Biogeochemistry (2025) 168:79
https://doi.org/10.1007/s10533-025-01246-3

Identifying soil N_2O sources by combining laboratory experiments with process-based models

Zhifeng Yan · Zhaopei Chu · Balázs Grosz · Baoxuan Chang · Narasinha Shurpali · Gang Liu · Zhaolei Li · Jinsen Zheng · Si-liang Li · Klaus Butterbach-Bahl

Received: 10 February 2025 / Accepted: 22 May 2025 / Published online: 9 October 2025
© The Author(s) 2025

Abstract Nitrification and denitrification are two important biological processes producing N_2O in soils, but their contributions to N_2O emissions are not well understood, hindering precise mitigation measures. Here, we developed process-based models (PBM) with and without transport (T) to partition N_2O sources by tracking nitrogen flows (NF) through different reaction pathways. The model with transport (PBM-T-NF) well predicted N_2O production from nitrification and denitrification in two different repacked soils with a shallow depth of 8 mm under moisture conditions ranging from 40 to 100% water-filled pore space (WFPS), demonstrating its robustness and reliability. In comparison, the model without transport (PBM-NF) failed to capture the N_2O dynamics and the relative contribution of denitrification to N_2O production (C_D), highlighting the need of including mass transport in predicting N_2O dynamics. The PBM-T-NF model was further employed to

Responsible Editor: Naomi Wells.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s10533-025-01246-3.

Z. Yan (✉) · Z. Chu · B. Chang · S. Li
Institute of Surface-Earth System Science, School of Earth System Science, Tianjin University, Tianjin 300072, China
e-mail: yanzf17@tju.edu.cn

Z. Yan · S. Li
Critical Zone Observatory of Bohai Coastal Region, Tianjin Key Laboratory of Earth Critical Zone Science and Sustainable Development in Bohai Rim, Tianjin University, Tianjin 300072, China

B. Grosz
Thünen Institute of Climate-Smart Agriculture, 38116 Brunswick, Germany

N. Shurpali
Natural Resources Institute Finland, Halolantie 31 A, 71750 Maaninka, Finland

G. Liu
College of Management and Economics, Tianjin University, Tianjin 300072, China

Z. Li
Key Laboratory of Low-Carbon Green Agriculture in Southwestern China, Ministry of Agriculture and Rural Affairs, Interdisciplinary Research Center for Agriculture Green Development in Yangtze River Basin, College of Resources and Environment, Southwest University, Chongqing, China

J. Zheng
Crop, Livestock and Environment Division, Japan International Research Center for Agricultural Sciences, Ohwashi 1-1, Tsukuba, Ibaraki 305-8686, Japan

K. Butterbach-Bahl
Institute of Meteorology and Climate Research, Atmospheric Environmental Research, Karlsruhe Institute of Technology, Garmisch-Partenkirchen, Germany

K. Butterbach-Bahl
Pioneer Center Land-CRAFT, Agroecology, Aarhus University, Aarhus C, Denmark

investigate the effects of soil properties on N_2O emissions and sources. Increased NH_4^+ concentration significantly decreased C_D under relatively low moisture conditions, while increased NO_3^- slightly promoted C_D over different moisture contents, emphasizing the importance of substrate availability and moisture conditions in controlling C_D. Furthermore, the PBM-T-NF model was used to quantify N_2O sources from an artificial soil core of 80 mm depth. Soil depth was shown to be important in mediating C_D by controlling O_2 diffusivity, which is highly dependent on moisture content. Given the long-standing challenge in experimental quantification of N_2O sources from soils, our developed model provides a novel way to estimate N_2O production from different nitrogen processes, which is key for accurately targeting mitigation of N_2O emissions from soils.

Keywords Nitrification · Denitrification · Nitrous oxide · Process-based model · Mass transport

Introduction

Nitrous oxide (N_2O) plays an important role in driving global warming and depleting stratospheric ozone (IPCC 2021). Natural and managed soils are major sources of atmospheric N_2O, accounting for about half (7.9 Tg yr^{-1}) of emissions from 2007 to 2016 (Tian et al. 2020). However, this estimate of N_2O emissions is highly uncertain, ranging from 6.3–10.3 Tg yr^{-1} (Tian et al. 2020), largely because the complicated processes that produce, transport and consume N_2O are difficult to be accurately characterized and incorporated in models that estimate global N_2O emissions (Butterbach-Bahl et al. 2013; Müller et al. 2014).

Nitrification and denitrification are the two primary biological processes that produce N_2O in soils (Wang et al. 2023). The magnitude of N_2O emission and its attribution from nitrification and denitrification are influenced by various environmental factors, such as substrate availability (Laville et al. 2011), O_2 concentration (Song et al. 2019), and soil structure and texture (Lucas et al. 2023). Soil moisture is a key regulator of N_2O emissions and sources, mainly by modulating substrate and O_2 availability (Smith 2017). As soil moisture increases, the rate of N_2O production is expected to decrease after reaching a maximum value, and the moisture tipping point (i.e., the optimal water content) at which the maximum N_2O flux rate occurs varies with soil properties (Davidson et al. 2000). In general, nitrification dominates soil N_2O emissions under relatively low moisture conditions, while denitrification dominates under high moisture conditions (Han et al. 2024; Kool et al. 2011, 2007). Therefore, quantification of N_2O production from the two processes under different moisture conditions is critical for accurate estimation of soil N_2O emissions.

Several approaches have been employed to quantify soil N_2O production from nitrification and denitrification (Bateman and Baggs 2005; Groffman et al. 2006; Heinen 2006). Inhibitors, such as acetylene (C_2H_2), have been widely used to separate nitrification and denitrification due to their simplicity and low cost (Bateman and Baggs 2005; Watts and Seitzinger 2000). However, this approach has been reported to systematically underestimate N_2O production from denitrification (Watts and Seitzinger 2000). In comparison, isotopic techniques, including natural and enrichment abundance approaches, have been shown to be more reliable in distinguishing different nitrogen (N) processes (Kool et al. 2011; Wang et al. 2024; Yang et al. 2019; Zhu et al. 2013). In particular, enrichment approaches with $^{15}N\text{-}NH_4^+$ and/or $^{15}N\text{-}NO_3^-$ additions have been widely used especially in agricultural soils (Bateman and Baggs 2005; Wang et al. 2023; Zhang et al. 2015), providing valuable insights into N_2O sources and the underlying mechanisms (Friedl et al. 2021; Wang et al. 2024). Furthermore, the natural isotope techniques, such as ^{15}N site preference, can quantify N_2O emissions from different N processes without interfering with soil N cycling (Butterbach-Bahl et al. 2013), and are mostly applied in the field (Wei et al. 2023). Although N isotope data have been applied to constrain soil N_2O emissions at global scale (Harris et al. 2022), conclusions derived from ^{15}N signals are often site- or soil-specific, and the high cost of isotopic techniques also limits their applications on a large scale (Ruser et al. 2006; Wei et al. 2023). The use of models provides another effective means of deriving regional or global N_2O estimates and to target mitigation options.

A large number of models have been developed to simulate N_2O emissions from soils (Butterbach-Bahl et al. 2013; Heinen 2006; Tian et al. 2018). Emission factor approaches are often used at regional or global scales, when the data needed to calculate N_2O

emissions across spatial and temporal scales are not available (Wang et al. 2020a). These approaches are straightforward, but also have large uncertainties (Del Grosso et al. 2020). In comparison, process-based models are typically more accurate when applied at the site or farm scale (Ehrhardt et al. 2018; Yue et al. 2019). Numerous process-based models have been proposed to simulate N_2O emissions with varying complexities (Del Grosso et al. 2020; Tian et al. 2019). The simplified ones, such as DAISY, often correlate N_2O flux with estimates of soil N cycling (Hansen 2002); the detailed ones, such as SLIM, further account for the effects of soil structure on gas diffusion (Vinten et al. 1996); and the advanced ones, such as DNDC, explicitly quantify the dynamics of different microbial functional groups (Li et al. 2000). Most process-based models include N_2O production from nitrification and denitrification (Butterbach-Bahl et al. 2013; Tian et al. 2019), where empirical relationships between N_2O flux and environmental factors are widely applied (Del Grosso et al. 2000; Wang et al. 2021). In particular, the response of N_2O fluxes to changes in soil moisture, i.e., moisture reduction functions, are highly soil specific (Friedl et al. 2021), and their empirical application in process-based models is a major source of uncertainty in N_2O estimate (Heinen 2006). Furthermore, although ^{15}N signals have been employed in models to distinguish various N processes and quantify N_2O emissions, these models, including a variety of N trace models (Jansen-Willems et al. 2022; Müller et al. 2014; Zheng et al. 2023), often use optimization technique such as Markov Chain Monte Carlo (MCMC) to quantify different N processes. By contrast, the combination of process-based models and N isotopic approaches are less explored.

Soil N_2O emission is an integral consequence of N_2O production, transport, and consumption (Butterbach-Bahl et al. 2013; Müller et al. 2014). As water is relatively stagnant in soils, the transport of N_2O inside soils is mainly determined by gas diffusivity (Yan et al. 2018b), since gas diffuses in air approximately ten thousand faster than in water (Stumm and Morgan 1996). Moreover, the gas diffusivity is highly dependent on soil structure and moisture contents (Fu et al. 2024; Yan et al. 2016), whose interactions make soil N_2O emissions difficult to predict (Rabot et al. 2015). Current models, including DLEM, APSIM and DayCent, often neglect the transport of N_2O in soils by assuming that the produced N_2O is directly released to atmosphere, partly because the soil gas diffusivity is difficult to quantify experimentally (Tian et al. 2018; Parton et al. 1996; Del Grosso et al. 2020). This assumption is generally valid under low moisture condition but likely overestimates N_2O emissions under high moisture conditions, in which part of N_2O is reduced to N_2 due to their long retention time (Baggs 2011). Only a few models directly quantify the gas diffusion in soil profile (Li et al. 2000; Klier et al. 2011). For example, the DNDC model quantifies O_2 diffusion to determine the redox potential in soils, but neglects N_2O diffusion along soil profiles (Li et al. 2000). Therefore, it is necessary to incorporate N_2O transport in model simulations.

To better simulate soil N_2O emissions and sources, here we: (1) developed process-based models (PBM) with and without transport (T) to quantify N_2O production from nitrification and denitrification by tracking nitrogen flows (NF) in their reaction pathways; (2) evaluated the developed models by using incubation experiments, in which enriched ^{15}N techniques were applied to measure N_2O emissions from nitrification and denitrification under six moisture levels (40–100% WFPS) (Wang et al. 2023); and (3) used the model with transport (PBM-T-NF) to investigate the effects of soil conditions on N_2O emissions and sources. The PBM-T-NF model explicitly quantified the transport of solutes (i.e., dissolved N species, dissolved organic carbon, and dissolved O_2) and gases (i.e., NO, N_2O, N_2, and O_2) inside soils as well as their impacts on N_2O production and consumption, which together determine N_2O emissions. To focus on diffusion process, other transport processes such as advection were not included in the models, and the diffusion was described by Fick's Law (Yan et al. 2018a). By tracking the N flows through nitrification and dentification based on ^{15}N signals, the developed models are also able to reliably quantify the contribution of nitrification and dentification to N_2O emissions under different environmental conditions. Consequently, the developed model is able to evaluate the effects of solutes and gases diffusion on N_2O emissions, and quantify their attributions from nitrification and denitrification, which may reduce the uncertainty of estimating N_2O emissions from soils.

Methods

Partition of N_2O sources by tracking nitrogen flows

To simplify the model development and avoid over-parameterization, the developed models only accounted for the two most important pathways producing N_2O (i.e., nitrification and denitrification) and neglected other microbial processes such as anaerobic ammonium oxidation (ANAMMOX) and chemical processes such as chemodenitrification. The reaction pathways used in the models are depicted in Fig. 1a.

Accordingly, the stoichiometry of each reaction pathway is described as follows, where Eqs. (1a–4a) represent nitrification and Eqs. (5a–8a) represent denitrification (Maggi et al. 2008):

$$2NH_4^+ + 3O_2 \rightarrow 2NO_2^- + 4H^+ + 2H_2O \quad (1a)$$

$$5NO_2^- + NH_4^+ \rightarrow 6NO + 4OH^- \quad (2a)$$

$$8NO + 2NH_4^+ \rightarrow 5N_2O + 2H^+ + 3H_2O \quad (3a)$$

$$2NO_2^- + O_2 \rightarrow 2NO_3^- \quad (4a)$$

$$2NO_3^- + CH_2O \rightarrow 2NO_2^- + CO_2 + H_2O \quad (5a)$$

$$4NO_2^- + CH_2O + 4H^+ \rightarrow 4NO + CO_2 + 3H_2O \quad (6a)$$

$$4NO + CH_2O \rightarrow 2N_2O + CO_2 + H_2O \quad (7a)$$

$$2N_2O + CH_2O \rightarrow 2N_2 + CO_2 + H_2O \quad (8a)$$

Microbes (i.e., nitrifier and denitrifier communities) were explicitly considered in the models. Dual Michaelis–Menten kinetic equations were used to calculate the reaction rate (r) for each pathway, and the r of denitrification was inhibited by O_2 concentration (Chang et al. 2022).

$$r_{NH_4^+-NO_2^-,N} = -\mu_{NH_4^+-NO_2^-} \cdot X_{AOB} \cdot \frac{C_{NH_4^+}}{C_{NH_4^+} + K_{NH_4^+,NH_4^+-NO_2^-}} \cdot \frac{C_{O_2,a}}{C_{O_2,a} + K_{O_2,NH_4^+-NO_2^-}} \quad (1b)$$

$$r_{NO_2^--NO,N} = -\mu_{NO_2^--NO,N} \cdot X_{AOB} \cdot \frac{C_{NO_2^-}}{C_{NO_2^-} + K_{NO_2^-,NO_2^--NO,N}} \cdot \frac{C_{NH_4^+}}{C_{NH_4^+} + K_{NH_4^+,NO_2^--NO}} \quad (2b)$$

$$r_{NO-N_2O,N} = -\mu_{NO-N_2O,N} \cdot X_{AOB} \cdot \frac{C_{NO,a}}{C_{NO,a} + K_{NO,NO-N_2O,N}} \cdot \frac{C_{NH_4^+}}{C_{NH_4^+} + K_{NH_4^+,NO-N_2O}} \quad (3b)$$

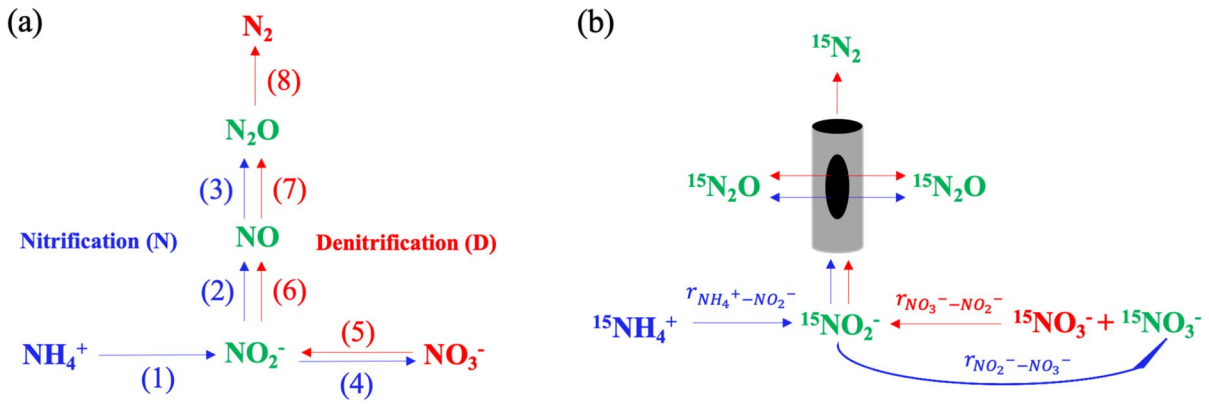

Fig. 1 **a** Reaction pathways of nitrification and denitrification considered in this study. **b** Illustration showing the N flows of producing N_2O as indicated by ^{15}N. The numbers in **a** refer to the corresponding reaction pathways in the text, and the reactions (r) in **b** can be found in the text

$$r_{NO_2^- - NO_3^-,N} = -\mu_{NO_2^- - NO_3^-} \cdot X_{NOB} \cdot \frac{C_{NO_2^-}}{C_{NO_2^-} + K_{NO_2^-,NO_2^- - NO_3^-}} \cdot \frac{C_{O_2,a}}{C_{O_2,a} + K_{O_2,NO_2^- - NO_3^-}} \quad (4b)$$

$$r_{NO_3^- - NO_2^-,D} = -\mu_{NO_3^- - NO_2^-,D} \cdot X_{DEN} \frac{C_{NO_3^-}}{C_{NO_3^-} + K_{NO_3^-,NO_3^- - NO_2^-}} \cdot \frac{C_{DOC}}{C_{DOC} + K_{DOC,NO_3^- - NO_2^-}} \cdot \frac{I_{O_2,NO_3^- - NO_2^-,D}}{C_{O_2,a} + I_{O_2,NO_3^- - NO_2^-,D}} \quad (5b)$$

$$r_{NO_2^- - NO,D} = -\mu_{NO_2^- - NO,D} \cdot X_{DEN} \cdot \frac{C_{NO_2^-}}{C_{NO_2^-} + K_{NO_2^-,NO_2^- - NO,D}} \cdot \frac{C_{DOC}}{C_{DOC} + K_{DOC,NO_2^- - NO,D}} \cdot \frac{I_{O_2,NO_2^- - NO}}{C_{O_2,a} + I_{O_2,NO_2^- - NO}} \quad (6b)$$

$$r_{NO - N_2O,D} = -\mu_{NO - N_2O,D} \cdot X_{DEN} \cdot \frac{C_{NO,a}}{C_{NO,a} + K_{NO,NO - N_2O,D}} \cdot \frac{C_{DOC}}{C_{DOC} + K_{DOC,NO - N_2O,D}} \cdot \frac{I_{O_2,NO - N_2O}}{C_{O_2,a} + I_{O_2,NO - N_2O}} \quad (7b)$$

$$r_{N_2O - N_2} = -\mu_{N_2O - N_2} \cdot X_{DEN} \cdot \frac{C_{N_2O,a}}{C_{N_2O,a} + K_{N_2O,N_2O - N_2}} \cdot \frac{C_{DOC}}{C_{DOC} + K_{DOC,N_2O - N_2}} \cdot \frac{I_{O_2,N_2O - N_2}}{C_{O2,a} + I_{O_2,N_2O - N_2}} \quad (8b)$$

where r_{A-B} represents the production rate of B from A, μ_{A-B} represents the maximum reaction rate of B from A, $K_{C,A-B}$ represents the half-saturation concentration of C during the conversion from A to B, $I_{C,A-B}$ represents the inhibition constant of C during the conversion from A to B, X_{AOB}, X_{NOB} and X_{DEN} represent the biomass content of ammonia-oxidizing bacteria, nitrite-oxidizing bacteria and denitrifiers, respectively. C_A represents the concentration of A. The subscripts of N and D refer to the individual process. The subscript of a represents aqueous gases, i.e., dissolved gases, to distinguish them from gaseous gases. More descriptions about the parameters can be found in the Supplementary information (SI) Table S1 and Table S2.

The contribution of nitrification and denitrification to N_2O production is assumed to depend on the NO_2^- content derived from nitrification and denitrification, that is, the N fluxes from NH_4^+ and NO_3^- to NO_2^-. Since NO_3^- is simultaneously consumed by denitrification and replenished by nitrification (Fig. 1b), the total N fluxes from initial NH_4^+ to NO_2^- (F_N) and from initial NO_3^- to NO_2^- (F_D) can be calculated as

$$F_N = \int_0^{t_1} r_{NH_4^+ - NO_2^-} dt \quad (9)$$

$$F_D = \int_0^{t_1} r_{NO_3^- - NO_2^-} dt \cdot \frac{m_{NO3,0}}{m_{NO3,0} + \int_0^{t_1} r_{NO_2^- - NO_3^-} dt \cdot \frac{\int_0^{t_1} r_{NH_4^+ - NO_2^-} dt}{\int_0^{t_1} (r_{NH_4^+ - NO_2^-} + r_{NO_3^- - NO_2^-}) dt}} \quad (10)$$

where t_1 is the reaction time, and $m_{NO3,0}$ is the initial amount of NO_3^-. The numerical calculation of F_N and F_D during the simulations can be found in the Supplemental information "Methods" section.

Therefore, the contribution ratio of denitrification (C_D) to N_2O production can be calculated by

$$C_D = \frac{F_D}{F_N + F_D} \quad (11)$$

and the contribution ratio of nitrification to N_2O production equals 1-C_D.

Process-based models quantifying N_2O production and emissions

The developed process-based models accounted for the production and consumption of different N species, including NH_4^+, NO_3^-, NO_2, and N_2O, during the processes of nitrification and denitrification. The two models with and without transport were developed to examine the effect of transport on N_2O production and emissions from soils.

In the process-based model with transport (PBM-T-NF), the vertical transport is included. Since the soil water was stagnant during the simulation, the advection process related to water movement was neglected in our study. Only the diffusion of aqueous and gaseous species within soils was included in the developed models. The governing equations are described by

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x}\left(D_i \frac{\partial C_i}{\partial x}\right) + r_i \quad (12)$$

where C_i is concentration of dissolved (i.e., aqueous, including NH_4^+, NO_3^-, NO_2, and DOC) or gaseous species (including NH_3, NO, N_2O, N_2, and O_2), and D_i is the effective diffusion coefficient. The values of D_i for aqueous ($D_{i,a}$) and gaseous ($D_{i,g}$) species can be calculated by following equations (Hamamoto et al. 2010)

$$\frac{D_{i,a}}{D_{i,a,0}} = \phi^{m_a - n_a}\theta^{n_a} \tag{13}$$

$$\frac{D_{i,g}}{D_{i,g,0}} = \phi^{m_g - n_g}(\phi - \theta)^{n_g} \tag{14}$$

where $D_{i,a,0}$ and $D_{i,g,0}$ are the corresponding diffusion coefficients in pure water and air, ϕ is porosity, θ is volumetric soil moisture, m_a, n_a and m_g, n_g are empirical parameters accounting for the effect of tortuosity and pore connectivity on diffusion of aqueous and gases species, respectively, in soils.

The term r_i is the sources or sinks of C_i, and was calculated according to the reaction pathways (Eqs. 1a–8a) and the corresponding reaction rates (Eqs. 1b–8b). $r_i = 0$ for the gaseous species. The gaseous and aqueous gases, including NH_3, NO, N_2O, N_2, and O_2, are assumed to reach equilibrium in each numerical voxel following the Henry's law (Sander 2015):

$$C_{i,a} = \frac{C_{i,g}}{K_{i,eqi}} \tag{15}$$

where $C_{i,a}$ is the concentration of aqueous gas, $C_{i,g}$ is the concentration of gaseous gas, and $K_{i,eqi}$ is Henry's law constant.

In the process-based model without transport (PBM-NF), the mass transport was ignored and all physicochemical constituents were assumed to be uniformly distributed in the soil. The governing equations can be simplified into

$$\frac{\partial C_i}{\partial t} = r_i \tag{16}$$

The gas exchange rates between the soil and the headspace, including N gases emissions and O_2 uptake, are calculated using Fick's law (Yan et al. 2018a):

$$R_i = D_{i,a}\frac{C_{i,a,top} - C_{i,headspace}K_{i,eq}}{\Delta x/2} + D_{i,g}\frac{C_{i,g,top} - C_{i,headspace}}{\Delta x/2} \tag{17}$$

where $C_{i,a,top}$ and $C_{i,g,top}$ are the concentrations of aqueous and gaseous gas in the top numerical voxel, respectively. $C_{i,headspace}$ is the concentration of gas in the headspace, $K_{i,eq}$ is the Henry constant, and Δx is the spatial resolution of numerical voxel.

The adsorbed and dissolved NH_4^+ in each numerical voxel are assumed to be in equilibrium according to the Langmuir model [see Supplemental information equation (S1)]. The dissolved NH_4^+ and dissolved NH_3 are assumed to reach equilibrium in each numerical voxel as a function of pH [see Supplemental information equation (S2)]. The dissolved organic carbon (DOC) is assumed to be replenished by adsorbed organic carbon (SOC) via desorption [see Supplemental information equation (S3)].

Model calibration and validation

The process-based models with and without transport, in which N_2O sources are partitioned by tracking N flows, were calibrated and validated using laboratory incubation experiments.

The experiments measured N_2O production from nitrification and denitrification in two different fluvo-aquic soils by using the enriched ^{15}N tracing technique (Wang et al. 2023). Soil samples (0–15 cm) were collected in October 2020 from two long-term agricultural experimental sites [Luan Cheng (LC), Hebei (37°53′ N, 114°41′E) and Shang Zhuang (SZ), Beijing (39°48′N, 116°28′E)] in the North China Plain. The cropping system was rotated with winter wheat and summer maize in the LC and SZ. The physicochemical properties of the two soils are listed in Table 1.

Incubation experiments were conducted to quantify N_2O production from nitrification and denitrification under 40–100% water-filled pore space (WFPS). $K^{15}NO_3$ (10.16 atom%) was applied at a rate of 50 mg NH_4^+-N kg^{-1} to identify the source of N_2O-N, and additional NH_4Cl was added at a rate of 50 mg NO_3^--N kg^{-1}. Soil (20 g oven-dry equivalent) was added to each 120 mL incubation flask, with a bulk density of 1 g cm^{-3} and a soil depth of 8 mm. The flasks were pre-incubated in dark at 25 °C for 7 days, and then incubated for another 48 h after ^{15}N

Table 1 Soil properties used for model calibration and validation

	Soil texture			pH	SOC (g kg^{-1})	NO_3^--N (mg kg^{-1})	NH_4^+-N (mg kg^{-1})
	Sand (%)	Silt (%)	Clay (%)				
LC soil (model calibration)	29.2	64.1	6.7	7.92	19.82	30.49	2.08
SZ soil (model validation)	36.1	56.4	7.5	7.89	10.93	22.50	3.07

application. Concentrations and ^{15}N isotopic signatures of NH_4^+, NO_3^-, and N_2O were measured after 12, 24, and 48 h. The concentrations of NH_4^+-N and NO_3^--N were measured using a continuous-flow analyzer (Skalar Analytical, Breda, The Netherland), and the concentrations of N_2O were measured using gas chromatography (Agilent 7890, Santa Clara, CA, USA). Isotope analysis of NH_4^+-N and NO_3^--N were performed on aliquots of the extracts using a diffusion technique (Brooks et al. 1989) and the ^{15}N isotopic signature was measured by isotope ratio mass spectrometry (IRMS 20–22, Sercon, Crewe, UK). The ^{15}N signature of N_2O was determined using a Thermo Finnigan MAT-253 spectrometer (Thermo Fisher Scientific, Waltham, MA, USA).

The contribution ratios of nitrification, $1 - C_D$, and denitrification, C_D, to N_2O production were calculated from the changes in ^{15}N atom% of NH_4^+, NO_3^-, and N_2O by using the following equation (Stevens et al. 1997)

$$C_d = \frac{\left(a_{N_2O} - a_{NH_4}\right)}{\left(a_{NO_3} - a_{NH_4}\right)} \quad \text{with} \quad a_{NO_3} \neq a_{NH_4} \tag{18}$$

where aN_2O is the ^{15}N atom% enrichment of the N_2O produced by nitrification and denitrification, and aNO_3 and aNH_4 are the ^{15}N atom% enrichment of soil NO_3^- and NH_4^+ at the time of gas sampling. More details about the experiments can be found in our previous experimental study (Wang et al. 2023).

The developed models were first calibrated with the experimental measurements of the LC soil and then validated with those of the SZ soil. The simulated concentrations of NH_4^+ and NO_3^- as well as the N_2O fluxes and C_D were compared with the measured values (n = 48). The simulated NH_4^+ and NO_3^- concentrations as well as C_D were averaged over the soil profile in the PBM-T-NF model for comparisons with the measured ones. To minimize the effect of gas accumulation and transport inside soils on the PBM-NF model, the experimental measurements over the first 12 h were used to calibrate and validate the developed models.

The eight maximum reaction rates (μ_i, see Table S1) for nitrification and denitrification were first determined based on manual fitting. The values of μ_i were then optimized using Markov Chain (MC) approach by randomly changing the parameter values 1000 times from half to twice the initial values (see Supplemental information "Results" section for parameterization) (Brooks 1998). The parameter values that produced the minimum accumulated normalized root mean square error (*nRMSE*) of NH_4^+, NO_3^-, and N_2O concentrations during the 10,000 times of simulations, were chosen, for which (Abdalla et al. 2020)

$$nRMSE = \frac{RMSE}{\overline{M}} \tag{19}$$

and

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(S_i - M_i)^2}{n}} \tag{20}$$

where $\overline{M}$ is the average of the measured values, S_i and M_i are the simulated and measured values under different moisture contents, and n is the treatment number of moisture contents (i.e., $n = 6$).

Effects of model parameters and soil conditions

The influence of the maximum reaction rates (μ_i) on N_2O emissions and sources (C_D) under different soil moisture contents was evaluated by using the PBM-T-NF model. The experimental setup for model calibration was employed for the model sensitivity analysis (see Table 1). The effects of NH_4^+ and NO_3^- concentrations, bulk density, and soil depth on N_2O production and C_D were further investigated using the PBM-T-NF model. All parameters except the investigated factors remained unchanged during

the simulations (see Table 1, S1 and S2). The N_2O emissions and sources under different soil moisture contents were analyzed.

To further investigate the effect of vertical mass transport on N_2O emissions and sources, we created an artificial soil core with a height of 80 mm. The physicochemical properties in the artificial soil core were assigned the same as those of the experimental soil samples, and they were uniformly distributed along the soil depth. The dissolved and gaseous species could diffuse between adjacent layers, while the soil moisture was kept constant during the simulations. Unlike the closed system in other simulations, the soil core was assumed to be open to the atmosphere, mimicking the field situation, and its N_2O flux was calculated by Eq. (17), where the gas concentration in the headspace is the atmospheric concentration.

Numerical setup and procedure

Matlab codes were developed to solve the governing equations. The simulated soils were considered as a single numerical voxel in the PBM-NF model and uniformly stratified in the PBM-T-NF model. The finite-difference method was used for the spatial discretization, and the spatial resolution of the numerical voxels (Δx) was 1 mm. The explicit Euler method was used for the temporal evolution, and a small time step (Δt = 0.125 s) was used to avoid negative values during the simulations. The initial concentrations of NH_4^+ and NO_3^- used in the model evaluation simulation are presented in Supplemental information Table S3. The initial concentrations of SOC, O_2, and different N species and pH value were either obtained from literature or given by the experiments (see Supplemental information Table S1). The initial concentration of DOC was assumed to reach equilibrium with SOC, and the pH remained unchanged.

Results

Model calibration

Both the models with and without transport (PBM-T-NF and PBM-NF) overpredicted NH_4^+ concentrations at low soil moisture conditions (24.18% larger at WFPS = 0.4 and 20.05% larger at WFPS = 0.6), but underpredicted them at high soil moisture levels conditions except for WFPS = 1.0 (13.12% smaller at WFPS = 0.8 and 38.52% larger at WFPS = 0.9, Fig. 2a). In contrast, the two models predicted NO_3^- concentrations well over the different soil moisture levels except for WFPS = 1.0 (averaged nRMSE is 13.58%, Fig. 2b). Compared to the PBM-NF model, the PBM-T-NF model produced much better N_2O concentrations across different soil moisture levels by capturing the low N_2O concentrations at low moisture levels (i.e., WFPS = 0.4) and the high N_2O concentrations at high moisture levels (i.e., WFPS = 0.9) (averaged nRMSE is 64.38% for PBM-NF and 36.35% for PBM-T-NF, Fig. 2c). The PBM-T-NF model also predicted the increasing trends of C_D with increasing soil moisture content (Fig. 2d), although it underestimated C_D under intermediate moisture conditions and overestimated it under low (WFPS = 0.4) and saturated (WFPS = 1.0) moisture conditions. In contrast, the PBM-NF model produced nearly constant C_D except for WFPS = 1.0.

Model validation

Both the PBM-T-NF and PBM-NF models captured the changing trends of NH_4^+, NO_3^-, and N_2O concentrations with increasing soil moisture in the SZ soil. Compared with the PBM-NF model, the PBM-T-NF model produced more accurate NH_4^+ (Fig. 3a) and NO_3^- (Fig. 3b) concentrations under high moisture conditions and much better N_2O concentration under relatively low soil moisture conditions (Fig. 3c). Furthermore, the PBM-T-NF model reliably reproduced the C_D as WFPS $\geq$ 0.7 (averaged nRMSE is 10.64%, Fig. 3d). By contrast, the PBM-NF model failed to capture the gradual increase in C_D as WFPS $\geq$ 0.7 (averaged nRMSE is 53.87%, Fig. 3d). Overall, the PBM-T-NF model well predicted the changes in all the four variables when soil moisture varied in a wide range, and it was used to evaluate the effects of model parameters and soil conditions on N_2O emissions and sources in the following sections.

Sensitivity analysis

The maximum reaction rates, $\mu_{NH_4^+-NO_2^-}$ and $\mu_{NO_3^--NO_2^-}$, were found to significantly affect C_D.

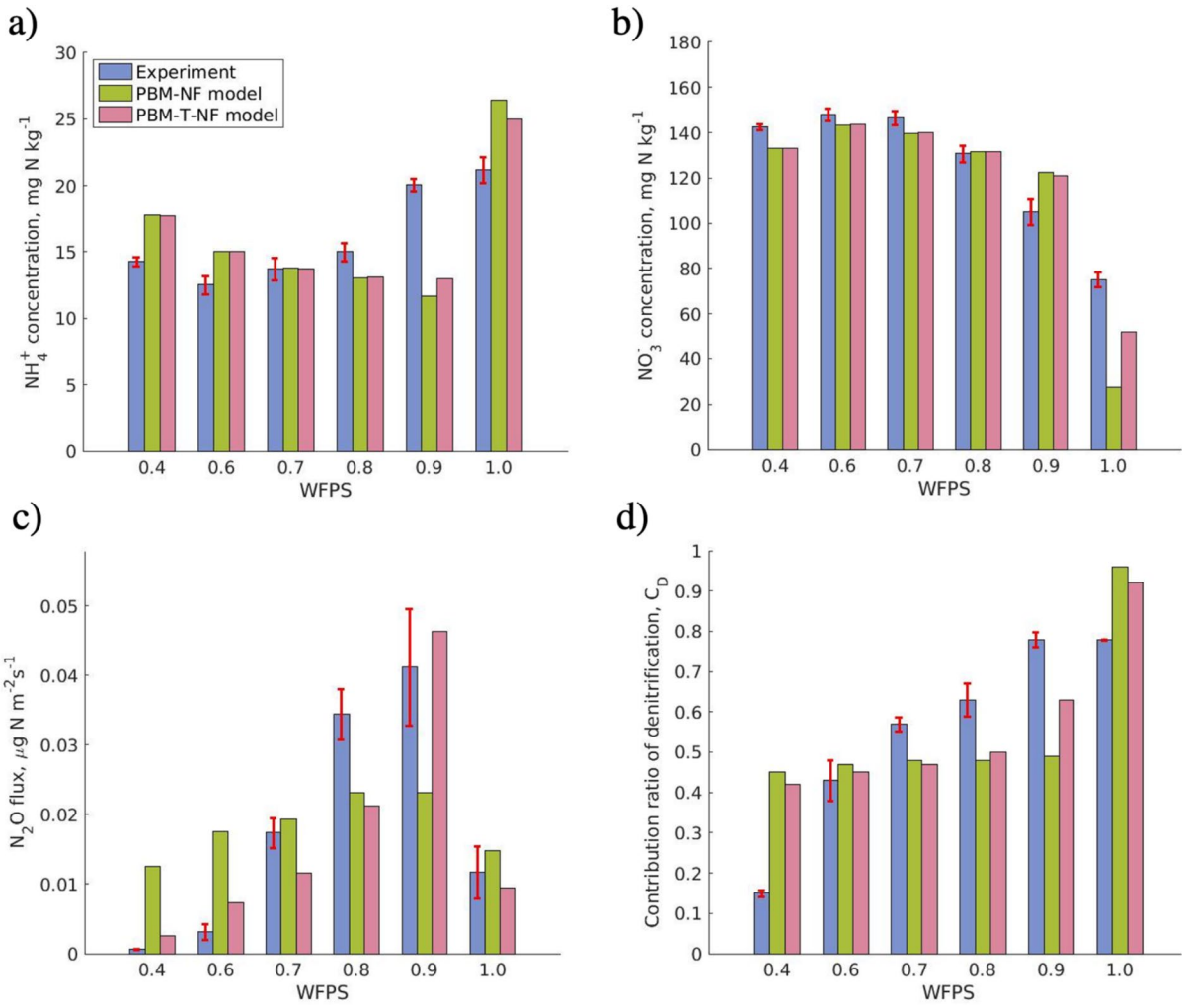

Fig. 2 Comparisons of the measured and modelled **a** total NH_4^+ concentration and **b** dissolved NO_3^- concentration at 12 h, and **c** soil N_2O fluxes and **d** contribution ratio of denitrification to N_2O production, C_D, over the first 12 h. The experimental results of the LC soil were used to calibrate the developed models

Increased $\mu_{NH_4^+-NO_2^-}$ reduced C_D especially under low soil moisture conditions (Fig. 4a). Conversely, increased $\mu_{NO_3^--NO_2^-}$ promoted C_D across different moisture levels (Fig. 4b). By comparison, the effects of other maximum reactions rates, including $\mu_{NO_2^--NO,N}$, $\mu_{NO-N_2O,N}$, $\mu_{NO_2^--NO_3^-}$, $\mu_{NO_2^--NO,D}$, $\mu_{NO-N_2O,D}$, $\mu_{N_2O-N_2}$, can be neglected (results not show). However, all these parameters except $\mu_{NO-N_2O,N}$ modulated N_2O emissions (Fig. S1). Especially, increased $\mu_{NO_2^--NO,N}$ (Fig. S1b) and $\mu_{NO_3^--NO_2^-}$ (Fig. S1e) strongly promoted N_2O emissions, while increased $\mu_{NO_2^--NO_3^-}$ (Fig. S1d) substantially depressed N_2O emissions.

Effects of soil conditions on N_2O sources and emissions

All four soil physiochemical properties clearly influenced C_D. Increased NH_4^+ concentration significantly decreased C_D under relatively low moisture conditions, i.e., WFPS $<=0.8$ (Fig. 5a). In comparison, increased NO_3^- concentration slightly increased C_D over the different moisture conditions (Fig. 5b). Increased soil depth also promoted C_D, especially under high moisture conditions except for WFPS = 1.0 (Fig. 5d). In contrast to the consistent effects of the above three soil properties across

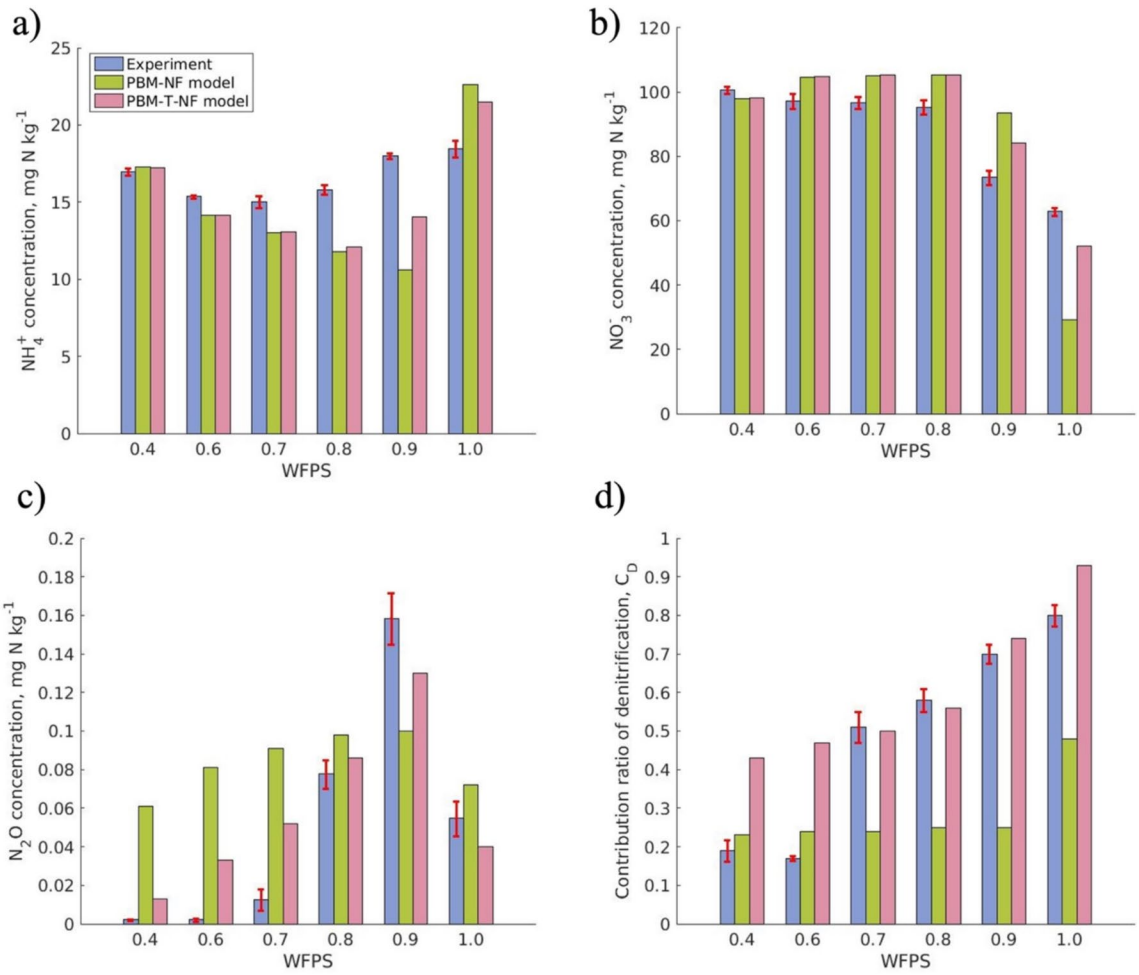

Fig. 3 Comparisons of the measured and modelled **a** total NH_4^+ concentration and **b** dissolved NO_3^- concentration at 12 h, and **c** soil N_2O flux and **d** contribution ratio of denitrification to N_2O production, C_D, over the first 12 h. The experimental results of the SZ soil were used to validate the developed models

different moisture levels, increased bulk density reduced C_D at relatively low moisture levels but increased C_D under high moisture levels (Fig. 5c). Correspondingly, the increased NH_4^+ concentration significantly increased N_2O emissions (Fig. S2a), while the other three soil properties have minor effects on N_2O emissions (Fig. S2b-d).

N_2O emissions and sources from the artificial soil core under different moisture levels

The N_2O flux from the artificial soil core first increased and then decreased with increasing soil moisture, reaching the maximum under WFPS = 0.8 (Fig. 6a). The N_2O flux approaches zero at WFPS = 1.0, because the N_2O was almost completely denitrified to N_2 under saturated conditions. Correspondingly, C_D increased with soil moisture, reaching almost 100% at WFPS = 1.0 (Fig. 6b). With time, the N_2O flux decreased and C_D increased. In particular, N_2O decreased by 113% at WFPS = 0.7 from 12 to 24 h, while C_D increased by no more than 13% at all the moisture levels.

Figure 7 shows the profile distributions of NH_4^+, NO_3^-, N_2O and O_2 concentrations along the soil depth under different soil moisture conditions at 12

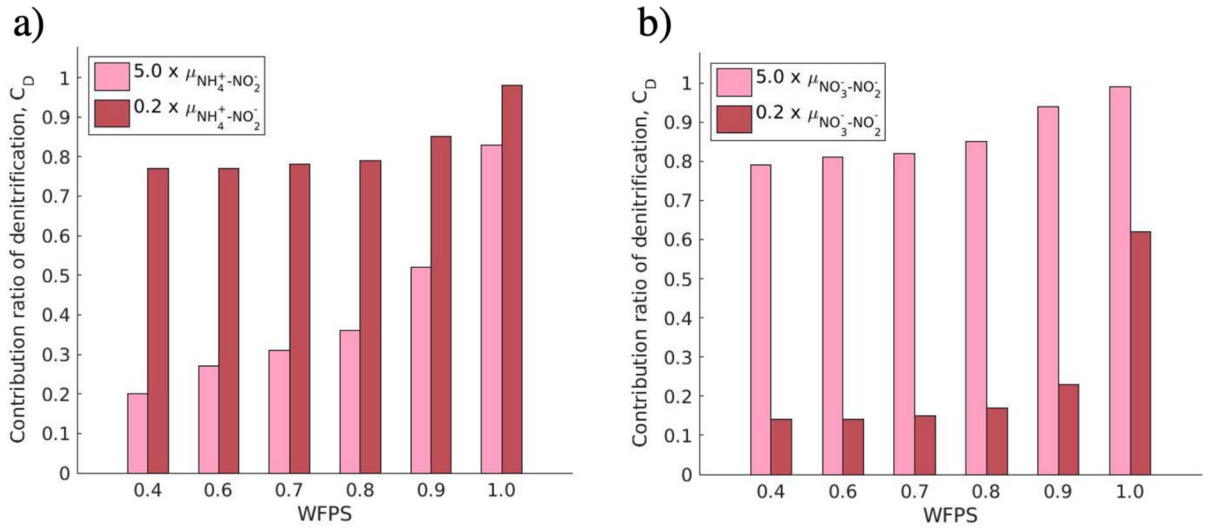

Fig. 4 Simulated contribution ratio of denitrification to N_2O production, C_D, by the PBM-T-NF model as **a** $\mu_{NH_4^+-NO_2^-}$ and **b** $\mu_{NO_3^--NO_2^-}$ were increased (5.0 ×) or decreased (0.2 ×) by five times. The LC soil was used to evaluate the effects of model parameters

and 24 h. The NH_4^+ was depleted rapidly throughout the soil profile under relatively low moisture conditions, and was mainly consumed in the top soil under high moisture conditions (Fig. 7a). Conversely, NO_3^- was mainly consumed at the bottom of the soil profile under high moisture conditions (Fig. 7b). Correspondingly, N_2O concentration is high under high moisture conditions (Fig. 7c), while O_2 concentration is high under low moisture conditions (Fig. 7d).

Discussion

Performance of the developed models

The model with transport (PBM-T-NF) well predicted the concentrations of different N species under a wide range of soil moisture conditions. In particular, the PBM-T-NF model almost exactly predicted C_D under relatively high moisture conditions in the SZ soil, illustrating the robustness and accuracy of our proposed conceptual model based on N flows through the reaction pathways of nitrification and denitrification. By contrast, the model without transport (PBM-NF) could not capture the increasing trends in C_D with increasing soil moisture content, and failed to predict the rapid changes in N_2O concentrations around the maximum values, although the studied soil core is shallow (i.e., 8 mm). The results illustrate the necessity of considering mass transport in the simulation of soil N processes (Gilhespy et al. 2014; Tian et al. 2019). In particular, in contrast with the model neglecting diffusion, the model with transport predicted the N_2O emissions under high moisture contents much better, since the latter successfully captured the sharp decrease in O_2 concentration along the soil profile, which favored denitrification and N_2O production (Rohe et al. 2021). The large N_2O fluxes have been widely demonstrated difficult to be predicted by models (Abdalla et al. 2020; Klier et al. 2011; Yue et al. 2019). Incorporating mass transport in these models may improve the prediction of N_2O emissions from soils.

The developed model assumed that NO_2^- derived from nitrification and denitrification was reduced indistinguishably to NO and then to N_2O by nitrifiers and denitrifiers. This explains why the simulated C_D was mainly sensitive to $\mu_{NH_4^+-NO_2^-}$ and $\mu_{NO_3^--NO_2^-}$, whose values are crucial to control the contributing N flows from nitrification or denitrification. This assumption is appropriate in quasi-stationary conditions, where N_2O is produced by either nitrification or denitrification (Zhu et al. 2013), and is also the theoretical basis for the enriched isotopic approaches

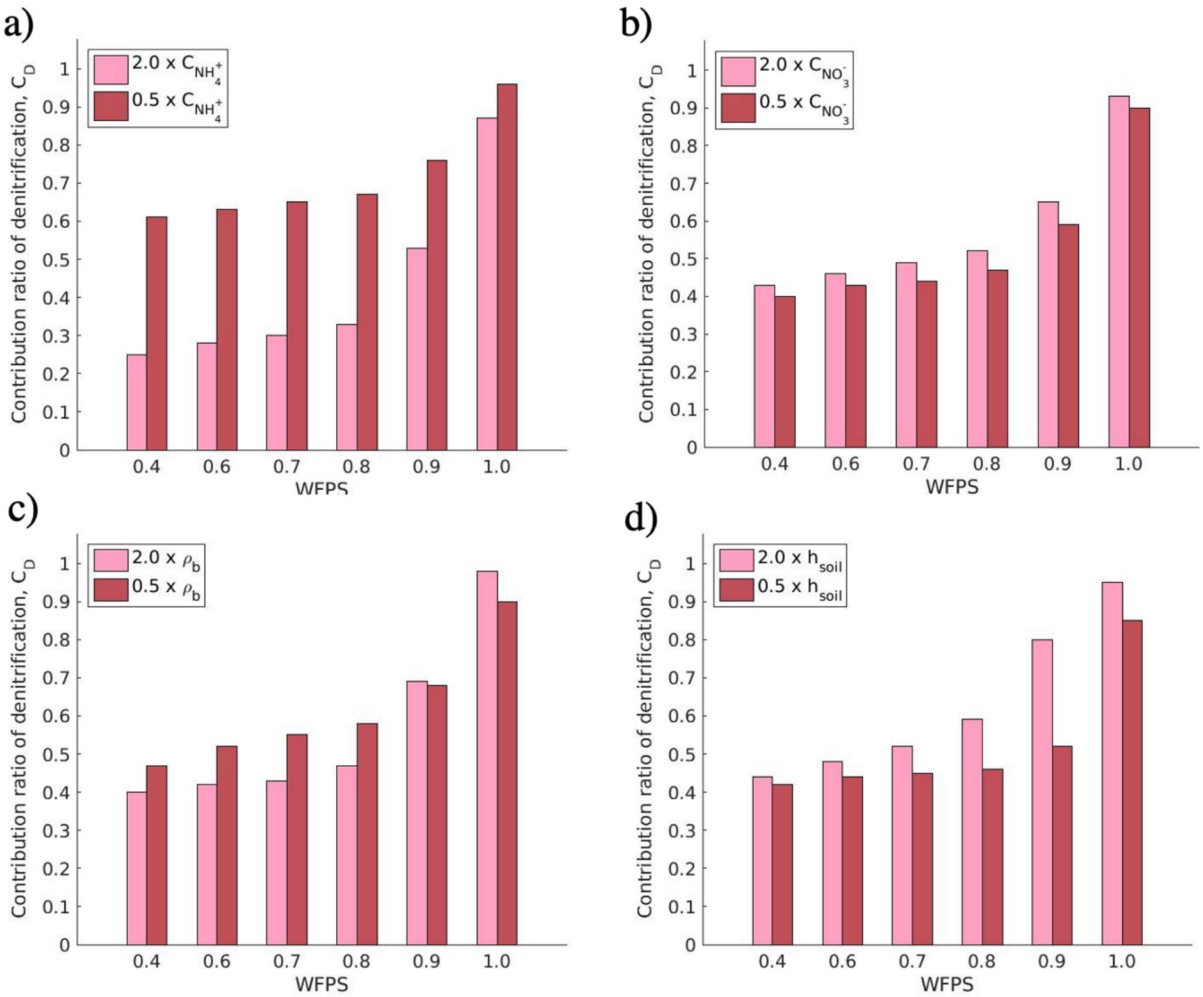

Fig. 5 Simulated contribution ratio of denitrification to N_2O production, C_D, by the PBM-T-NF model as **a** NH_4^+ concentration, **b** NO_3^- concentration, **c** bulk density, or **d** soil depth was doubled (2.0 ×) or halved (0.5 ×). The LC soil was used to evaluate the effects of soil conditions

(Bateman and Baggs 2005; Wang et al. 2023; Zhu et al. 2013). However, in dynamic environments such as riverine or coastal wetlands with fluctuating groundwater tables, NO_2^- produced by nitrification under aerobic conditions is often reduced by denitrification under anerobic conditions (Deegan et al. 2012; Li et al. 2023). The coupled nitrification and denitrification process may be important for N_2O production in these land–water transition zones (Baggs 2011; Butterbach-Bahl et al. 2013), and complicates the modeling accuracy of our developed models.

Effects of soil conditions on N_2O sources and emissions

The PBM-T-NF model provides a feasible way to evaluate the effects of soil conditions on N_2O sources. NH_4^+ concentration is found to be the key to regulating C_D in the soils studied (Fig. 5a), mainly because NH_4^+ is the rate-limiting substrate for nitrification, whose product (i.e., NO_3^-) is substrate of denitrification (Li et al. 2024). If another soil with insufficient NO_3^- is used, NO_3^- may outperform NH_4^+ in regulating C_D (Harris et al. 2021). Therefore, the baseline soil chemistry is critical to controlling the source of N_2O. In addition, elevated NH_4^+ concentration not

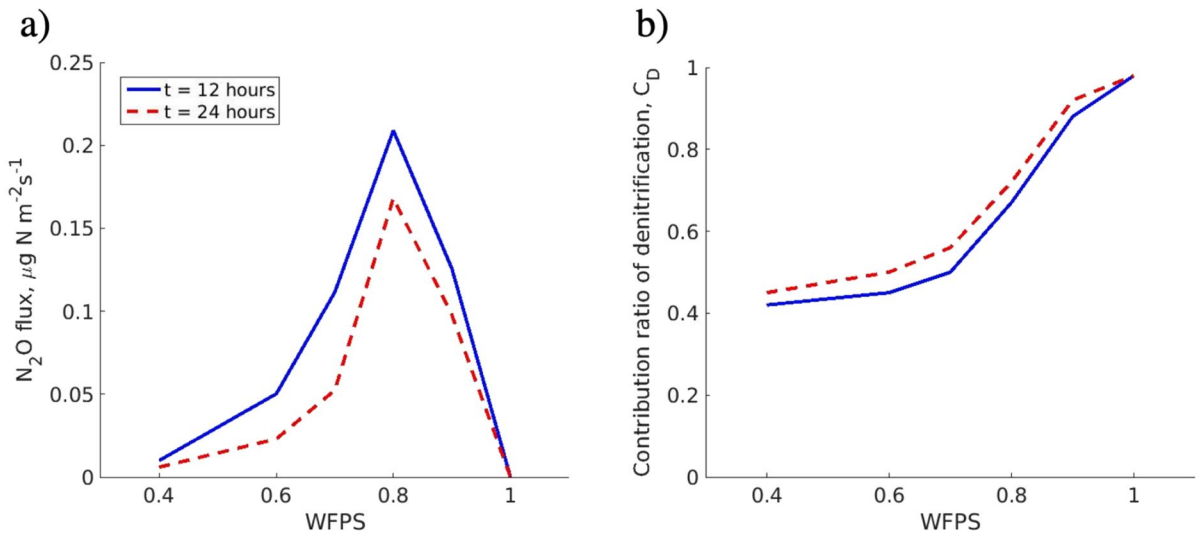

Fig. 6 Simulated **a** N_2O fluxes and **b** contribution of denitrification to N_2O production, C_D, by the PBM-T-NF model in the artificial soil core under different soil moisture conditions at 12 and 24 h

only directly increases N_2O production by promoting nitrification, but also indirectly stimulates it via denitrification by depleting O_2, as NH_4^+ oxidation consumes O_2 and creates preferential conditions for denitrification (Song et al. 2019; Yang et al. 2021). This stimulation can become significant under high soil moisture conditions, where O_2 diffusion is limited (Smith 2017), and partially explains why the N_2O flux increased by 6.8-folds at WFPS = 0.9, though NH_4^+ concentration increased only by 4-folds (Fig. S2a).

It is plausible to assume that soil conditions regulating N_2O production and its emission into the atmosphere interact. For example, elevated soil bulk density stimulated nitrification by increasing the rate-limiting NH_4^+ concentration under low moisture conditions, resulting in a smaller C_D (Fig. 5c). However, the elevated bulk density increased C_D under high moisture conditions by promoting denitrification, because it decreased O_2 diffusion into soils (Yan et al. 2016). The moisture-dependent effects of soil bulk density illustrate that the conclusions derived from controlled experiments under optimal moisture levels should be interpreted carefully in the field (Huang et al. 2015; Li et al. 2024). It is worth noting that the effect of soil compaction on N_2O emission is currently attracting much attention (Ren et al. 2020). Similarly, increasing soil depth increased C_D by promoting denitrification, as it created a more anaerobic soil layer favorable for denitrification at the bottom of the soil core. The magnitude of the depth effect also depended on soil moisture content and was maximized at relatively high moisture contents (Fig. 5d), as O_2 was not limited at low soil moisture contents and N_2O was reduced to N_2 under high moisture contents (Wang et al. 2020b). Therefore, multifactorial experiments with varying soil moisture are essential to unravel the underlying mechanisms behind the spatial and temporal changes in N_2O fluxes and sources.

Effects of mass transport on N_2O sources

The large differences between the simulation results from the PBM-T-NF model and the PBM-NF model underscore the importance of mass transport within soils, as do the simulated discrepancies from the shallow soil and the artificial soil core. For example, the simulated C_D from the soil core is apparently increased at WFPS = 0.8 and 0.9 (Fig. 6b), compared to the results from the shallow soil. This is mainly because O_2 concentration was depleted at the bottom of the soil core, which strongly stimulated denitrification (Fig. 7 and Fig. S2). Compared with the shallow soil, the greater depth in the soil core also shifted the optimal moisture, under which the maximum N_2O

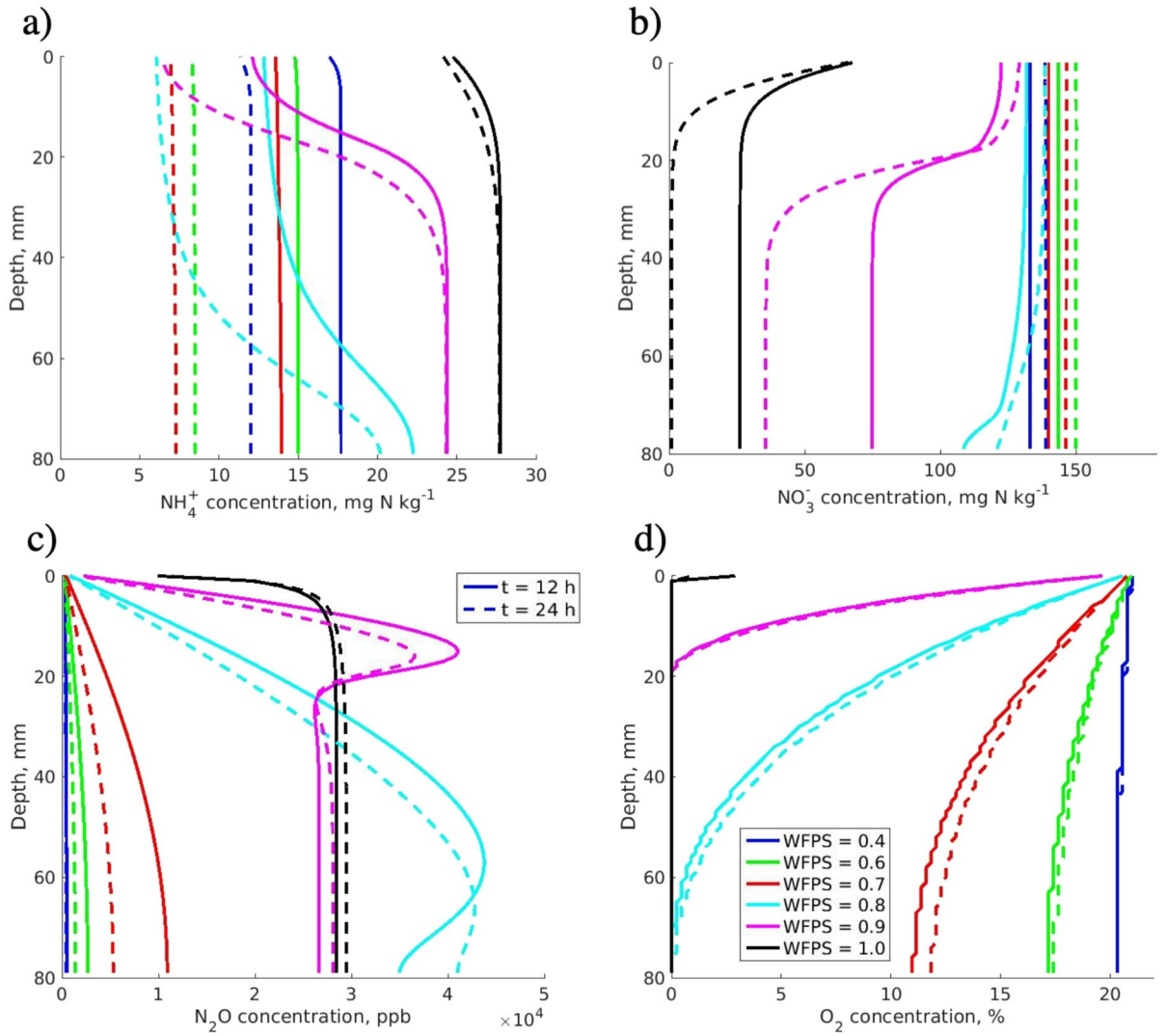

Fig. 7 Simulated profile concentrations of **a** total NH_4^+, **b** dissolved NO_3^-, **c** gaseous N_2O, and **d** gaseous O_2 by the PBM-T-NF model along the artificial soil core under different soil moisture conditions

flux occurred, from WFPS = 0.9 to 0.8, as it further reduced N_2O to N_2 via complete denitrification under the high moisture conditions (Fig. 7 and Fig. S2) (Hu et al. 2015; Xia et al. 2018). The anaerobic condition at the bottom of the soil core also explains why the N_2O flux approached zero at WFPS = 1.0 (Fig. 6a). The decline in gas diffusivity due to elevated soil moisture content has been found to well explain N_2O fluxes under different soil moisture conditions (Chamindu Deepagoda et al. 2019). The incorporation of gas diffusion in models have been found to improve the prediction of N_2O emissions especially under high precipitation (Klier et al. 2011). Furthermore, soil water movement (i.e., advection) has been reported to affect N cycling and N_2O emissions by modulating substrate availability and moisture distribution (Gao et al. 2023), and its inclusion in process-based models could improve the simulation accuracy of N processes (Smith et al. 2020). Our model should also account for water movement in the future especially under high moisture conditions, where water is supposed to move downward due to gravity.

Soil N_2O emissions into the atmosphere are a function of N_2O production, transport and consumption

(Klier et al. 2011; Signor and Cerri 2013). A large number of studies have investigated N_2O production under different environmental conditions by using different types of soils (Bateman and Baggs 2005; Wang et al. 2023; Zhu et al. 2013). While N_2O production pathways are relatively better understood, the effects of transport on N_2O emissions are poorly understood, mainly because soil is an invisible and complex matrix containing solid, water, and gaseous phases (Rabot et al. 2018; Yan et al. 2023). Solute transport in soils regulates N_2O production through affecting substrate availability (Kravchenko et al. 2017), while gas transport in soils affects N_2O consumption by determining its residence time (Chang et al. 2022; Niu et al. 2016). Furthermore, the gas transport determines both N_2O production and consumption by controlling O_2 availability (Van der Weerden et al. 2012). Therefore, mass transport is extremely important for soil N_2O emissions, and it is regulated by many factors including soil structure and moisture content (Kravchenko et al. 2017). Although the well-mixed soils (without structure) were employed to simulate N_2O emissions, the large difference between the simulation results from the developed models with and without transport indicate the importance of mass transport. This importance is expected to increase for N_2O emissions from natural soils, which contain structures such as different sizes of aggregates and complex pore connectivity (Van der Weerden et al. 2012; Fu et al. 2024).

Model limitations and future directions

The PBM-T-NF model provides a feasible way to quantify N_2O sources, overcoming the shortcomings of isotopic approaches, which are costly and subject to uncertainty (Denk et al. 2017). However, the model we developed focuses on nitrification and denitrification, which can lead to misleading results due to the incomplete N processes (Yan et al. 2024). For example, dissimilatory nitrate reduction to ammonium (DNRA) and anaerobic ammonium oxidation (ANAMMOX) may be important sources of N_2O under high moisture conditions (Shi et al. 2024), and their omission may lead to an overestimation of the N_2O contribution from nitrification or denitrification. Besides, abiotic processes including NH_2OH decomposition and chemodenitrification may contribute significantly to N_2O production (Zhu-Barker et al. 2015). Nitrogen mineralization and assimilation can also modulate N_2O production by regulating substrate and O_2 availability (Xu et al. 2024; Yan et al. 2024; Zhang et al. 2022, 2018). Therefore, future models should incorporate these N processes in quantifying N_2O sources by tracking N flows in the corresponding reaction pathways, similar to what we did in this study. By combing with more sensitive experiments, including dual isotope approaches and site preference techniques (Kool et al. 2007; Wei et al. 2023), the advanced model can be calibrated and the comprehensive N processes are simulated more accurately.

Although the PBM-T-NF model was well validated on independent soils in this study, the model should be tested on more different types of soil in the future. For example, the contribution of denitrification to N_2O in acid soils may be significantly different from the alkaline soils as used in this study (Kool et al. 2011; Zhu et al. 2013), given that pH significantly impacts the community structures of nitrifiers and denitrifiers (Han et al. 2024). In addition, more experiments investigating the effects of soil and environmental conditions are needed to further constrain model parameters, and the dynamics of N processes should be experimentally quantified. Once our model is calibrated and validated by more experimental data using different soils and under various environmental conditions, it has the potential to be applied to a wide range of soil types and to capture the spatiotemporal variability of N_2O sources, which can guide us to take more precise measures to mitigate N_2O emissions from soils.

Conclusions

We developed process-based models to quantify N_2O attributions from nitrification and denitrification by tracking the N flows through the reaction pathways. Compared to the model without transport, the model accounting for solute and gas diffusions better predicted the N_2O fluxes and sources under a wide range of soil moisture levels in two different soils, highlighting the importance of including mass transport in predicting N_2O emissions. Therefore, combining N_2O production, transport, and consumption in process-based models is able to improve prediction of N_2O emissions. Furthermore, the effects of soil conditions on N_2O sources were found to depend on substrate

availability and moisture status. Multifactorial experiments and modeling are needed to unravel the mechanisms underlying the large spatial and temporal variabilities in soil-to-atmosphere N_2O fluxes. Overall, we provide a feasible way to quantify N_2O production from nitrification and denitrification, complementing current experimental approaches in the study of N processes.

Acknowledgements We would like to thank Dr. Christoph Müller from the Justus Liebig University Giessen, Institute of Plant Ecology, Germany, for constructive suggestions. This work was financially supported by the National Natural Science Foundation of China (42293262, 42077009), the National Key Research and development Program of China (2022YFF1301002), and Haihe Laboratory of Sustainable Chemical Transformations and Tianjin Municipal Science and Technology Bureau (No. 21ZYJDJC00090). K.B.B. received additional funding by the Pioneer Center for Research in Sustainable Agricultural Futures (Land-CRAFT), DNRF grant number P2, Aarhus University, Denmark. N.J.S. acknowledges the financial support from EU projects (SUS-SOIL, NPower and CSR).

Author contributions All co-authors contributed to the study. Z. Y. incubated the idea, wrote the numerical codes, run the simulations, and wrote the first draft. Z. C. helped to analyze the data, and all authors contributed to improve the manuscript.

Funding The authors have not disclosed any funding.

Data availability The data used in this study are publicly available in the Zenodo repository at https://doi.org/10.5281/zenodo.15230929.

Declarations

Conflict of interest The authors declare no conflict of interest.

Open Access This article is licensed under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International License, which permits any non-commercial use, sharing, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if you modified the licensed material. You do not have permission under this licence to share adapted material derived from this article or parts of it. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by-nc-nd/4.0/.

References

Abdalla M, Song X, Ju X, Topp CFE, Smith P (2020) Calibration and validation of the DNDC model to estimate nitrous oxide emissions and crop productivity for a summer maize-winter wheat double cropping system in Hebei, China. Environ Pollut 262:114199

Baggs EM (2011) Soil microbial sources of nitrous oxide: recent advances in knowledge, emerging challenges and future direction. Curr Opin Environ Sustain 3(5):321–327

Bateman EJ, Baggs EM (2005) Contributions of nitrification and denitrification to N_2O emissions from soils at different water-filled pore space. Biol Fertil Soils 41(6):379–388

Brooks S (1998) Markov chain Monte Carlo method and its application. J R Stat Soc Ser D Stat 47(1):69–100

Brooks P, Stark JM, McInteer B, Preston T (1989) Diffusion method to prepare soil extracts for automated nitrogen-15 analysis. Soil Sci Soc Am J 53(6):1707–1711

Butterbach-Bahl K, Baggs EM, Dannenmann M, Kiese R, Zechmeister-Boltenstern S (2013) Nitrous oxide emissions from soils: How well do we understand the processes and their controls? Philos Trans R Soc B Biol Sci 368(1621):20130122

Chamindu Deepagoda TKK, Jayarathne JRRN, Clough TJ, Thomas S, Elberling B (2019) Soil–gas diffusivity and soil–moisture effects on NO emissions from intact pasture soils. Soil Sci Soc Am J 83(4):1032–1043

Chang B, Yan Z, Ju X, Song X, Li Y, Li S, Fu P, Zhu-Barker X (2022) Quantifying biological processes producing nitrous oxide in soil using a mechanistic model. Biogeochemistry 159:1–14

Davidson EA, Keller M, Erickson HE, Verchot LV, Veldkamp E (2000) Testing a conceptual model of soil emissions of nitrous and nitric oxides. Bioscience 50(8):667–680

Deegan LA, Johnson DS, Warren RS, Peterson BJ, Fleeger JW, Fagherazzi S, Wollheim WM (2012) Coastal eutrophication as a driver of salt marsh loss. Nature 490(7420):388–392

Del Grosso SJ, Parton WJ, Mosier AR, Ojima DS, Kulmala AE, Phongpan S (2000) General model for N_2O and N_2 gas emissions from soils due to dentrification. Glob Biogeochem Cycles 14(4):1045–1060

Del Grosso SJ, Smith W, Kraus D, Massad RS, Vogeler I, Fuchs K (2020) Approaches and concepts of modelling denitrification: increased process understanding using observational data can reduce uncertainties. Curr Opin Environ Sustain 47:37–45

Denk TRA, Mohn J, Decock C, Lewicka-Szczebak D, Harris E, Butterbach-Bahl K, Kiese R, Wolf B (2017) The nitrogen cycle: a review of isotope effects and isotope modeling approaches. Soil Biol Biochem 105:121–137

Ehrhardt F, Soussana J-F, Bellocchi G, Grace P, McAuliffe R, Recous S, Sándor R, Smith P, Snow V, de Antoni MM, Basso B, Bhatia A, Brilli L, Doltra J, Dorich CD, Doro L, Fitton N, Giacomini SJ, Grant B, Harrison MT, Jones SK, Kirschbaum MUF, Klumpp K, Laville P, Léonard J, Liebig M, Lieffering M, Martin R, Massad RS, Meier E, Merbold L, Moore AD, Myrgiotis V, Newton P, Pattey E, Rolinski S, Sharp J, Smith WN, Wu L, Zhang Q (2018) Assessing uncertainties in crop and pasture ensemble

model simulations of productivity and N_2O emissions. Glob Change Biol 24(2):e603–e616
Friedl J, Scheer C, De Rosa D, Müller C, Grace PR, Rowlings DW (2021) Sources of nitrous oxide from intensively managed pasture soils: the hole in the pipe. Environ Res Lett 16(6):065004
Fu Z, Yan Z, Li S-l (2024) Effects of soil pore structure on gas diffusivity under different land uses: characterization and modelling. Soil Tillage Res 237:105988
Gao Y, Tian Y, Zhan W, Li L, Sun H, Zhao T, Zhang H, Meng Y, Li Y, Liu T, Ding J (2023) Characterizing legacy nitrogen-induced time lags in riverine nitrogen reduction for the Songhuajiang River Basin: source analysis, spatio-seasonal patterns, and impacts on future water quality improvement. Water Res 242:120292
Gilhespy SL, Anthony S, Cardenas L, Chadwick D, Del Prado A, Li C, Misselbrook T, Rees RM, Salas W, Sanz-Cobena A, Smith P, Tilston EL, Topp CFE, Vetter S, Yeluripati JB (2014) First 20 years of DNDC (DeNitrification DeComposition): model evolution. Ecol Model 292:51–62
Groffman PM, Altabet MA, Böhlke H, Butterbach-Bahl K, David MB, Firestone MK, Giblin AE, Kana TM, Nielsen LP, Voytek MA (2006) Methods for measuring denitrification: diverse approaches to a difficult problem. Ecol Appl 16(6):2091–2122
Hamamoto S, Moldrup P, Kawamoto K, Komatsu T (2010) Excluded-volume expansion of Archie's law for gas and solute diffusivities and electrical and thermal conductivities in variably saturated porous media. Water Resour Res 46(6):6514
Han B, Yao Y, Liu B, Wang Y, Su X, Ma L, Liu D, Niu S, Chen X, Li Z (2024) Relative importance between nitrification and denitrification to N_2O from a global perspective. Global Change Biol 30(1):e17082
Hansen S (2002) Daisy, a flexible soil–plant–atmosphere system model. Report Dept Agric 615:1
Harris E, Diaz-Pines E, Stoll E, Schloter M, Schulz S, Duffner C, Li K, Moore KL, Ingrisch J, Reinthaler D, Zechmeister-Boltenstern S, Glatzel S, Brüggemann N, Bahn M (2021) Denitrifying pathways dominate nitrous oxide emissions from managed grassland during drought and rewetting. Sci Adv 7(6):eabb7118
Harris E, Yu L, Wang YP, Mohn J, Henne S, Bai E, Barthel M, Bauters M, Boeckx P, Dorich C, Farrell M, Krummel PB, Loh ZM, Reichstein M, Six J, Steinbacher M, Wells NS, Bahn M, Rayner P (2022) Warming and redistribution of nitrogen inputs drive an increase in terrestrial nitrous oxide emission factor. Nat Commun 13(1):4310
Heinen M (2006) Simplified denitrification models: overview and properties. Geoderma 133(3):444–463
Hu H, Chen D, He J (2015) Microbial regulation of terrestrial nitrous oxide formation: understanding the biological pathways for prediction of emission rates. FEMS Microbiol Rev 39(5):729–749
Huang T, Gao B, Hu X, Lu X, Well R, Christie P, Bakken LR, Ju X (2015) Ammonia-oxidation as an engine to generate nitrous oxide in an intensively managed calcareous Fluvo-aquic soil. Sci Rep 4(1):03950
IPCC (2021) Climate change 2021
Jansen-Willems AB, Zawallich J, Müller C (2022) Advanced tool for analysing ^{15}N tracing data. Soil Biol Biochem 165:108532
Klier C, Gayler S, Haberbosch C, Ruser R, Stenger R, Flessa H, Priesack E (2011) Modeling nitrous oxide emissions from potato-cropped soil. Vadose Zone Journal 10(1):184–194
Kool DM, Wrage N, Oenema O, Dolfing J, Van Groenigen JW (2007) Oxygen exchange between (de)nitrification intermediates and H_2O and its implications for source determination of NO_3^- and N_2O: a review. Rapid Commun Mass Spectrom 21(22):3569–3578
Kool DM, Dolfing J, Wrage N, Van Groenigen JW (2011) Nitrifier denitrification as a distinct and significant source of nitrous oxide from soil. Soil Biol Biochem 43(1):174–178
Kravchenko AN, Toosi ER, Guber AK, Ostrom NE, Yu J, Azeem K, Rivers ML, Robertson GP (2017) Hotspots of soil N_2O emission enhanced through water absorption by plant residue. Nat Geosci 10:496–500
Laville P, Lehuger S, Loubet B, Chaumartin F, Cellier P (2011) Effect of management, climate and soil conditions on N_2O and NO emissions from an arable crop rotation using high temporal resolution measurements. Agri For Meteorol 151(2):228–240
Li C, Aber J, Stange F, Butterbach-Bahl K, Papen H (2000) A process-oriented model of N_2O and NO emissions from forest soils: 1. Model development. J Geophys Res Atmos 105(D4):4369–4384
Li X, Gao D, Li Y, Zheng Y, Dong H, Liang X, Liu M, Hou L (2023) Increased nitrogen loading facilitates nitrous oxide production through fungal and chemodenitrification in estuarine and coastal sediments. Environ Sci Technol 57(6):2660–2671
Li Y, Wang Z, Ju X, Wu D (2024) Disproportional oxidation rates of ammonia and nitrite deciphers the heterogeneity of fertilizer-induced N_2O emissions in agricultural soils. Soil Biol Biochem 191:109325
Lucas M, Gil J, Robertson GP, Ostrom NE, Kravchenko A (2023) Changes in soil pore structure generated by the root systems of maize, sorghum and switchgrass affect in situ N_2O emissions and bacterial denitrification. Biol Fertil Soils. https://doi.org/10.1007/s00374-023-01761-1
Maggi F, Gu C, Riley WJ, Hornberger GM, Venterea RT, Xu T, Spycher N, Steefel C, Miller NL, Oldenburg CM (2008) A mechanistic treatment of the dominant soil nitrogen cycling processes: model development, testing, and application. J Geophys Res Biogeosci 113(G2):G02016
Müller C, Laughlin RJ, Spott O, Rütting T (2014) Quantification of N_2O emission pathways via a ^{15}N tracing model. Soil Biol Biochem 72:44–54
Niu S, Classen AT, Dukes JS, Kardol P, Liu L, Luo Y, Rustad L, Sun J, Tang J, Templer PH, Thomas RQ, Tian D, Vicca S, Wang Y-P, Xia J, Zaehle S (2016) Global patterns and substrate-based mechanisms of the terrestrial nitrogen cycle. Ecol Lett 19(6):697–709
Parton WJ, Mosier AR, Ojima DS, Valentine DW, Schimel DS, Weier K, Kulmala AE (1996) Generalized model for N_2 and N_2O production from nitrification and denitrification. Global Biogeochem Cycles 10(3):401–412
Rabot E, Cousin I, Hénault C (2015) A modeling approach of the relationship between nitrous oxide fluxes from

soils and the water-filled pore space. Biogeochemistry 122(2–3):395–408
Rabot E, Wiesmeier M, Schlüter S, Vogel HJ (2018) Soil structure as an indicator of soil functions: a review. Geoderma 314:122–137
Ren X, Zhu B, Bah H, Raza ST (2020) How tillage and fertilization influence soil N_2O emissions after forestland conversion to cropland. Sustainability 12(19):7947
Rohe L, Apelt B, Vogel H-J, Well R, Wu G-M, Schlüter S (2021) Denitrification in soil as a function of oxygen availability at the microscale. Biogeosciences 18(3):1185–1201
Ruser R, Flessa H, Russow R, Schmidt G, Buegger F, Munch JC (2006) Emission of N_2O, N_2 and CO_2 from soil fertilized with nitrate: effect of compaction, soil moisture and rewetting. Soil Biol Biochem 38(2):263–274
Sander R (2015) Compilation of Henry's law constants (version 4.0) for water as solvent. Atmos Chem Phys 15(8):4399–4981
Shi Z, She D, Pan Y, Abulaiti A, Huang Y, Liu R, Wang F, Xia Y, Shan J (2024) Ditch level-dependent N removal capacity of denitrification and anammox in the drainage system of the Ningxia Yellow River irrigation district. Sci Total Environ 916:170314
Signor D, Cerri CEP (2013) Nitrous oxide emissions in agricultural soils: a review. Pesquisa Agropecuária Tropical 43(3):322–338
Smith KA (2017) Changing views of nitrous oxide emissions from agricultural soil: key controlling processes and assessment at different spatial scales. Eur J Soil Sci 68(2):137–155
Smith W, Grant B, Qi Z, He W, VanderZaag A, Drury CF, Helmers M (2020) Development of the DNDC model to improve soil hydrology and incorporate mechanistic tile drainage: a comparative analysis with RZWQM2. Environ Model Softw 123:104577
Song X, Ju X, Topp CFE, Rees RM (2019) Oxygen regulates nitrous oxide production directly in agricultural soils. Environ Sci Technol 53(21):12539–12547
Stevens RJ, Laughlin RJ, Burns LC, Arah JRM, Hood RC (1997) Measuring the contributions of nitrification and denitrification to the flux of nitrous oxide from soil. Soil Biol Biochem 29(2):139–151
Stumm W, Morgan JJ (1996) Aquatic chemistry: chemical equilibria and rates in natural waters. Wiley, New York
Tian H, Yang J, Lu C, Xu R, Canadell JG, Jackson RB, Arneth A, Chang J, Chen G, Ciais P (2018) The global N_2O model intercomparison project. Bull Am Meteorol Soc 99(6):1231–1251
Tian H, Yang J, Xu R, Lu C, Canadell JG, Davidson EA, Jackson RB, Arneth A, Chang J, Ciais P, Gerber S, Ito A, Joos F, Lienert S, Messina P, Olin S, Pan S, Peng C, Saikawa E, Thompson RL, Vuichard N, Winiwarter W, Zaehle S, Zhang B (2019) Global soil nitrous oxide emissions since the preindustrial era estimated by an ensemble of terrestrial biosphere models: magnitude, attribution, and uncertainty. Global Change Biol 25(2):640–659
Tian H, Xu R, Canadell JG, Thompson RL, Winiwarter W, Suntharalingam P, Davidson EA, Ciais P, Jackson RB, Janssens-Maenhout G, Prather MJ, Regnier P, Pan N, Pan S, Peters GP, Shi H, Tubiello FN, Zaehle S, Zhou F, Arneth A, Battaglia G, Berthet S, Bopp L, Bouwman AF, Buitenhuis ET, Chang J, Chipperfield MP, Dangal SRS, Dlugokencky E, Elkins JW, Eyre BD, Fu B, Hall B, Ito A, Joos F, Krummel PB, Landolfi A, Laruelle GG, Lauerwald R, Li W, Lienert S, Maavara T, Macleod M, Millet DB, Olin S, Patra PK, Prinn RG, Raymond PA, Ruiz DJ, Van Der Werf GR, Vuichard N, Wang J, Weiss RF, Wells KC, Wilson C, Yang J, Yao Y (2020) A comprehensive quantification of global nitrous oxide sources and sinks. Nature 586(7828):248–256
Van der Weerden TJ, Kelliher FM, de Klein CAM (2012) Influence of pore size distribution and soil water content on nitrous oxide emissions. Soil Research 50(2):125–135
Vinten A, Castle K, Jrm A (1996) Field evaluation of models of denitrification linked to nitrate leaching for aggregated soil. Eur J Soil Sci 47(3):305–317
Wang Q, Zhou F, Shang Z, Ciais P, Winiwarter W, Jackson RB, Tubiello FN, Janssens-Maenhout G, Tian H, Cui X, Canadell JG, Piao S, Tao S (2020a) Data-driven estimates of global nitrous oxide emissions from croplands. Natl Sci Rev 7(2):441–452
Wang R, Pan Z, Zheng X, Ju X, Yao Z, Butterbach-Bahl K, Zhang C, Wei H, Huang B (2020b) Using field-measured soil N_2O fluxes and laboratory scale parameterization of $N_2O/(N_2O+N_2)$ ratios to quantify field-scale soil N_2 emissions. Soil Biol Biochem 148:107904
Wang C, Amon B, Schulz K, Mehdi B (2021) Factors that influence nitrous oxide emissions from agricultural soils as well as their representation in simulation models: a review. Agronomy 11(4):770
Wang H, Yan Z, Ju X, Song X, Zhang J, Li S, Zhu-Barker X (2023) Quantifying nitrous oxide production rates from nitrification and denitrification under various moisture conditions in agricultural soils: laboratory study and literature synthesis. Front Microbiol. https://doi.org/10.3389/fmicb.2022.1110151
Wang H, Yan Z, Chen Z, Song X, Zhang J, Li S-L, Müller C, Ju X, Zhu-Barker X (2024) Microbial ammonium immobilization promoted soil nitrogen retention under high moisture conditions in intensively managed fluvo-aquic soils. Biol Fertil Soils. https://doi.org/10.1007/s00374-024-01831-y
Watts SH, Seitzinger SP (2000) Denitrification rates in organic and mineral soils from riparian sites: a comparison of N_2 flux and acetylene inhibition methods. Soil Biol Biochem 32(10):1383–1392
Wei H, Song X, Liu Y, Wang R, Zheng X, Butterbach-Bahl K, Venterea RT, Wu D, Ju X (2023) In situ 15N–N2O site preference and O2 concentration dynami. Global Change Biol 2023:1
Xia L, Lam SK, Wolf B, Kiese R, Chen D, Butterbach-Bahl K (2018) Trade-offs between soil carbon sequestration and reactive nitrogen losses under straw return in global agro-ecosystems. Global Change Biol 24(12):5919–5932
Xu P, Li G, Zheng Y, Fung JCH, Chen A, Zeng Z, Shen H, Hu M, Mao J, Zheng Y, Cui X, Guo Z, Chen Y, Feng L, He S, Zhang X, Lau AKH, Tao S, Houlton BZ (2024) Fertilizer management for global ammonia emission reduction. Nature 626(8000):792–798
Yan Z, Liu C, Todd-Brown KE, Liu Y, Bond-Lamberty B, Bailey VL (2016) Pore-scale investigation on the response of

heterotrophic respiration to moisture conditions in heterogeneous soils. Biogeochemistry 131(1):121–134

Yan Z, Bond-Lamberty B, Todd-Brown KE, Bailey VL, Li S, Liu C, Liu C (2018a) A moisture function of soil heterotrophic respiration that incorporates microscale processes. Nat Commun 9(1):2562

Yan Z, Wang T, Wang L, Yang X, Smith P, Hilpert M, Li S, Shang J, Bailey V, Liu C (2018b) Microscale water distribution and its effects on organic carbon decomposition in unsaturated soils. Sci Total Environ 644:1036–1043

Yan Z, Wang Z, Fu Z, Zhang Y, Peng X, Zheng J (2023) Microscale heterogeneity controls macroscopic soil heterotrophic respiration by regulating resource availability and environmental stress. Biogeochemistry. https://doi.org/10.1007/s10533-023-01044-9

Yan Z, Chang B, Song X, Wang G, Shan J, Yang L, Li S-l, Butterbach-Bahl K, Ju X (2024) A microbial-explicit model with comprehensive nitrogen processes to quantify gaseous nitrogen production from agricultural soils. Soil Biol Biochem 189:109284

Yang Y, Zhang H, Shan Y, Wang J, Qian X, Meng T, Zhang J, Cai Z (2019) Response of denitrification in paddy soils with different nitrification rates to soil moisture and glucose addition. Sci Total Environ 651:2097–2104

Yang L, Zhang X, Ju X, Wu D (2021) Oxygen-depletion by rapid ammonia oxidation regulates kinetics of N_2O, NO and N_2 production in an ammonium fertilised agricultural soil. Soil Biol Biochem 163:108460

Yue Q, Cheng K, Ogle S, Hillier J, Smith P, Abdalla M, Ledo A, Sun J, Pan G (2019) Evaluation of four modelling approaches to estimate nitrous oxide emissions in China's cropland. Sci Total Environ 652:1279–1289

Zhang J, Müller C, Cai Z (2015) Heterotrophic nitrification of organic N and its contribution to nitrous oxide emissions in soils. Soil Biol Biochem 84:199–209

Zhang Y, Ding H, Zheng X, Cai Z, Misselbrook T, Carswell A, Müller C, Zhang J (2018) Soil N transformation mechanisms can effectively conserve N in soil under saturated conditions compared to unsaturated conditions in subtropical China. Biol Fertil Soils 54(4):495–507

Zhang B, Zhou M, Zhu B, Xiao Q, Zheng X, Zhang J, Müller C, Butterbach-Bahl K (2022) Soil clay minerals: an overlooked mediator of gross N transformations in Regosolic soils of subtropical montane landscapes. Soil Biol Biochem 168:108612

Zheng J, Fujii K, Koba K, Wanek W, Müller C, Jansen-Willems AB, Nakajima Y, Wagai R, Canarini A (2023) Revisiting process-based simulations of soil nitrite dynamics: tighter cycling between nitrite and nitrate than considered previously. Soil Biol Biochem 178:108958

Zhu X, Burger M, Doane TA, Horwath WR (2013) Ammonia oxidation pathways and nitrifier denitrification are significant sources of N_2O and NO under low oxygen availability. Proc Natl Acad Sci 110(16):6328–6333

Zhu-Barker X, Cavazos AR, Ostrom NE, Horwath WR, Glass JB (2015) The importance of abiotic reactions for nitrous oxide production. Biogeochemistry 126(3):251–267

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.