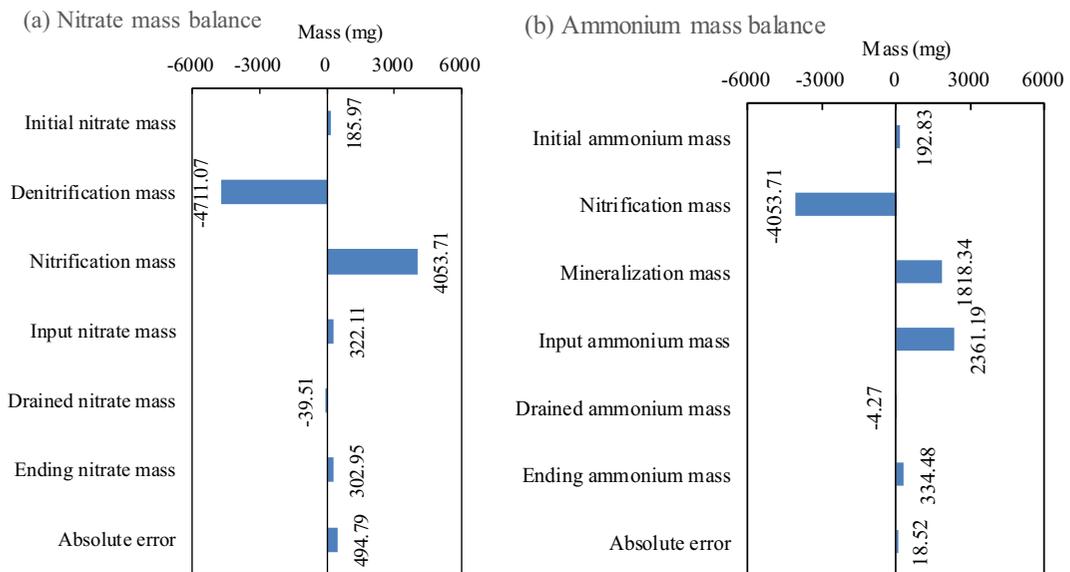


Fig. 10 The mass balance for **a** nitrate and **b** ammonium. The positive sign (+) means the nitrogen input, and the negative sign (-) means the nitrogen removal

3.3 Model Validation Results

The calibrated model parameters are validated by simulating the soil nitrogen concentration measured during the irrigation events using the secondary sewage water. The simulated and measured drainage water, spatial

variation of nitrogen concentrations, and temporal variation of nitrogen concentrations are shown in Figs. 11, 12 and 13, respectively. The figures show that the calibrated parameters can yield satisfactory results for simulating drainage water and nitrogen concentrations irrigated with the secondary sewage water. The *RMSE*

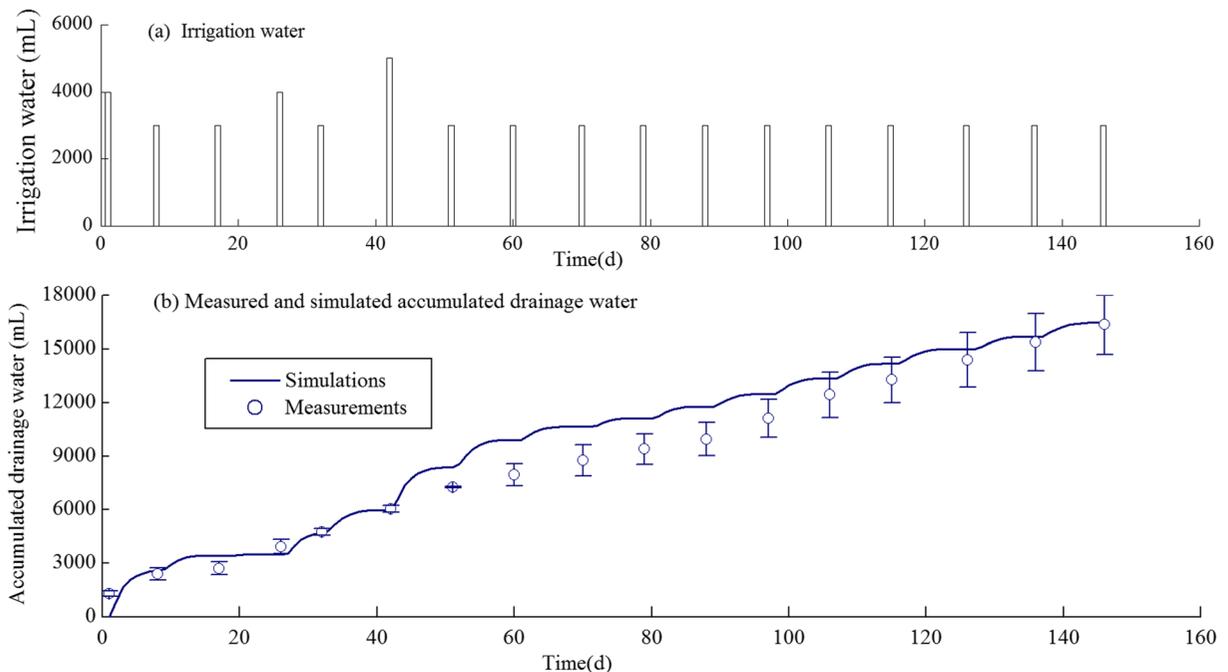
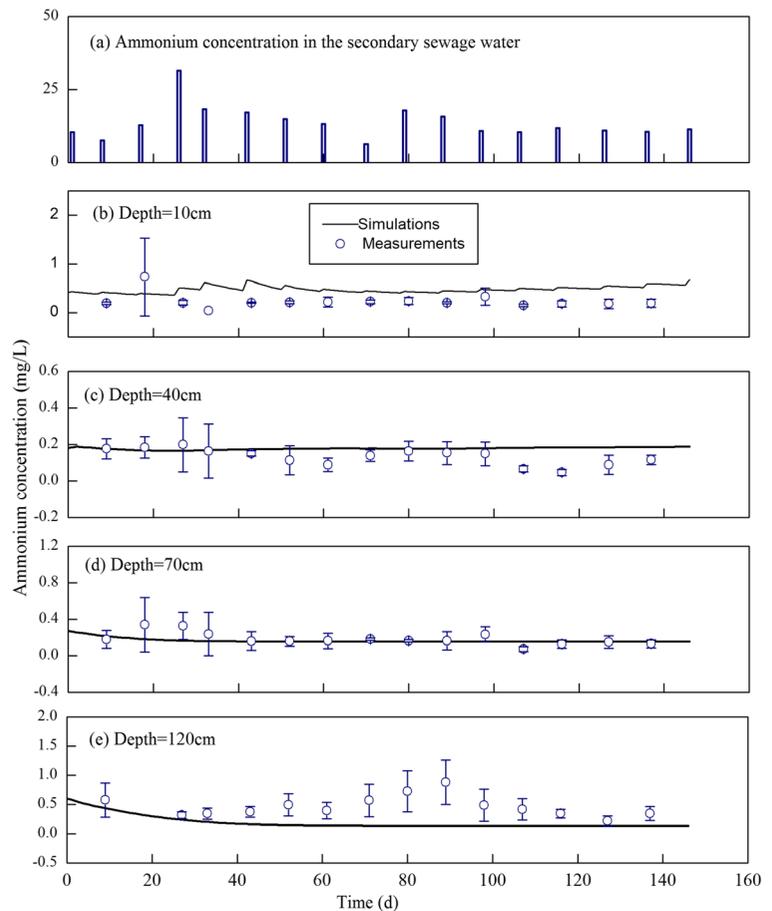


Fig. 11 The simulated and measured drainage water accumulation process with the secondary sewage water irrigation

Fig. 12 The simulated and measured ammonium concentration over time at the depths of **a** 10, **b** 40, **c** 70, and **d** 120 cm



values of the simulated and measured ammonium concentration are 0.061 to 0.33 mg/L at the four depths, and it is 0.23 mg/L for all samples, as listed in Table 3. The *ME* values range from -0.015 to 0.27 mg/L, and it is 0.17 mg/L for all samples. For nitrate concentration, the *RMSE* values range from 0.7 to 14.71 mg/L at the four depths, and 7.66 mg/L for all samples. The *ME* values are -1.97 to 13.79 mg/L, and it is 4.89 mg/L for all samples. The largest errors for both nitrate and ammonium occur at the top soil.

3.4 Model Discussion

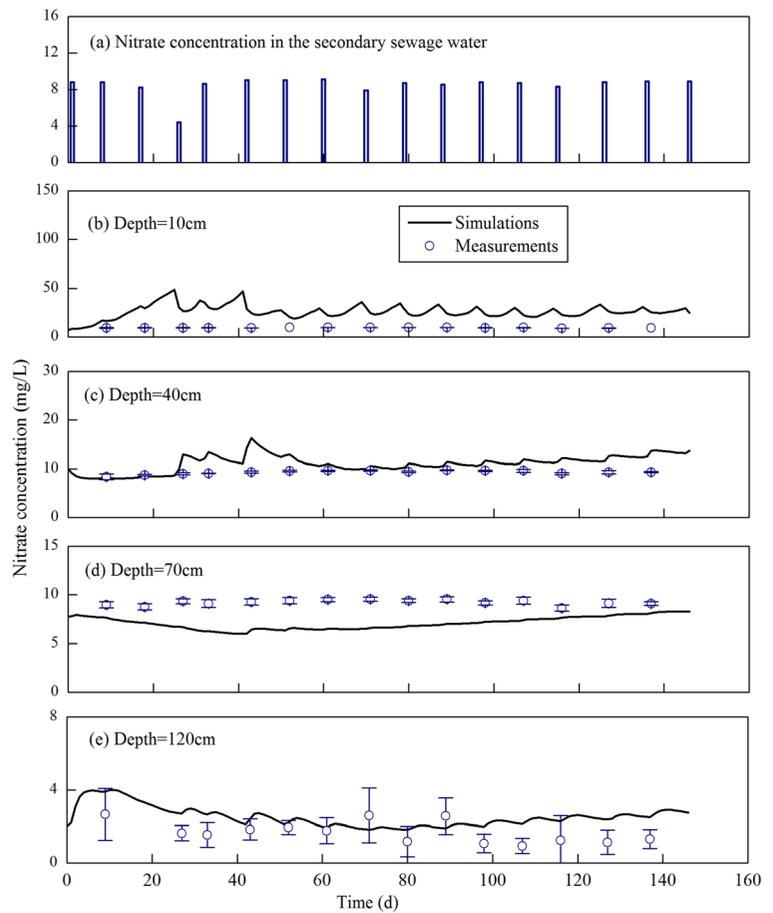
In order to compare the simulation accuracy by the original model and the improved model by adding the impact term of organic carbon, the two models are used to simulate the soil nitrogen concentration with the primary sewage water irrigation. Figure 14 shows the simulated and measured concentration profiles of ammonium and nitrate by the original Nitrogen-2D model

and the improved model. The results show that the organic carbon effect function does not greatly impact the simulated soil ammonium concentration. The *RMSE* of the simulated ammonium concentration only decreases from 0.34 to 0.31 mg/L when using the improved model, and the *ME* decreases from 0.24 to 0.21 mg/L. However, the simulated soil nitrate concentration has been improved significantly by the improved Nitrogen-2D model. The *RMSE* of the improved model is 9.63 mg/L, and the *ME* is 4.59 mg/L, while the corresponding values for the original model are 35.59 and 21.56 mg/L. Therefore, adding the impact term of organic carbon in the nitrogen model can make the model more reasonable.

4 Numerical Simulation Experiments

The calibrated model is used to simulate the nitrogen dynamics under four different scenarios of organic

Fig. 13 The simulated and measured nitrate concentration over time at the depths of **a** 10, **b** 40, **c** 70, and **d** 120 cm



carbon concentration in the sewage water. The simulation used the calibrated parameter values of soil water flow and nitrogen transformation and transport. The organic carbon concentrations of the four scenarios are 0.25, 0.5, 1.0, and 2.0 times of the primary sewage water simulation. The spatial and temporal variation of simulated nitrate and ammonium are shown to analyze the nitrogen dynamics under the impact of organic carbon.

4.1 Simulation Results Under the Four Scenarios of Organic Carbon Concentration

Figure 15 shows the simulated ammonium concentration profiles under the four scenarios of organic carbon concentration in the sewage water at different times. The figure shows that the higher organic carbon concentration slightly increases the ammonium concentration in

Table 3 The observed and simulated values, and the *ME* and *RMSE* of the simulated and measured nitrogen concentration in the model validation

Depth (cm)	Sample	Nitrate (mg/L)				Ammonium (mg/L)			
		Measurement	Simulation	<i>ME</i>	<i>RMSE</i>	Measurement	Simulation	<i>ME</i>	<i>RMSE</i>
10	17	9.40	23.18	13.79	14.71	0.23	0.51	0.27	0.33
40	17	9.22	11.89	2.67	3.21	0.14	0.18	0.041	0.061
70	17	9.14	7.17	-1.97	2.15	0.18	0.17	-0.015	0.069
120	16	1.66	1.87	0.20	0.70	0.45	0.20	-0.25	0.33
Total	67	7.44	11.16	3.72	7.66	0.25	0.26	0.02	0.23

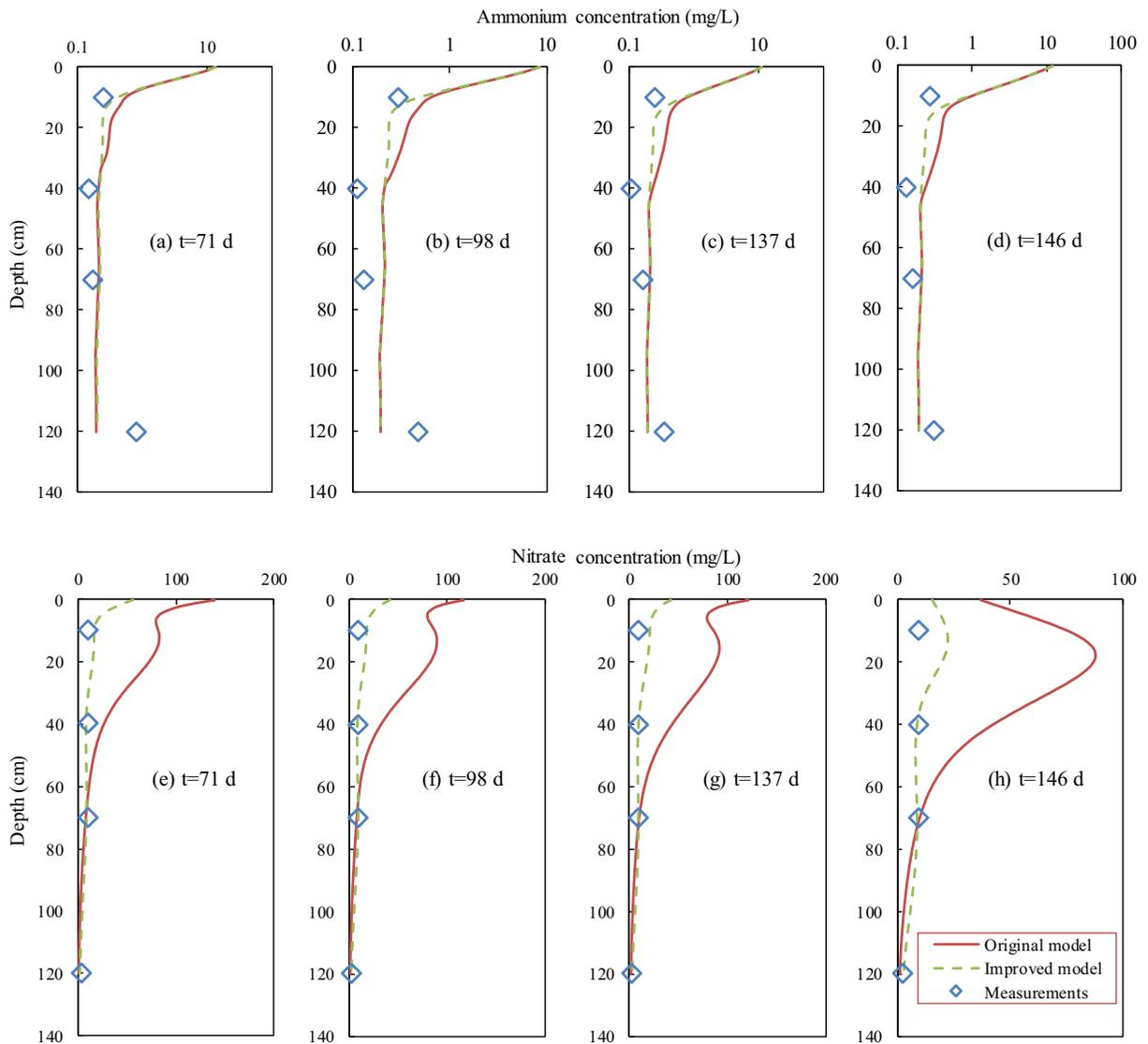


Fig. 14 Simulated and measured ammonium and nitrate concentration profiles at different times by the original and improved Nitrogen-2D model

the soil profile. The reason is that, the input organic carbon sources speeds up the mineralization process, which contributes to the increase of ammonium concentration. In addition, other studies found that adding organic carbon can decrease nitrification rates, which also leads to the increase of ammonium concentration (Strauss and Lamberti 2000; Zhu and Chen 2001). Figure 16 shows the simulated nitrate concentration profiles under the four scenarios of organic carbon concentration in the sewage water at different times. The figure shows that higher organic carbon concentration causes the decrease of the nitrate concentration in the soil profile

above the depth of 60 cm. The reason why denitrification rate is different at different depths is related to the organic carbon distribution in the soil profile. The input organic carbon is mainly distributed in the shallow soil profile, and higher organic carbon speeds up the denitrification rate and decreases the nitrate concentration.

Figures 17 and 18 show the temporal variation of ammonium and nitrate concentration at different depths under the four scenarios of organic carbon concentrations in the sewage water. Figure 17 shows that increasing organic carbon concentration in the sewage water has negligible impact on the ammonium concentration

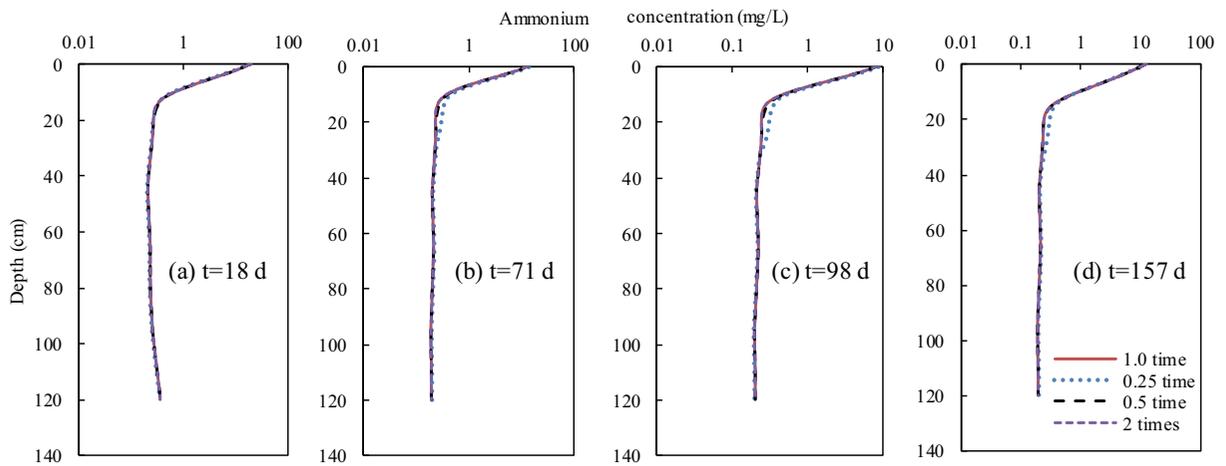


Fig. 15 Simulated ammonium concentration profiles under the four scenarios of organic carbon concentration in the sewage water at different times

over the entire simulation period. However, the nitrate concentration has been greatly affected by the organic carbon concentrations of the four scenarios, as shown in Fig. 18. High nitrate concentrations are resulted when the organic carbon concentration decreases in the sewage water at different depths especially above 70 cm. The lower organic carbon concentration corresponds to the larger nitrate concentration value.

4.2 Mass Balance Analysis Under Four Scenarios of Organic Carbon Concentration

Table 4 lists the mass balance data of ammonium and nitrate under the four scenarios of organic carbon

concentration in the sewage water. The drained nitrate mass decreases with the increase of organic carbon concentration in the sewage water. Similarly, the nitrate mass in the soil profile at the end of the simulation decreases with the increase of organic carbon concentration in the sewage water. As the increase of organic carbon concentration in the sewage water, the drained nitrate mass decreases from 40.93 to 37.92 mg, and the ending nitrate mass decreases from 732.64 to 189.79 mg, which is close to the initial nitrate mass in the profile. These results indicate that increasing organic carbon concentration in the sewage water can decrease the nitrate contamination risk in soils. However, increasing organic carbon concentration has negligible impact

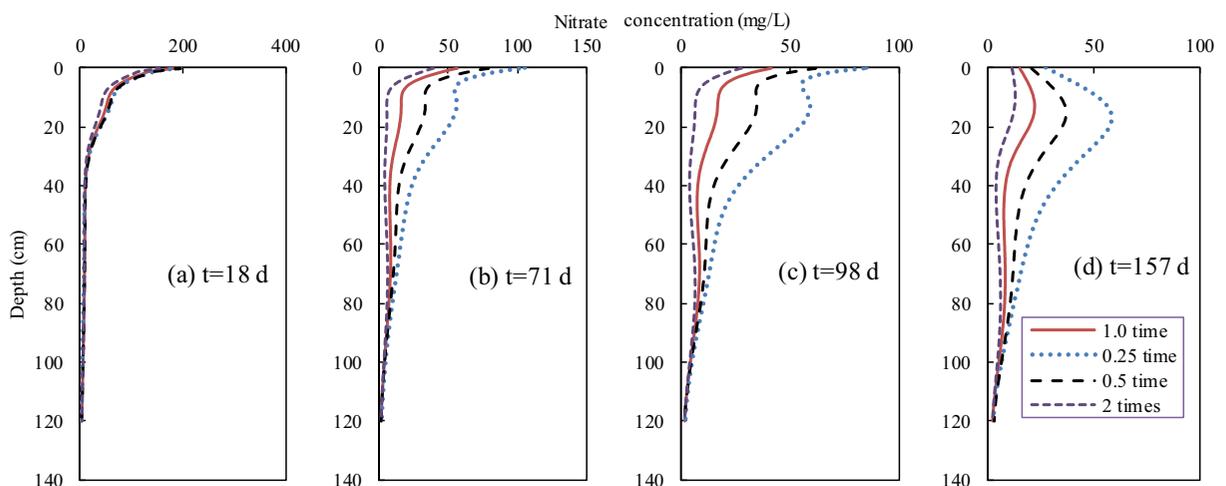
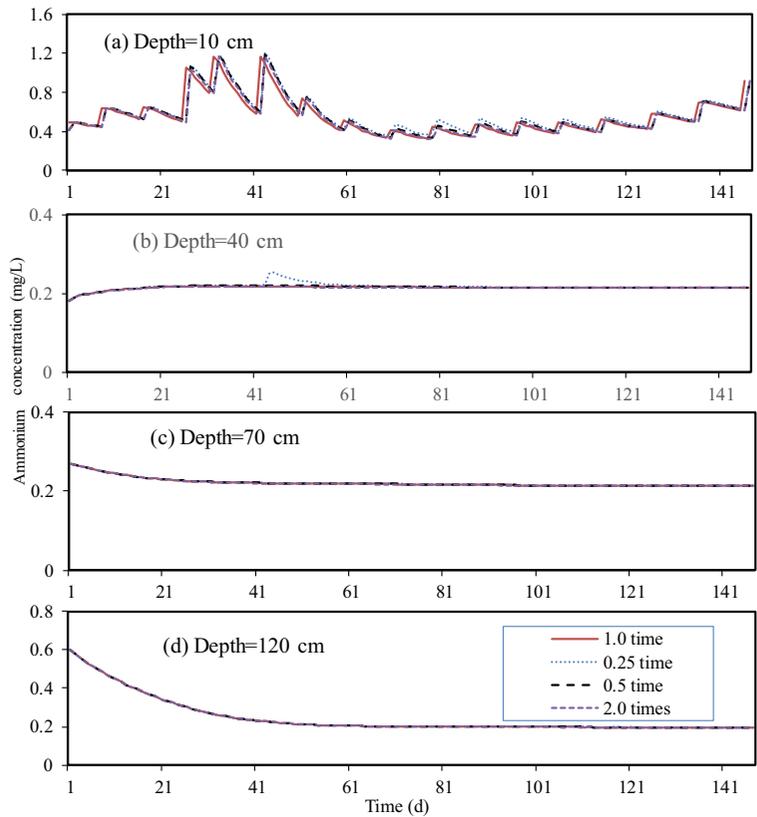


Fig. 16 Simulated nitrate concentration profiles under the four scenarios of organic carbon concentration in the sewage water at different times

Fig. 17 The simulated ammonium concentration trends over time under the four scenarios of organic carbon concentration in the sewage water at the depths of 10, 40, 70, and 120 cm



on the drained ammonium mass and the ammonium mass in soil at the end of the simulation period.

5 Sensitivity Analysis

The model includes 10 major parameters ($K_s, \alpha, n, \alpha_L, \alpha_T, (K_0)_l, (K_0)_h, K_{dn}, K_n, K_d$) related to soil water retention and nitrogen transformation and transport. From the original dataset obtained from calibration, different simulations are conducted for the parameters within the limitation of the database. Base on the simulation, the local sensitivity analysis is performed to assess how the parameters affect model outputs, including accumulated drainage water amount, nitrate, and ammonium concentrations in soil profiles and drainage water.

5.1 Sensitivity Analysis Method

The local sensitivity analysis method is used in this study. Assume a model, f ,

$$o = f(\beta_1, \beta_1, \dots, \beta_n), \tag{14}$$

where o is the model output, and β_i is a parameter of model f . The variation of the output value caused by the variation of a model parameter is evaluated as,

$$\begin{aligned} \delta_i &= \frac{\Delta o}{\Delta \beta_i} \\ &= \frac{f(\beta_1, \dots, \beta_{i-1}, \beta_i + \Delta \beta_i, \beta_{i+1}, \dots, \beta_n) - f(\beta_1, \dots, \beta_{i-1}, \beta_i, \beta_{i+1}, \dots, \beta_n)}{\Delta \beta_i}, \end{aligned} \tag{15}$$

where δ_i is the change of model output variable caused by the variation $\Delta \beta_i$ of parameter β_i . In order to compare the sensitivity of different parameters, the normalized sensitivity index (SI) is calculated as,

$$SI_i = \frac{\Delta o}{\Delta \beta_i} \frac{\beta_i}{o}. \tag{16}$$

The sensitivity index can be classified to five levels listed in Table 5 for evaluating relative sensitivity of the parameters (Oliver and Christakos 1996; Hamby 1994; Castillo et al. 2004).

Fig. 18 Simulated nitrate concentration trends over time under the four scenarios of organic carbon concentration in the sewage water at the depths of 10, 40, 70, and 120 cm

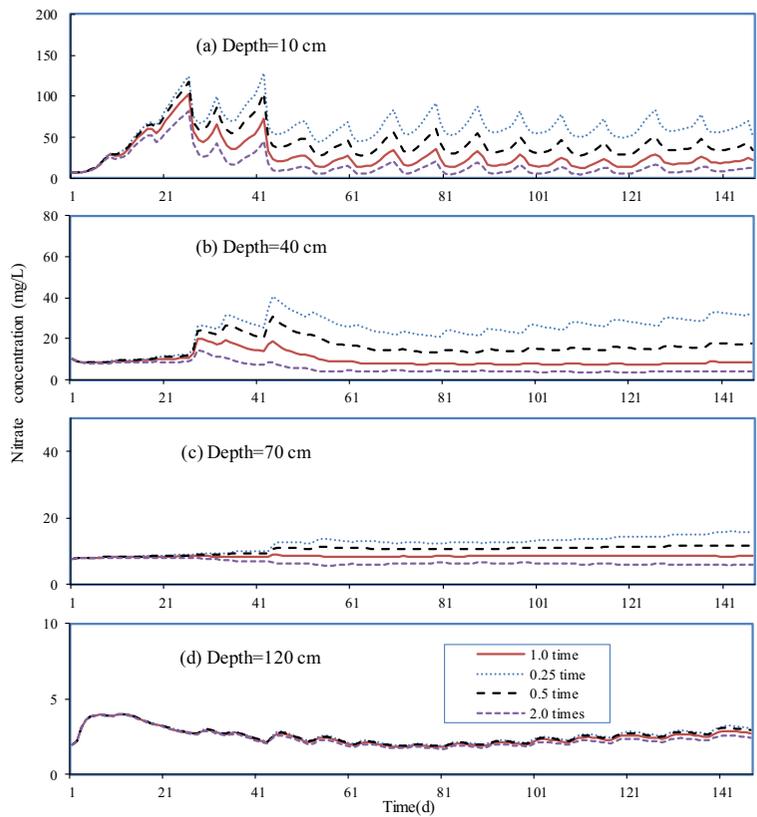


Table 4 Soil nitrate and ammonium mass balance analysis under the four scenarios of organic carbon concentration in the sewage water. Negative numbers are the amount of nitrate removed from the modeling system

Items		Scenarios			
		0.25 time	0.5 time	1 time	2 times
Nitrate	Initial nitrate mass (mg)	185.97	185.97	185.97	185.97
	Denitrification mass (mg)	-4608.58	-4689.87	-4711.07	-4696.94
	Nitrification mass (mg)	4053.71	4057.25	4053.7	4043.1
	Input nitrate mass (mg)	322.11	322.11	322.11	322.11
	Drained nitrate mass (mg)	-40.93	-40.43	-39.51	-37.92
	Ending nitrate mass (mg)	732.64	479.59	302.95	189.79
	Absolute error (mg)	818.52	645.34	494.79	372.15
	Relative error (%)	8.23	6.60	5.15	3.93
Ammonium	Initial ammonium mass (mg)	192.83	192.83	192.83	192.83
	Nitrification mass (mg)	-4053.71	-4057.25	-4053.7	-4043.1
	Mineralization mass (mg)	1824.70	1822.58	1818.34	1809.86
	Input ammonium mass (mg)	2361.19	2361.19	2361.19	2361.19
	Drained ammonium mass (mg)	-4.27	-4.27	-4.27	-4.27
	Ending ammonium mass (mg)	339.00	334.58	334.48	334.48
	Absolute error (mg)	18.51	18.63	18.52	18.58
	Relative error (%)	0.21	0.21	0.21	0.21

Table 5 Sensitivity index levels

Level	Value	Sensitivity
I	[0.00,0.05)	Not sensitive
II	[0.05,0.20)	Slight sensitive
III	[0.20,1.00)	Normal sensitive
IV	[1.00, ∞)	Very sensitive

5.2 Sensitivity Analysis Results

The sensitivity analysis considers three model outputs, and they are the amount of drainage water, the ammonium, and nitrate concentration in the drainage water at the end of the simulation. The sensitivity index evaluated for the three outputs are plotted in Fig. 19. Figure 19a shows that the drainage water amount is very sensitive to the exponent n , normal sensitive to the coefficient α , and the saturated hydraulic conductivity K_s . Soil water characteristic parameters have very slight impact on ammonium concentration, because soil has strong adsorption to ammonium. The ammonium concentration is very sensitive to the first-order nitrification rate constant K_n , normal sensitive to the zero-order rate coefficient in humus pool $(K_0)_h$, and the distribution coefficient for adsorbing solute K_d . It is not sensitive to the zero-order rate coefficient in litter pool $(K_0)_l$. The nitrate concentration is sensitive to many parameters except the transverse dispersivity α_T , the first-order nitrification rate constant K_n . It is very sensitive to n and K_{dn} , normal sensitive to α , $(K_0)_l$, and $(K_0)_h$, and slight sensitive to α_L . The reason

why the nitrate concentration is sensitive to n , α is that they can affect water content greatly in the soil profile.

6 Conclusions

This study focuses on investigating the effects of organic carbon in sewage irrigation water on soil nitrogen transformation and transport with the improved nitrogen model Nitrogen-2D by adding the organic carbon impact term. The improved model is calibrated and validated by the soil nitrogen dynamics under primary and secondary sewage water irrigation with high and low organic carbon concentration. The impact of organic carbon concentration and parameter sensitivity is analyzed using the calibrated model. The major conclusions are as follows.

- (1) The modified nitrogen model Nitrogen-2D with the added impact term of organic carbon yields more satisfactory simulation of the dynamics of soil water content and nitrate under the sewage water irrigation than the original model.
- (2) The Nitrogen-2D-based model simulations for the four different scenarios of organic carbon concentration indicate that higher organic carbon concentration can greatly decrease nitrate concentration in soil profiles. The mass balance analysis shows that the nitrate mass in the drained water decreases as the organic carbon concentration increases in sewage water.

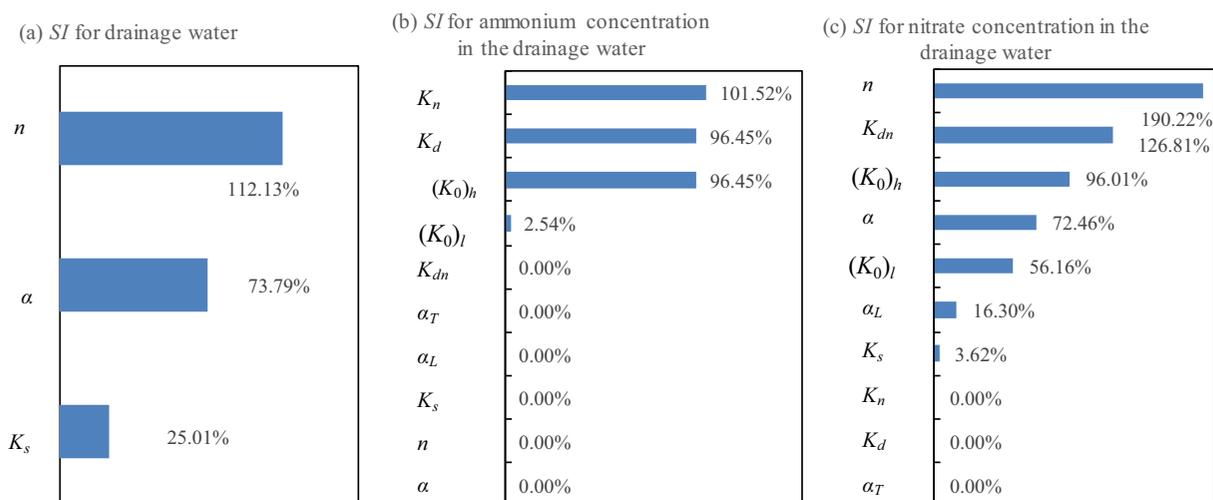


Fig. 19 Sensitivity index for the model outputs of **a** the drainage water amount, **b** the ammonium concentration in the drainage water in the end, **c** the nitrate concentration in the drainage water in the end

- (3) The results of local sensitivity analysis show that the drainage water amount is very sensitive to soil water retention function parameters and is normal sensitive to the saturated hydraulic conductivity. The ammonium concentration is very sensitive to the first-order nitrification rate constant, zero-order rate coefficient in humus pool, and the distribution coefficient for adsorbing solute. Many parameters impact the nitrate simulation results, which are soil water retention function parameters α and n and nitrogen transformation parameters K_{dn} , $(K_0)_i$, and $(K_0)_h$.

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