Using data mining to improve the prediction of key nitrogen loss from terrestrial ecosystems

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To my family…

ABSTRACT

Nitrogen (N) losses through nitrification, denitrification and nitrate leaching in terrestrial ecosystems reduce fertiliser N use efficiency (NUE), affect environmental quality and human health. In the past 50 years, many individual studies both on-site and in laboratories have been carried out to investigate N loss from different pathways. However, field measurements of N dynamics are generally time-consuming, expensive and some of the processes are quite difficult to measure. Process-based models have been developed to better understand the complex, multivariate and unpredictable N cycling by integrating soil, environmental and management factors. However, the performances of these models are restricted by data availability, inconsistent responses of N loss to key drivers, difficulty of parameter derivation and limited capacity of larger scale simulation. To address the limitations of process-based models, improve the prediction and simulation of N losses, increase NUE and environmental quality, comprehensive databases of N loss from different pathways were compiled by data mining, advanced machine learning models were developed to reflect the linkage between N loss pathways and soil, environmental and climatic conditions on global scale. The research reported in this thesis quantified the nitrification rate (R_{nit}) and the fraction of nitrous oxide (N_2O) from nitrification ($f_{N_2O_Nit}$), denitrification and associated N_2O, dinitrogen (N_2) production, the contribution of N_2O from autotrophic nitrification, heterotrophic nitrification and denitrification and nitrate (NO_3^-) leaching, investigated the key drivers of N loss from each pathway, performed global performed accurate global prediction of nitrification rate and the fraction of N_2O from nitrification, NO_3^-N leaching with fewer input variables using data mining, machine learning models and ^{15}N tracing experiment. Findings include:

Data mining and machine learning were integrated to predict R_{nit} and $f_{N_2O_Nit}$. According to the compiled global database on R_{nit} and $f_{N_2O_Nit}$, the average potential R_{nit} in the topsoil was 1.4 kg N ha^{-1} d^{-1}, and $f_{N_2O_Nit}$ was from 0.004 to 9.19% (average 0.46%). The machine-learning based stochastic gradient boosting (SGB) model outperformed three widely used process-based models in estimating R_{nit} and N_2O emission from nitrification by using the same input variables. SGB technique was then applied for global prediction of R_{nit} and $f_{N_2O_Nit}$ with only a few input variables (R^2 = 0.76 and 0.55, respectively). The potential R_{nit} was driven by long-term mean annual temperature, soil C/N ratio and soil pH, whereas $f_{N_2O_Nit}$ by mean annual precipitation, soil clay content, soil pH, soil total N. The global $f_{N_2O_Nit}$ varied by over 200 times (0.006-1.2%), it should be adjusted according to edaphic and environmental conditions when used in process-based models or global climate models in projecting N_2O emissions.

A global assessment of soil denitrification rate, $N_2O/(N_2O+N_2)$, and their driving factors and mitigation strategies was conducted based on 225 studies (3367 observations). N loss through denitrification varied greatly across land uses and climatic regions with an average of 0.25 kg N $ha^{-1}d^{-1}$. The average emission factor of denitrification (EF_D) was 4.8%. The wide range of $N_2O/(N_2O+N_2)$ (mean: 0.33) demonstrated that the adoption of a fixed ratio in some process-based models for estimating N_2 emissions from denitrification is not suitable. N_2 loss accounted for 67% of total denitrification. N loss as N_2, although harmless to the environment, deserves more attention from the perspective of improving NUE. Soil denitrification rate was significantly related to soil WFPS, NO_3^- content and soil temperature and soil oxygen (O_2) content. N_2 emissions were significantly correlated with latitude, WFPS, soil mineral N and soil oxygen content. Soil oxygen content, NO_3^- content, organic C, C/N ratio and WFPS were the key driver of $N_2O/(N_2O+N_2)$ ratio. The meta-analysis showed that optimizing N application rates,

using ammonium-based fertilizers compared to nitrate-based fertilizers, biochar amendment and application of nitrification inhibitors could effectively reduce soil denitrification rate and associated N_2 emissions by 34-219% and 15-226%, respectively. These findings highlight that N loss via soil denitrification and N_2 emissions cannot be neglected, and that mitigation strategies should be adopted to reduce N loss and improve N use efficiency. Our study provides a solid foundation to large-scale estimations of denitrification and the refinement of relevant parameters used in the submodels of denitrification in process-based models.

The contribution of N_2O production pathways and its driving factors in forest soils were investigated by global data analysis and an incubation experiment with ^{15}N tracing technique in both Australia and worldwide. Based on 13 forest soils sampled within Australia, forest soils in temperate areas had the highest N_2O emission rate (19.5 μg N kg^{-1} soil d^{-1}), followed by subtropical and arid soils (3.84 and 3.80 μg N kg^{-1} soil d^{-1}, respectively). Heterotrophic nitrification dominated N_2O production in Australian forest soils; its contribution followed the order of arid (78%) > subtropical (69%) > temperate (59%). N_2O from heterotrophic nitrification was negatively related to MAT and the contribution of heterotrophic nitrification to N_2O production was negatively related to soil TN and TC. These results partially agreed with the global literature data synthesis, which showed that in addition to heterotrophic nitrification (42%), denitrification (43.5%) was also a key pathway of N_2O production in global forest soils. Globally, soil pH, moisture content, total N content, total C content and MAT contributed to heterotrophic nitrification and denitrification to N_2O production.

A machine learning model (NL_{NO3} model) was developed based on a global literature-based database of NO_3-N leaching from field experiments (1818 observations) to predict NO_3-N leaching from agroecosystems. The NL_{NO3} model can reliably predict NO_3-N

leaching using a few easily accessible input variables (R^2=0.75). According to the model estimation of NO_3-N leaching, the global spatiotemporal pattern and hotspots were identified. The total NO_3-N leaching in agroecosystems increased from 23.2 Tg N yr^{-1} in 1961 to 32.8 Tg N yr^{-1} in 2000 and 39.7 Tg N yr^{-1} in 2010. Hotspots of NO_3-N leaching in agroecosystems expanded from Europe in 1961 to China, South Asia and Brazil in 2000 and 2010. The high spatiotemporal heterogeneity of NO_3-N leaching was mainly driven by soil properties (soil TN, soil pH, soil texture), aridity index and farming practices (N fertilization and irrigation).

Results of the present research demonstrate that the capacities of data mining in better understanding the complex N cycling in terrestrial ecosystems and informing potential mitigation strategies to reduce N loss. Data mining coupled with advanced machine learning methods could not only address the limitations of process-based models and improve model simulation performances, but also provide an alternative approach in predicting N dynamics at a larger scale.

Keywords: nitrogen loss, data mining, nitrification, denitrification, nitrate leaching, N_2O production pathways, machine learning, process-based model.

DECLARATION

This is to certify that:

(i) the thesis comprises only my original work towards the Ph.D. except where indicated in the Preface,

(ii) due acknowledgement has been made in the text to all other material used, and

(iii) the thesis is fewer than 100,000 words in length, exclusive of tables, figures, bibliographies and appendices.

Baobao Pan

October 2021

PREFACE AND CONTRIBUTION OF AUTHORS

This thesis is comprised of eight chapters. The first chapter is the introduction, followed by the literature review (Chapter 2) which outlines the available literature on the related research questions and identifies current knowledge gaps. A general methodology chapter (Chapter 3) provides detailed information on the materials and methods used in the studies included in this thesis. Four results Chapters 4 to 7 were written in manuscript format according to the specific format of individual journals. Finally, the general discussion and conclusions (Chapter 8) highlight the key findings of this thesis and suggest areas for further research.

The candidate is the primary author of all manuscripts; co-authors include Shu Kee Lam, Deli Chen, Enli Wang, Longlong Xia, Yushu Zhang, Arvin Mosier and Xia Liang. The candidate is responsible for designing and conducting the research, collecting and analyzing the data, and writing the manuscripts. The co-authors assisted in improving the methodology, data quality and polishing the manuscripts.

The manuscripts included in this thesis are presented in the following order:

Chapter 4

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Chapter 6

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TABLE OF CONTENTS

ABSTRACT i
DECLARATION v
PREFACE AND CONTRIBUTION OF AUTHORS vi
PUBLICATIONS viii
ACKNOWLEDGEMENTS ix
TABLE OF CONTENTS xi
LIST OF TABLES xiv
LIST OF FIGURES xvii
LIST OF ABBREVIATIONS xx
CHAPTER 1 1
1INTRODUCTION 1
1.1 Research background 1
1.2 Conceptual framework 5
1.3 Research objectives and organization of thesis 7
1.4 Significance of the study 10
CHAPTER 2 14
LITERATURE REVIEW 14
2.1 N loss pathways and their driving factors 14
2.2 Different scales of models to predict N cycle in agroecosystems 20
2.3 Limitations of existing process-based models 21
2.4 The need of data mining and machine learning 25
2.5 Summary of current knowledge gaps 29
CHAPTER 3 43
GENERAL METHODOLOGY 43
3.1 Data mining and database compilation 43
3. 2 Machine learning models (Chapters 4 and 6) 45
3.3 Linear mixed-effect models (Chapter 5) 48
3.4 Meta-analysis (Chapter 5) 49
3.5 Incubation study (Chapter 6) 51
CHAPTER 4 60
Nitrification and its N_2O emission in terrestrial ecosystems: a global prediction based on data mining and a machine leaning model 60

4.1 Introduction....61
4.2 Methods....62
4.3 Results....64
4.4 Discussion....65
4.5 Conclusion....69
4.6 Supplementary information....71
Chapter 5....83
Nitrogen loss via denitrification in terrestrial ecosystems: a global synthesis based on data mining....83
5.1 Introduction....84
5.2 Data and Methods....85
5.3 Results....86
5.4 Discussion....89
5.5 Conclusion....91
5.6 Supplementary information....95
CHAPTER 6....146
N_2O production pathways in forest soils: coupling Australian case study and global literature data....146
6.1 Introduction....148
6.2 Materials and methods....150
6.3 Results....154
6.4 Discussion....159
6.5 Conclusion....162
CHAPTER 7....168
Spatiotemporal trend of nitrate leaching in global agroecosystems: a machine learning model evaluation....168
7.1 Introduction....169
7.2 Materials and methods....172
7.3 Results....176
7.4 Discussion....180
7.5 Conclusion....185
7.6 Supplementary information....186
CHAPTER 8....226
GENERAL DISCUSSION AND CONCLUSIONS....226

8.1 Major findings 226
8.2 Implications of the study 229
8.3 Limitations of the study 231
8.4 Suggestions for further study 232

LIST OF TABLES

Chapter 2

Table 1. Comparison of three widely used models for N loss pathway algorithms 22
Table 2. Comparison of widely used supervised machine learning models 28

Chapter 3

Table 1. Keywords and selection criteria used in the global literature search.....44

Chapter 4

Table 1. Summary statistics of nitrification, the fraction of associated N_2O emissions and environmental variables from global literature 65
Table 2. Comparison of observed and predicted nitrification and associated N_2O emissions by SGB1, APSIM, Crop-DNDC and WNMM models.....66
Table 3. Model prediction of *R*nit by stepwise selection of variables.....67
Table 4. Model prediction of *f*N2ONit by stepwise selection of variables.....67

Chapter 5

Supporting Information

Table S1. Studies used in the meta-analysis to examine the effects of mitigation strategies on soil denitrification rate.....123
Table S2. Studies used in the meta-analysis to examine the effects of mitigation strategies on soil N_2 production.....139

Chapter 6

Table 1. Soil properties of sampling sites in Australia 154
Table 2. Soil properties from literature-based data.....157
Table 3. Spearman's correlation analysis of the contribution of N_2O production with climatic variables and soil properties globally. * $p < 0.05$ ** $p < 0.01$.....158

Chapter 7

Table 1. Summary statistics of NO_3-N leaching database from collected literatures...176
Table 2. The N fertilization and simulated NO_3-N leaching worldwide in 1961, 2000 and 2010 178

Supporting Information

Table S1. Model prediction of NL_{NO3} by stepwise selection of variables 193

Table S2. The global N application and simulated NO_3^--N leaching loss in cropland and grassland in 1961, 2000 and 2010 194

LIST OF FIGURES

Chapter 1

Figure 1. Soil N transformation and loss pathways 1

Figure 2. Using data mining to improve the prediction of key N loss from terrestrial ecosystems 4

Figure 3. Conceptual framework of this study 5

Chapter 4

Figure 1. The effects of soil pH, soil temperature and WFPS on nitrification in process-based models (ASPIM, WNMM and CropDNDC).....63

Figure 2. Global distribution of *R*nit predicted by SGB (A2) model 6

Figure 3. Global distribution of *f*N2ONit predicted by SGB (B3) model 68

Supporting Information

Figure S1. Mean square errors (MSE) reduction at different levels of the parameters against number of trees. a) Learn rate, b) subsample fraction and c) Minimum terminal node 75

Figure S2. Mean square errors (MSE) reduction of training and validation datasets for SGB1. a) nitrification rate, b) N_2O emission from nitrification 76

Figure S3. Distribution of nitrification, the fraction of associated N_2O emissions from global literature.....77

Figure S4. Scatter plot of the observed against predicted nitrification rate by SGB1, APSIM, crop-DNDC and WNMM models 77

Figure S5. Scatter plot of the observed against predicted nitrification associated N_2O emission by SGB1, APSIM, crop-DNDC and WNMM models 78

Figure S6. Global soil and climate basic information used in predicting R_{nit}. a) Soil pH, b) Mean annual temperature, c) Soil C/N ratio 79

Figure S7. Global soil and climate basic information used in predicting $f_{\mathrm{N_2O_Nit}}$. a) Soil pH, b) Mean annual precipitation, c) Soil clay content, d) Soil total nitrogen 80

Figure S8. Global map of land cover–type climatology. Adapted from "A global land cover climatology using MODIS data", Broxton *et al* 2014 © American Meteorological Society. Used with permission 80

Chapter 5

Figure 1. Location of study sites included in the database 86
Figure 2. Denitrification rate across land uses and climatic zones from field experiments. Values in parentheses represent the number of observations. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$ 87
Figure 3. EF_D across land uses and climatic zones from field experiments. Values in parentheses represent the number of observations. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$.....87
Figure 4. The $N_2O/(N_2O+N_2)$ ratio of denitrification across climatic zones and land use types. Values in parentheses represent the number of observations. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$ 88
Figure 5. Relationships between soil denitrification rate and a) soil WFPS, b) soil O_2 concentration, c) soil NO_3 content, d) soil mineral N, e) soil pH and f) soil temperature. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.....88
Figure 6. Relationships between soil N_2 emissions and a) latitude (circles: the Northern Hemisphere; triangles: Southern Hemisphere.), b) soil mineral N, c) soil WFPS and d) soil O_2 concentration. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies 89
Figure 7. Relationships between soil $N_2O/(N_2O+N_2)$ ratio and a) soil pH, b) soil WFPS, c) soil O_2 concentration, d) soil C/N ratio, e) soil NO_3^- content and f) soil organic carbon. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.....90
Figure 8. Effect of N application, fertilizer type, amendment, management practice and nitrification inhibitor on a) denitrification rate and b) N_2 production. Means and 95% confidence intervals are depicted. Numbers of experimental observations are in

parentheses. DCD: dicyandiamide, DMPP: 3,4-dimethylpyrazole phosphate, Nitrapyrin: 2-chloro-6-(trichloromethyl) pyridine 92
Figure 9. The effects of management practices on soil denitrification rate, N_2 emission and associated soil N transformations. Values presented were obtained from the meta-analysis in this study. '+' and '–'denote increased and decreased responses to management practices. DNRA: dissimilatory nitrate reduction to ammonium.....92

Supporting Information

Figure S1. Relationships between soil denitrification rate and a) latitude (circles: the Northern Hemisphere; triangles: Southern Hemisphere.), b) SOC, c) soil C/N ratio and d) N application rate. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies 97
Figure S2. Relationships between soil N_2 production and a) soil NO_3^- content, b) soil pH, c) soil C/N ratio, d) soil organic carbon e) N application rate and f) soil temperature. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.....98
Figure S3. Relationships between soil N_2O /(N_2O+N_2) ratio and a) soil mineral N, b) latitude (circles: the Northern Hemisphere; triangles: Southern Hemisphere) and c) N application rate and d) soil temperature. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.....99
Figure S4. Relationships between soil denitrification rate and soil pH in different land use types. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies 100
Figure S5. Measurement of denitrification rate between AIT and ^{15}N-gas flux method. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$ 101

Chapter 6

Figure 1. The location of soils collected for the incubation study.....151
Figure 2. N_2O production rates a) and pathways b) in Australian forest soils from different climatic zones. N_2O_h, N_2O_a and N_2O_d are N_2O emission rates via heterotrophic nitrification, autotrophic nitrification and denitrification, respectively. n_h, n_a and n_d are the contribution of heterotrophic nitrification, autotrophic nitrification and denitrification to N_2O production, respectively. Different letters in Figure 2a on the top indicate that the total values are significantly different at $p< 0.05$.....155
Figure 3. Correlation analysis of N_2O production rate and contribution with climatic variables and soil properties. * $p < 0.05$ ** $p < 0.01$.....156
Figure 4. The N_2O production pathways from forest soils from global studies (n=39), n_h, n_a and n_d are the contribution of heterotrophic nitrification, autotrophic nitrification and denitrification to N_2O production, respectively.....158

Chapter 7

Figure 1. Study site distribution in the database.....173
Figure 2. a) Scatter plot of the observed against predicted NO_3-N leaching by NL_{NO3} model (n=1818, R^2=0.75, MSE=0.4, MAPE = 0.27); b) relative importance of variables in NL_{NO3} model.....177
Figure 3. Global distribution of NO_3-N leaching in agroecosystems in year 1961, 2000 and 2010 179
Figure 4. Relationships between NO_3-N leaching and (a) N_IN, (b) AI, (c) IR, (d) LONG, (e) S_PH and (f) S_TN. Shaded bands in figures indicate 95% confidence intervals, and the dashed lines represent the average NO_3-N leaching.....180

Supporting Information

Figure S1. Distribution of a) soil pH and b) TN in agroecosystems 194
Figure S2. Global distribution of arid index (AI).....195
Figure S3. Global distribution of N application rate (N_IN) 196
Figure S4. Global distribution of irrigation application rate (IR) 197
Figure S5. Average NO_3^--N leaching rate in different soil texture. Different letters in the figure indicate that the values are significantly different at p< 0.05.....198

LIST OF ABBREVIATIONS

4R	Right nutrient source, Right rate, Right time, Right place,
AI	Aridity Index
ANN	Artificial Neural Networks
APSIM	Agricultural Production Systems Simulator Model
BRT	Boosted Regression Tree
C	Carbon
CO_2	Carbon Dioxide
CV	Cross Validation
DAYCENT	Daily time-step version of the CENTURY biogeochemical model
DNDC	DeNitrification DeComposition
DNRA	Dissimilatory nitrate (NO_3^-) reduction to ammonium (NH_4^+)
EF	Emission Factor
FASSET	Farm Assessment Tool
$f_{N_2O_Nit}$	the fraction of N_2O from nitrification
GHG	Green House Gas
GLEAMS	Groundwater Loading Effects of Agricultural Management Systems
IPCC	International Panel on Climate Change
IR	Irrigation Rate
IRMS	Isotope Ratio Mass Spectrometer
ISRIC	International Soil Reference and Information Centre
LATI	Latitude
LEACHM	the Leaching Estimation and Chemistry Model
LLM	Linear Mixed Models
LONG	Longitude
LR	Learn Rate
LUS	Land Use System
MAP	Mean Annual Precipitation
MAPE	Mean Absolute Percentage Error
MAT	Mean Annual Temperature
ML	Machine Learning
MINCHILD	Minimum terminal node
MSE	Mean Square Error
N	Nitrogen
N_IN	N application rate
N_2	Dinitrogen
N_2O	Nitrous Oxide
NH_3	Ammonia

NH_4^+	Ammonium
NO	Nitric Oxide
NO_3^-	Nitrate
NOE model	Nitrous Oxide Emission Model
Nr	reactive Nitrogen
NSE	Nash–Sutcliffe model efficiency
NT	Number of trees
NUE	Nitrogen Use Efficiency
O2	Oxygen
PET	Potential Evapotranspiration Rate
R^2	Regression coefficients of determination
RF	Random Forest
RMSE	Root Mean Square Error
Rnit	Nitrification rate
RZWQM	Root Zone Water Quality Model
S_TEX	Soil Texture
SF	Subsample Fraction
SGB	Stochastic Gradient Boosting
SVR	Support Vector Regression
TC	Total Carbon
TN	Total Nitrogen
WFPS	Water Filled Pore Space
WISE	World Inventory of Soil Emission
WNMM	Water and Nitrogen Management Model

CHAPTER 1

1INTRODUCTION

1.1 Research background

Nitrogen (N) is a key element of species composition, diversity, dynamics and functioning of different terrestrial, freshwater and marine ecosystems (Vitousek *et al.*, 1997; Erisman *et al.*, 2013). More than half of the world's population is nourished by food produced using synthetic nitrogen fertilisers (Erisman *et al.*, 2008). However, due to the increased accessibility to N fertilizer, N cycling has been greatly affected locally, regionally, nationally and globally. A large fraction of the N applied (up to 86% in vegetable production systems, Ju *et al.* (2011)) is lost from the plant-soil system, via pathways such as ammonia (NH_3) volatilisation, nitrate (NO_3^-) leaching, denitrification and nitrous oxide (N_2O) emission (Figure 1).

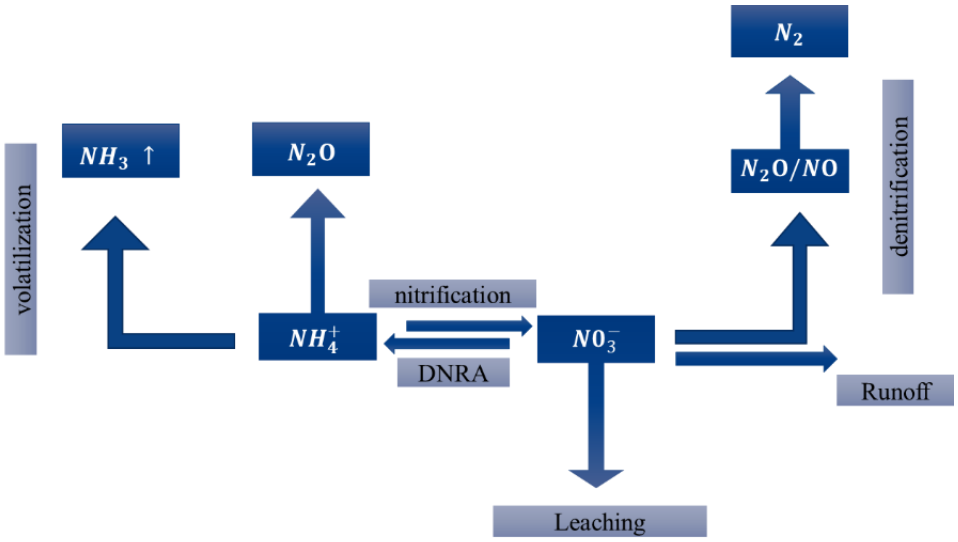


Figure 1. Soil N transformation and loss pathways, DNRA is dissimilatory nitrate (NO_3^-) reduction to ammonium (NH_4^+)

N losses may not only lead to a significant economic loss for farmers, but also adversely affect the environment and human health (Galloway *et al.*, 2008). Therefore, understanding N cycling in terrestrial ecosystems is essential to improve N management and sustain the environmental quality.

In the past 50 years, many studies have been conducted to investigate N loss from different pathways under laboratory and field conditions. However, findings from individual site-specific experiments cannot be easily extrapolated to a larger scale. However, individual experiments have provided tremendous amounts of data for developing advanced technical approaches to guide N management with a better understanding of the complex, multivariate and unpredictable N cycling. To date, different types of models have been developed such as simple empirical models, process-based models and scaling models (Chen *et al.*, 2008). Process-based models, such as such as APSIM (The Agricultural Production Systems sIMulator, Keating *et al.*, 2003; Holzworth *et al.*, 2014), DNDC (Denitrification-Decomposition (DNDC) model, Li *et al.*, 1992; Li *et al.*, 2000; Zhang *et al.*, 2002), WNMM (Water and Nitrogen Management Model, Li *et al.*, 2007), DAYCENT (Daily Centruy Model, Parton *et al.*, 1998; Del Grosso *et al.*, 2000), etc., are developed to simulate the complex interactions of water, carbon and N cycles of terrestrial ecosystems (Giltrap *et al.*, 2015b). However, such models have limitations that sometimes result in a mismatch between the observed and simulated data for certain N loss pathways. Firstly, most process-based models consist of different configurations. Based on all the configurations, the required data can be categorized as soil and crop modules, climatic reference data and management characteristics. Data unavailability in particular modules involved in the model often affects the initialisation and operation of the model. Secondly, for most process-based models, not all biophysical processes involved have been validated due to the limited

availability of experimental data. One well-known limitation is the use of a constant parameter in these models to estimate the N_2O emission from nitrification. However, it is not ideal to use a fixed parameter given that the most appropriate parameter value can vary with soils, climates and geographical locations. Thirdly, different responses of N losses to edaphic and environmental factors among models lead to inconsistent simulation results, and sometimes these results are beyond sensible ranges or without biological meaning. Lastly, process-based models have the potential capacity to provide global simulations; however, the large uncertainty of simulation would limit the performance because process-based models are normally calibrated for localized areas. In addition, the algorithms in process-based models were developed from limited empirical data, and the original boundary condition could limit the model applied in a certain range.

Therefore, to improve simulation and prediction of N losses, the limitations of existing process-based models as discussed above should be addressed. Furthermore, the world is moving into a big data era, with a large amount of data becoming available from around the world and across different edaphic, environmental and climatic conditions. Therefore, this study aims to apply novel data mining and machine learning techniques to quantify N losses, address process-based model limitations and perform global prediction with fewer variables (Figure 2).

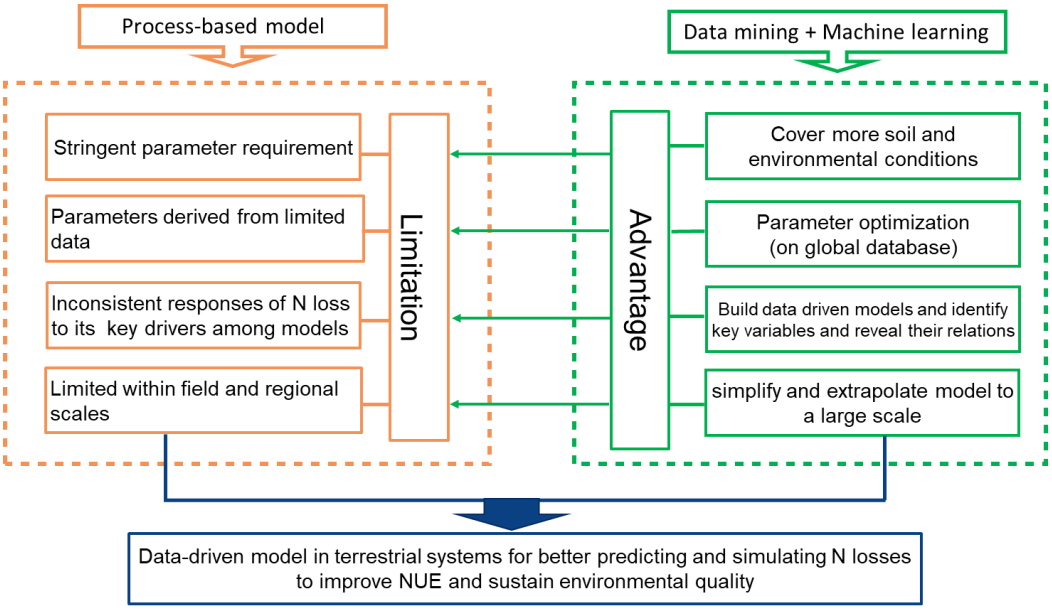


Figure 2 Using data mining to improve the prediction of key N losses from terrestrial ecosystems

First, the parameter availability and limited empirical data-based algorithms always constrain the accuracy of simulation performance in process-based models. The literature-based databases, covering a wide range of data under different soil, climate and management conditions across the world, can supplement such limitation in the process-based model. Moreover, data-driven models could identify the key variables and reveal their relationships to cover the shortage of inconsistent responses of N loss to its key drivers in process-based models. Data mining coupled with machine learning models could remove the redundant variables through variable selection techniques. In this case, the improved models can be much easier to extrapolate to a large scale. All the advantages of data mining and machine learning models can address the limitations of process-based models, achieve a better prediction and simulation of N losses, and improve nitrogen use efficiency (NUE) and environmental quality.

1.2 Conceptual framework

This thesis aimed to compile different databases of key N loss pathways (nitrification, denitrification and nitrate leaching) in terrestrial ecosystems. Based on these databases, different approaches were adopted to address the limitations of process-based models in simulating N loss from different pathways, to quantify N loss for each pathway, to investigate the key drivers of N loss, to improve the global prediction of N loss from different pathways by machine learning models and to propose potential mitigation strategies to reduce N loss. Figure 3 presents the conceptual framework of the study.

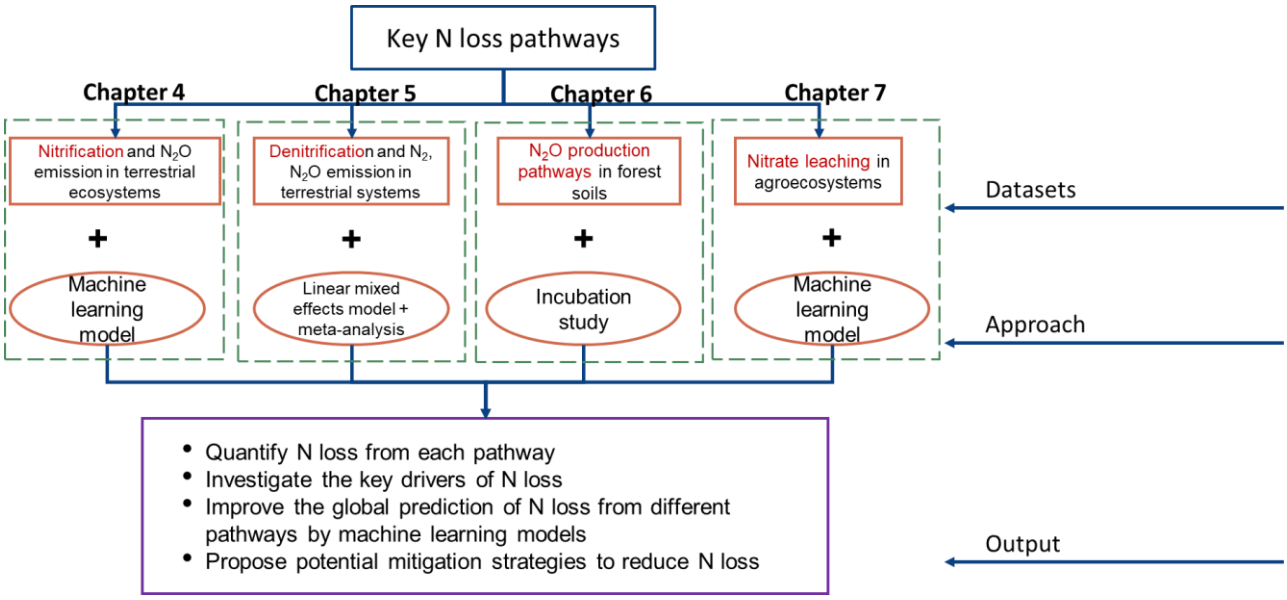


Figure 3 Conceptual framework of this study

Specifically, in Chapter 4, due to the difficulties of field measurement, process-based models have embedded modules to predict nitrification rate (R_{nit}) and N_2O from nitrification. However, these models adopted the equations generated from limited empirical data, and nitrification is calculated as a function of soil water content, soil pH, soil temperature and soil ammonium content. The use of a fixed fraction is known to be problematic in predicting N_2O emission because the proportion of N_2O emission from nitrification varies with soil and environmental conditions. In addition, process-based

models may not perform well when extrapolating from site-specific to a larger scale, mainly owing to their deficiency in capturing the key processes in response to driving factors, detailed parameterization, limited data availability and their limited capacity in handling complex interacting factors (Saha *et al.*, 2007; van der Laan *et al.*, 2014; Giltrap *et al.*, 2015a). Therefore, machine-learning based stochastic gradient boosting models (SGB) coupled with a literature-based database (186 observations) were adopted to address these issues in process-based models and the proposed SGB models could perform prediction of R_{nit} and associated N_2O emissions at a large scale.

Chapter 5 aimed to conduct a global synthesis to quantify soil denitrification rate and $N_2O/(N_2O+N_2)$, to investigate their driving factors and to evaluate potential mitigation strategies of denitrification and N_2 emission. Many empirical and process-based models estimated N_2O and N_2 emissions from denitrification based on a fixed N_2O to N_2 ratio or combined with functions of soil properties due to the measurement difficulties, resulting in high uncertainties of simulations. Based on the comprehensive database (3367 observations from 225 studies), the emission factor of denitrification was calculated, the linear mixed-effect models were applied to explore the relationships between soil denitrification rate, soil N_2 emissions and the $N_2O/(N_2O+N_2)$ ratio from denitrification with soil and environmental properties, and a meta-analysis was conducted to examine the effects of mitigation strategies on denitrification and N_2 emissions using this database.

Chapter 6 aimed to evaluate the contribution of N_2O production pathways and their driving factors in forest soils. The contribution of N_2O production pathways in forest soil varied with high uncertainties under different soil and environmental conditions at the global scale and only scattered studies were conducted in Australia. Therefore, a laboratory ^{15}N tracing experiment combined with a global literature search for N_2O production pathways in forest soils was carried out to separate and quantify the relative

contribution of N_2O from denitrification, autotrophic nitrification and heterotrophic nitrification pathways and to investigate the key drivers of N_2O production pathways.

In Chapter 7, a machine learning model based on a literature-based database was developed to address limitations of process-based models in simulating NO_3-N leaching in agroecosystems, e.g., detailed initialization, complex soil parameters and hydrodynamic coefficients. A comprehensive database of NO_3-N leaching was compiled (1818 observations from 286 studies) and used to develop the machine learning model (NL_{NO3} model). The NL_{NO3} model was applied to quantify global NO_3-N leaching in agroecosystems from a bottom-up perspective and to investigate the global spatiotemporal pattern of NO_3-N leaching.

1.3 Research objectives and organization of thesis

Many field and laboratory experiments have been carried out to quantify the N loss from different pathways and to investigate the key drivers and potential mitigation strategies in reducing N loss in terrestrial ecosystems. Process-based models have been adopted to simulating such complicated N processes in different scales. However, limitations of current process-based models were mainly associated with data availability, complex relationships between input and output variables and difficulty of model parameter estimation. To address the limitations of process-based models, and to improve the prediction and simulation of N losses, comprehensive databases of N loss from different pathways were compiled by data mining, and advanced machine learning models were developed to reflect the linkage between N loss pathways and soil, environmental and climatic conditions at larger scales. This thesis begins with an introductory Chapter 1, which covers the research background, conceptual framework, objectives and significance of the study. Chapter 2 presents a literature review on the main N loss pathways, their related factors and different approaches of N loss predictions. Chapter 3

summarizes the general methods used in each study. The specific objectives of the four results chapters were:

(i) Chapter 4: Nitrification as a main N loss pathway was discussed. Measurements of nitrification rate and how much nitrified N is emitted as N_2O are challenging. Many ecological process-based models adopt the equations generated from limited empirical data, and N_2O emission from nitrification is calculated as a fixed ratio.

Based on a comprehensive database compiled from global literature; this study first attempted:

- to establish SGB models to predict R_{nit} and the ratio of N_2O from nitrification;
- to compare the performance of SGB and process-based models in predicting nitrification rate and associated N_2O production; and
- to extend spatial simulation of nitrification rate and the ratio of N_2O from nitrification using the better performing model.

(ii) Chapter 5: Dinitrogen (N_2) and nitrous oxide (N_2O) produced via denitrification as a major N loss pathway was discussed. N_2O emissions contribute to global warming and stratospheric ozone depletion whereas N_2 losses decrease NUE. Many empirical and process-based models were developed to estimate N_2O and N_2 emissions from denitrification based on a fixed N_2O to N_2 ratio. However, the N_2O and N_2 emissions from soil denitrification showed a large spatial and temporal heterogeneity at a global scale. A better understanding of $N_2O/(N_2O+N_2)$ ratio and its driving factors could improve the estimation of denitrification in earth system models.

Based on a comprehensive database of soil denitrification rate, N_2 emissions and $N_2O/(N_2O+N_2)$ collected from global peer-reviewed studies:

- to quantify soil denitrification rate, N_2 emissions and $N_2O/(N_2O+N_2)$ across different land use systems and climatic zones;
- to further assess the relationships between soil/environmental factors and soil denitrification rate, N_2 emissions and $N_2O/(N_2O+N_2)$ ratio; and
- to evaluate the effects of mitigation strategies on soil denitrification rate and N_2 production.

(iii) Chapter 6: In Chapters 4 and 5, N loss pathway of nitrification and denitrification were investigated. To improve the simulation performance, it is necessary to evaluate the contribution of N_2O production pathways and their driving factors, especially the main pathways: denitrification, autotrophic nitrification and heterotrophic nitrification according to the hole in the pipe model. Forest soil is the largest source of N_2O emissions from terrestrial ecosystems and the contribution of N_2O production by different pathways varies widely. Besides, only scattered studies were conducted in Australia, an incubation study was conducted in chapter 6 to provide more data. Therefore, **Chapter 6** aimed to examine the patterns and driving factors of N_2O production pathways in both Australian and global forest soils by combining data mining and incubation study.

Based on a global literature-based database and an incubation study of N_2O production pathways in forest soils:

- to evaluate the contribution of N_2O production pathways of Australian forest soils in subtropical, temperate and arid regions;
- to quantify the N_2O production pathways from forest soils globally; and
- to investigate the relationships between dominant N_2O production pathways in forest soils and driving factors.

(iv) Chapter 7: In previous chapters (Chapters 4-6), two N loss (nitrification and denitrification) pathways were analyzed based on global synthesis and an incubation

study. The following **Chapter 7** aimed to explore another N loss pathway- nitrate leaching- using a global database of field studies in agroecosystems. In this chapter, global nitrate leaching is quantified by SGB models and the spatiotemporal pattern is investigated.

Based on a comprehensive database of global NO_3-N leaching

- to develop a more accurate machine learning model of NO_3-N leaching at a global scale;
- to investigate the global spatiotemporal trend of NO_3-N leaching in 1961, 2000 and 2010; and
- to identify the hotspots of NO_3-N leaching worldwide for setting priorities of mitigation.

In previous chapters, three global databases were compiled and analyzed, and an incubation study was conducted to quantify the N loss from three key pathways (Chapters 4 - 7). Two machine learning models were proposed in simulating nitrification and nitrate leaching globally (Chapters 4 and 6). This thesis concludes with Chapter 8, which presents major findings of the research, implications of the results, limitations of the study and suggestions for future research.

1.4 Significance of the study

By providing comprehensive literature-based databases and coupling with advanced machine learning models. The results in this research will provide accurate large-scale estimations of N loss from different pathways, empower the simulation capacity of process-based model by addressing the limitations of submodules in simulating N losses, improve the understanding of N dynamics in terrestrial ecosystems, help to improve NUE and sustain the environmental quality.

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CHAPTER 2

LITERATURE REVIEW

Nitrogen (N) losses not only represent a significant economic loss for farmers, but also adversely affect the environment and human health (Galloway et al., 2008). Examples include the eutrophication in Australia's Great Barrier Reef and China's Lake Tai. In this chapter, the main N loss pathways and their driving factors are reviewed. Nitrogen losses vary with soil and climatic conditions, land management practices and soil microbial interactions. Process-based models were developed to estimate the processes and interactions of water, carbon (C) and N cycles of terrestrial ecosystems (Chen *et al.*, 2008; Singh *et al.*, 2013). Nevertheless, limitations of models sometimes result in a mismatch between the observed and simulated data for certain N loss pathways. To improve simulation and prediction of agricultural N losses, we need to handle the limitations of existing process-based models, which are mainly associated with limited data availability, complex relationships between input and output variables and difficulty of model parameter estimation. The desired models are expected to inherit the advantages of existing models while addressing their limitations. Data mining coupled with machine learning models provides a novel way to reflect the linkage between N loss pathways and soil, environmental and climatic conditions to address the limitations of process-based models.

2.1 N loss pathways and their driving factors

N is a key element of species composition, diversity, dynamics and functioning of different terrestrial, freshwater and marine ecosystems (Vitousek *et al.*, 1997). The use of N fertilisers is also essential for profitable agricultural systems. However, a large fraction

of the N applied (up to 86% in vegetable production systems, Ju *et al.* (2011)) is lost from the plant-soil system, via pathways such as ammonia (NH_3) volatilization, nitrate (NO_3^-) leaching, N_2O emission from nitrification and denitrification. It should be noted that N_2 represents a substantial N loss accounting for up to 85% of total denitrification (year 2000) (Bouwman et al., 2013) but it is often overlooked and poorly studied as N_2 is non-reactive (Almaraz et al., 2020).

2.1.1 Ammonia volatilization

In terrestrial systems, NH_3 via volatilization is one of the main N loss pathways. More than 40% (up to 65%) of the applied N can be lost via NH_3 under particular environmental and soil conditions (Singh *et al.*, 2013). NH_3 can be volatilized and then returned to the earth's surface, leading to soil and water acidification and eutrophication (Schulze *et al.*, 1989). In addition, ammonia volatilization and its deposition also contribute to indirect N_2O greenhouse gas (GHG).

Higher ammonium (NH_4^+) concentration leads to higher potential NH_3 emitted into the air (Cameron *et al.*, 2013). Whitehead and Raistrick (1990) demonstrated that more ammonia gas production under high pH conditions (Reddy *et al.*, 1979). The temperature of both soil and atmosphere plays an essential role in urea hydrolysis after fertilization and NH_3 volatilization (Black *et al.*, 1985; McGarry *et al.*, 1987). A study showed that temperature is positively related to NH_3 emission from 5 to 45°C (He *et al.*, 1999). Soil moisture influences the NH_4^+/NH_3 equilibrium in soil solution through initiating urea hydrolysis and subsequent NH_3 volatilization (Sommer *et al.*, 2004). Al-Kanani *et al.* (1991) reported that when the soil moisture content was at field capacity, the volatilization rate has a positive correlation with water content and the maximum volatilization occurred. Pan *et al.* (2016) conducted a meta-analysis and showed that 4R nutrient stewardship, farming practices (e.g., irrigation, residue retention, amendment application),

fertilizer types could affect the rate of NH_3 volatilization by altering soil properties and microsite conditions.

2.1.2 Nitrate leaching

Nitrate leaching from the rootzone to groundwater is considered as a major N loss pathway that could reduce N use efficiency in agroecosystems (Di and Cameron, 2002). When water drains below the root-zone, nitrate leaching occurs (Burkitt, 2014). When dissolved organic N and NH_4^+ are lost to the surface and subsurface water flow, NO_3^- is the dominant form of dissolved N in the flowing water (Robertson and Vitousek, 2009). Nitrate leached to the groundwater would cause groundwater contamination, eutrophication (Robertson and Vitousek, 2009). Wang *et al.* (2017) estimated that N leaching in cropland accounted for 16% of N input using natural N isotope constraints combined with a global mass-balance model. Lin *et al.* (2001) used a global process-based simulation model to estimate that 19% of N fertilizer input was lost through N leaching in agricultural soils.

A previous study demonstrated less leaching loss in fine-textured soils than poorly structured sandy soils owing to slower water movement and potentially higher denitrification rate in fine-textured soils (Di and Cameron, 2002). Soil organic matter and mineralization process can both affect nitrate leaching. Nitrate leaching can be increased significantly on the bare soil (Cameron *et al.*, 2013) and most of the leached nitrate comes from organic N mineralization and fertilizer N remineralization. Soil depth is also considered an essential factor of nitrate leaching. Nitrate can reach to the groundwater faster in shallow soils than that in deep soils, resulting in higher nitrate leaching (Di and Cameron, 2002).

Climate and seasonal changes highly related to the amount of drainage are important factors for nitrate leaching. It has been studied that nitrate leaching increased due to the high soil drainage, for example, late-autumn, winter and early-spring with low evapotranspiration, high rainfall and low plant N uptake (cool condition) would lead to high soil drainage and subsequent high nitrate leaching (Cameron *et al.*, 2013; Burkitt, 2014). If soils are at the field capacity, excessive rainfall will cause nitrate leaching and surface runoff (Cameron *et al.*, 2002). In dry summer, nitrate might accumulate in the surface soil, leached in the subsequent winter. Furthermore, rainfall or irrigation applied after dry summer can cause more nitrate leaching (Goulding, 2000). Nitrate leaching in different land use varied widely. For example, extensively grazed grassland has relatively low nitrate leaching potential (Di and Cameron, 2002; Moir *et al.*, 2006; Cameron *et al.*, 2013). However, due to the high N input, short period of plant growth, high N concentration, shallow roots and low N use efficiency, intensive vegetable production systems have a high risk of nitrate leaching loss (Cameron *et al.*, 2013).

2.1.3 N_2O emissions from nitrification and denitrification

Nitrous oxide (N_2O) is a potent greenhouse gas with a global warming potential 298 times that of carbon dioxide (CO_2) over a 100-year timeframe (WMO, 2013), contributing to global warming and stratospheric ozone depletion (Ravishankara *et al.*, 2009). Soil systems, as dominant sources, account for approximately 60% of global N_2O emissions (Grace *et al.*, 2016). N_2O is produced mainly from microbial processes of nitrification and denitrification. Although the pathways of N loss as N_2O are well understood, the relative contribution of each pathway is not well known and its controlling factors including physical, chemical and biological parameters and environmental conditions should be further investigated (Chen *et al.*, 2008).

Nitrification is a microbial process, in which ammonium (NH_4^+) is converted into nitrate, a major pathway of N_2O production particularly in aerobic soils. The relationships between soil properties and nitrification rate have been well-documented (Maag and Vinther, 1996; Bengtsson *et al.*, 2003; Zebarth *et al.*, 2015). Li *et al.* (2020) conducted a global-scale study and demonstrated that soil N content, C/N ratio, soil pH and mean annual temperature (MAT) are the key drivers of nitrification rate. High soil C/N ratio may stimulate N immobilization, decrease availability of NH_4^+ for nitrification, and subsequently lower nitrification rate (Bengtsson *et al.*, 2003). Soil nitrification rate is regulated by microbial community structure and abundance which are affected by MAT in the long term (Avrahami *et al.*, 2003; Hu *et al.*, 2016). In addition, the environmental conditions on site can be reflected by MAT. The optimum temperature for nitrification varies under different environmental conditions but mostly within 20-35°C (Zaman and Chang, 2004; Ouyang *et al.*, 2017). Soil pH significantly affects the rate of nitrification and denitrification and the abundance and composition of functional genes of the N cycle (Shen *et al.*, 2008; Hu *et al.*, 2013). Generally, the maximum nitrification rate can be observed at pH 7-9 (Haynes, 2012). Soil moisture content is the main factor of N_2O emission because it can regulate the oxygen availability for soil nitrifiers (Butterbach-Bahl *et al.*, 2013). Maag and Vinther (1996) and Liu *et al.* (2017) demonstrated that N_2O from nitrification increased with increasing soil moisture and the optimum soil moisture for N_2O from nitrification was within the range of 30% to 70%.

Denitrification is a microbial process by which NO_3^- is gradually reduced to N_2O and dinitrogen (N_2) (Zaman *et al.*, 2012). It is a major pathway of nitrogen (N) loss in terrestrial ecosystems (Bouwman *et al.*, 2013). The N loss through soil denitrification from terrestrial ecosystems was estimated to be 105-185 Tg N yr^{-1} (Tiedje, 1988) and 166 Tg N yr^{-1} (Scheer *et al.*, 2020), with around 66-87 Tg N yr^{-1} in global croplands (Hofstra

and Bouwman, 2005; Seitzinger *et al.*, 2006) and 5.6 Tg N yr^{-1}in temperate grassland (Saggar *et al.*, 2013b). N_2 represents a substantial N loss accounting for up to 85% of total denitrification (year 2000) (Bouwman *et al.*, 2013) but it being non-reactive is often overlooked and poorly studied (Almaraz *et al.*, 2020). The quantification of N_2 emissions from terrestrial ecosystems remains challenging because of the high background atmospheric N_2 concentration (Zaman *et al.*, 2012; Saggar *et al.*, 2013a).

Soil denitrification and the emissions of its end-products (N_2O and N_2) are regulated by soil and environmental factors. For example, the availability of soil NO_3^- as the substrate affects denitrification rate (Abbasi and Adams, 1999). Higher carbon availability could increase denitrification rate but decrease N_2O/N_2 ratio (Clough *et al.*, 1998; Gillam *et al.*, 2008). The effect of soil pH on denitrification is controversial. It is generally accepted that acidic soil has lower soil denitrification rate (Sun *et al.*, 2012) and higher $N_2O/(N_2O+N_2)$ ratio (Zaman *et al.*, 2007; Zaman and Nguyen, 2010). Soil pH can also affect denitrification indirectly because nitrification is inhibited in acidic soils and subsequently less NO_3^- is available for denitrification (Tiedje, 1988). On the contrary, Simek and Cooper (2002) found no significant correlation between soil pH and denitrifying enzyme activity. Soil oxygen concentration, water content and rainfall are other important factors as anaerobic conditions favour denitrification (De Klein and Van Logtestijn, 1996; Ellis *et al.*, 1998; Hefting *et al.*, 2003; Saggar *et al.*, 2004). Generally, soil temperature is positively related to soil denitrification rate and associated N_2O emission (Knowles, 1982; De Klein and Van Logtestijn, 1996; Saggar *et al.*, 2013b). However, some studies demonstrated that N_2O emission may decrease with increasing temperature, because elevated temperature may enhance anaerobiosis and decrease the $N_2O:N_2$ ratio (Lai and Denton, 2018). These inconsistencies highlight the need of a

comprehensive assessment to identify the important driving factors of soil denitrification and associated gaseous loss.

2.2 Different scales of models to predict N cycle in agroecosystems

To date, numerous biogeochemical models have been developed based on empirical assumptions to simulate and estimate some unclear processes of soil-crop cycling. With the development of modelling, N dynamics under different climatic and management scenarios can be predicted. Models can be divided by scales as below:

a) Laboratory scales, which are dependent on the individual measurements, such as denitrification simulation in homogeneous systems (Leffelaar and Wessel, 1988), a mathematical model for denitrification in aggregated soils (Arah and Smith, 1989) and a zero-order kinetic model of N_2O and gaseous N (Focht, 1974).

b) Field and regional scales, which are scaled up from site-specific experiments to the landscape by using regression and empirical methods. Sozanska *et al.* (2002) established a regression model to predict N_2O emission from British soil; Roelandt *et al.* (2005) conducted two empirical models to predicting N_2O emissions from cropland and grassland in temperature climates. A simple regression model was used to estimate NH_3 emission after applying liquid cattle manure on grassland (Menzi *et al.*, 1998). However, due to the complex circumstances of agroecosystems, there is a large inconsistency in predicting N loss at the field and regional scales.

c) Nitrogen losses vary with soil and climatic conditions, land management practices and soil microbial interactions. As such, process-based models are developed to simulate the processes and interactions of water, carbon and N cycles of terrestrial ecosystems (Chen *et al.*, 2008). Because these models mainly focus on the field and regional scale simulations, it can bridge the gap between laboratory and global scale (Hu *et al.*, 2015).

Moreover, such models also allow combining individual processes as a complete system to give us a better understating of N cycling and management strategies to mitigate N losses (Vogeler *et al.*, 2013).

d) On the global scale, IPCC adopted the emission factor (EF) of 1% of added fertilizer to estimate the global inventory N_2O emission from agricultural land and a default value of 30% to represent the N fertilization induced NO_3-N leaching in the agricultural system (IPCC, 2006). This approach has the advantage of simplicity and is widely used in those countries without their country-specific emission factors (Giltrap *et al.*, 2015). However, this single value cannot represent the effects of soil, climate and management factors on N loss pathways.

2.3 Limitations of existing process-based models

Among these different scales of models, process-based model is considered as the most widely used and cost-effective tool for improving agricultural management practices, e.g. APSIM (Keating *et al.*, 2003), DNDC (Li *et al.*, 1992; Li, 2000), DAYCENT (Parton *et al.*, 1998; Del Grosso *et al.*, 2000), *ecosys* (Grant and Pattey, 1999), NLOSS (Riley and Matson, 2000), Expert-N (Engel and Priesack, 1993), WNMM (Li *et al.*, 2007), FASSET (Farm ASSEssment Tool, Olesen *et al.*, 2002). Data inputs for three commonly used process-based models for agricultural systems were listed in Table 1. These models have succeeded in simulating and predicting short-term and long-term N dynamics in various cropping systems. For example, they have been applied to simulate soil mineral N dynamics, N_2O emission and NO_3^- leaching (Li *et al.*, 2007; Chen *et al.*, 2010); crop yields and soil organic carbon sustainability (APSIM, (Luo *et al.*, 2017); greenhouse gas emissions (DNDC, (Li *et al.*, 2012). Nevertheless, such models have the following limitations that sometimes result in a mismatch between the observed and simulated data for certain N loss pathways.

Table 1. Comparison of three widely used models for N loss pathway algorithms

Models	WNMM	DNDC	APSIM
Nitrification	NH_4^+, pH, Temperature, water content	NH_4^+, pH Temperature, water content	NH_4^+, pH Temperature, water content
N_2O from Nitrification	Coefficient of nitrification rate 0.002 Temperature, water content	Coefficient of nitrification rate 0.0024	Coefficient of nitrification rate 0.002
Denitrification	NO_3^-, Temperature, water content, organic carbon	NO_3^- Microbial growth kinetics control N along NO_3^->NO_2->N_2O>N_2 pathway, water flow	NO_3^-, water content, temperature, active carbon
N_2O from denitrification	WFPS	Clay, WFPS, Temperature	N_2:N_2O ratio NO_3^-, CO_2, WFPS
Nitrate leaching	Water flow, NO_3^-, Temperature	NO_3^-, Water flow, clay content	Water flow, NO_3^-, bulk density
NH_3 volatilization	NH_4^+, wind speed, CEC, soil depth, Temperature	Temperature, Air-filled porosity, clay content, pH	N/A

2.3.1 Detailed data requirements

Stringent data requirement makes process-based model inapplicable without comprehensive measurement site-specific data. Firstly, precess-based models required site-specific data to initialise the model. A large amount of installation data including generic data and specific parameter data were needed. For example, when using process-based model to simulate future climate change scenarios, a large amount of data was required to support a historical long-term simulation, which is considered as a reference

for comparing with the output under future climate conditions. (Yang *et al.*, 2014; Li *et al.*, 2017). Secondly, most process-based models consisted of different configurations. Based on all the configurations, the required data can be categorized as soil and crop modules, climatic reference data and management characteristics (Keating *et al.*, 2003; Li *et al.*, 2007). Data unavailability in particular modules involved in the model often affects the initialisation and operation of the model, and subsequently model calibration and validation. In addition, for some particular modules, e.g., soil water balance and solute movement in APSIM model, soil physicochemical parameters in both APSIM and WNMM models, required user-defined data of the thickness of each layer and details of each layer as well. These data requirements restrict the wide application of process-based models because field measurement or monitoring of data is often impractical. Errors might arise from modelling with insufficient input data or the use of default values.

2.3.2 Parameters derived from limited data

For most process-based models, not all biophysical processes involved have been validated due to the limited availability of experimental data. Even if these processes were validated, only limited site-specific data were used and this largely hinders the derivation of parameters suitable for a wide range of agroecosystems, soils or climates (Giltrap *et al.*, 2015). One well-known limitation is the use of a constant parameter in these models to estimate the N_2O emission from nitrification (Chen *et al.*, 2008; Farquharson, 2016). Specifically, APSIM adopts a default parameter value of 0.002, without further considering any function of soil properties. WNMM considers soil temperature and soil water content for the calculation but sets a threshold of 0.002. Crop-DNDC fixes the parameter value at 0.0006 but considers the soil water-filled pore space (WFPS) and temperature in estimation. However, it is not ideal to have a constant parameter to

determine N_2O emission from nitrification, given that the most appropriate parameter value can vary with soils, climates and geographical locations.

2.3.3 Inconsistent responses of N loss to its key drivers among models

Different responses of N losses to edaphic and environmental factors among models lead to inconsistent simulation results, and occasionally these results are beyond sensible ranges or without biological meaning. For example, to simulate the nitrification process, same input parameters are used in different models and nitrification is calculated as a function of soil water content, soil pH, soil temperature and soil NH_4^+ content (Table 1). The responses of the function of nitrification to soil water content and soil temperature from APSIM, Crop-DNDC and WNMM were examined. Crop-DNDC covers a much wider range of soil water contents than APSIM and WNMM, with increasing nitrification up to 80% WFPS. In contrast, APSIM and WNMM models consider the threshold level of soil water content for nitrification to occur to be higher than DNDC. Nitrification increases with soil temperature in the WNMM and APSIM models, and plateaus over 30^{o}C but in DNDC, nitrification decreases when soil temperature exceeds 35 ^{o}C. These diverse responses reflect that the simulation results of these models may be inconsistent when the experimental data used to derive the parameters of these models were different and possibly limited.

2.3.4 Limited within field and regional scales

Process-based models are capable of global simulations; however, the large uncertainty of simulation would limit the performance because process-based models are typically calibrated for local areas (Chen *et al.*, 2008). As mentioned above, there are stringent data requirements for site-specific simulations, let alone larger-scale simulations, at regional or global scale. In addition, the algorithms in process-based models were developed from empirical data, the original boundary condition could limit the model applied in a certain

range. For example, the DNDC and APSIM can both predict soil nitrification, but if soil water content was out of the limit set in the algorithm, nitrification was assumed to be ceased in the model. However, according to measured data from global literature, nitrification did occur under 30% WFPS. Therefore, many studies were excluded from APSIM and WNMM which worsened the predictions (Maag and Vinther, 1996).

2.4 The need of data mining and machine learning

To improve simulation and prediction of N losses, the limitations of existing process-based models as discussed above should be handled, which are mainly associated with limited data availability, complex relationships between input and output variables, and difficulty of model parameter estimation. The desired techniques of data mining and machine learning are expected to address the limitations of existing models and improve the prediction performance.

Nowadays, the world has moved into a big data era, with a large amount of data becoming available across an array of edaphic, environmental and climatic conditions. Besides, unlike the time-consuming and expensive measurements of N dynamics, data mining technique is reliable at low cost that could be applied to uncover relevant insight and patterns by exploring and analysing large datasets. Data mining has four main goals including, identification (identify the patterns of data), classification (categorize data), optimization (optimize the utilization of limited resources) and prediction (determine the effect of attributes in the future) (Wu *et al.*, 2014). Machine learning (ML) provides a promising way, which allows complex relationships between input and output variables to be modelled in a data-driven way and enables important relationships never recognised previously to be revealed. As a novel way, the combination of data mining and ML models can be used to model the linkage between N loss pathways and soil, environmental and climatic conditions, addressing the limitations of process-based models.

In general, ML is defined as computational methods using available data to improve performance or to make accurate predictions (Mohri *et al.*, 2018). ML could easily identify the trends and patterns of datasets and handle multi-variety or multi-dimensional data. Besides, ML is an automatic process with continuous improvement in accuracy and efficiency by including more data (Khanzode and Sarode, 2020). Normally, ML tasks are typically classified into two categories depending on the learning type, including unsupervised and supervised learning (Liakos *et al.*, 2018). In unsupervised learning, the training data is unlabelled, and the algorithm will automatically learn from the data without supervision. The purpose of unsupervised learning is to discover hidden patterns among input variables instead of making prediction (Celebi and Aydin, 2016), i.e., there are only explanatory variables without dependent variables. Unsupervised learning, e.g., principal component analysis (PCA) (Pearson, 1901; Hotelling, 1933), K-means clustering (Hartigan and Wong, 1979) and hierarchical clustering (Zhang *et al.*, 1996), are usually applied for dimensionality reduction and clustering. For example, PCA was applied to reduce the numbers of monitoring stations for evaluating the effectiveness of river water quality monitoring network (Ouyang, 2005), and to analyze the correlation between the fluorescence spectra and paddy rice leaf N content (Yang *et al.*, 2016a); K-means clustering was used to classify synoptic meteorological conditions in a dry nitrogen deposition estimation study in South Korea (Park *et al.*, 2002).

Unlike unsupervised learning, supervised learning requests a user-defined dependent variable or target, and the given explanatory variables (Storm *et al.*, 2020). Such algorithms learn from training data and are used to predict outcomes for unforeseen data. Common advantages of supervised learning include simpler initialization processes (limited number of user-defined parameters), the capacity to predict non-linear relationships, and the ability to do regression and classification analysis (Breiman, 2001;

Friedman, 2001; Hastie *et al.*, 2009). A detailed description and comparison of widely used supervised machine learning models can be found in Table 2. Specifically, a supervised ML model, with a set of parameters governing the model formulation, is trained on an available set of pairs of input variables (soil properties, environmental conditions and climates, etc.) and output variables to derive the best model parameters. In this way, the model armed with those derived parameters can accurately predict output variables based on inputs, especially for those data not seen in the set used for training. Supervised machine learning has been widely applied in simulating soil N and C cycling. In recent decades, many studies showed that supervised machine learning models e.g., ANN (artificial neural networks), RF (random forest), SVR (support vector regression) and BRT (boosted regression tree), performed better than traditional models. For example, Yin *et al.* (2021) and Philibert *et al.* (2013) used RF to simulate global background N_2O emission in terrestrial ecosystems and N_2O emission from global cropland, respectively. ANN was applied to estimate soil nitric oxide (NO) emissions (Delon *et al.*, 2007) and global N_2O emissions from natural terrestrial ecosystems (Zhuang *et al.*, 2012). Yang *et al.* (2016b), Schillaci *et al.* (2017) and Wang *et al.* (2018) applied BRT models to simulate soil C concentration and stock, soil nutrient distribution in a regional scale. SVM has been widely used in predicting soil mineralization rate and N content in crops (Morellos *et al.*, 2016).

Table 2. Comparison of widely used supervised machine learning models

Supervised machine learning models	Description	Advantage	Disadvantage
Naive Bayesian classifier (Duda and Hart, 1973; Langley *et al.*, 1992)	• Based on Bayes' Theorem and the maximum posteriori hypothesis. • A probabilistic machine learning model used for classification task.	• Simple and easy to implement; • Handle both continuous and discrete data; • Fast (no iterations); • Perform well when the conditional Independence assumption holds.	• Possible to fail if the input data are not independent; • Zero probability problem.
Logistic Regression (Binomial logistics regression) (Cabrera, 1994; Peng *et al.*, 2002)	• A classification algorithm; • Used for binary dependent variable.	• Easy to implement, interpret, and very efficient to train; • Easy to extend to multiple classes or multinomial regression; • Inference about the importance of each input data; • Unlikely to overfit; • Efficient when input variables are linearly separable.	• May overfit with high dimensional datasets; Cannot handle non-linear problem, • Difficult to capture complex relationships, • Difficult to handle multicollinearity between input variables; • Difficult to handle irrelevant features; • Sensitive to outliers.
Support-vector machines (Cortes and Vapnik, 1995)	• Conduct one or more hyperplanes in a high- or infinite-dimensional space that is used for classification, regression and outliers detection (Bhavsar and Panchal, 2012).	• Effective in high dimensional spaces; • Handle small datasets; • Handle the nonlinear interactions of input variables; • Easy to generalization.	• Sensitive to missing value; • Not suitable for large data sets; • Difficult to select an appropriate Kernel function; • Requires feature scaling; • Difficult to interpret;
Random forests (Breiman, 2001)	• An ensemble learning method (bagging) for classification, regression by constructing a multitude of decision trees at training process.	• Unlikely to overfit; • Handle large datasets with high dimensionality; • Handle missing value; • Handle both categorical and continuous data; • Determine the importance of each input variable; • Handle the nonlinear interactions of input variables; • Robust to outliers.	• High computational expense; • May lead to overfit by noise; • Limited with Regression;
Gradient boosting decision tree (Multiple Additive Regression Tree) (Friedman, 2001, 2002)	• An ensemble learning method based on iterative decision tree algorithm. consist of multiple decision trees • Used for classification, regression	• Handle missing value; • Handles both categorical and continuous data; • Robust to outliers with optimizing different loss functions • High accuracy • No data pre-processing required; • Handle the nonlinear interactions of input variables; • Unlikely to overfit; • Determine the importance of each input variable	• May overemphasize outliers and cause overfitting (cross-validation needed); • High computational expense.
Artificial neural networks	• A series of algorithms that act like a human brain to recognize patterns among vast amounts of data.	• Robust to noise in the training data; • High accuracy; • Handle the nonlinear interactions of input variables; • No restrictions of the input variables • Better performance with large dataset	• Unexplained functioning of the network; • High computational expense; • Complicated model structure; • Difficult to interpret; • Large data requirement.

This successful application of machine learning models in N cycling demonstrated that data mining coupled with machine learning methods could capture the complex, dynamic, and nonlinear N processes more efficiently and effectively than traditional empirical and process-based models.

2.5 Summary of current knowledge gaps

Extensive literature review has been conducted on N loss from different pathways, their driving factors, different approaches of N loss predictions, and the limitations of existing process-based models. The identified knowledge gaps are:

1) Existing process-based models assume that the fractions of N_2O from nitrification and denitrification are fixed, resulting in inaccurate predictions of N loss in these pathways. More accurate parameters should be adopted to improve the prediction in process-based models.

2) The equations in submodules of process-based models in simulating N loss from different pathways are derived from limited experimental data and sometimes inconsistent, which could lead to simulation errors. More appropriate soil and environmental input variables should be identified and the complex relationships among input variables and N loss from different pathways should be explored and incorporated into submodules of process-based models.

3) Existing process-based models have limited capacities in extrapolating the simulation to a larger scale; detailed parameterization and comprehensive datasets are needed.

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CHAPTER 3

GENERAL METHODOLOGY

The research included data mining and machine learning model initializations and parameterizations in predicting N loss from different pathways. An incubation study was also conducted to investigate N_2O emission from different pathways and to identify the key factors affection N_2O emissions within Australia. The combination of methods was used to provide a comprehensive analysis of N loss from different pathways, their driving factors and mitigation strategies. The knowledge gained could help to improve the modelling capacity and understating of N cycling. In this chapter, the general materials and methods were described.

3.1 Data mining and database compilation

3.1.1 Literature search

In this project, extensive keywords were used for literature search from Web of Science (ISI), SCOPUS, CAB Abstracts (ISI), Academic Search Complete (EBSCO) and Google Scholar, the reference list of cited references were also cross-checked. The collected data were divided into different databases according to their N loss pathways. The keywords and selection criteria for each database are listed in Table 1. In summary, 186 observations (25 papers) published prior to 2018 were included in the nitrification database; a total of 3367 observations (225 studies) published between 1978 and 2019 were included in the denitrification database. 818 observations (286 studies) from 1986 to 2020 were included in the nitrate leaching database.

Table 1. Keywords and selection criteria used in the global literature search

Database	Keywords for literature search	Selection criteria
nitrification database	nitrification; N_2O/nitrous oxide emission; agriculture; cropping; pastures; forest and their combinations.	a) acetylene blockage technique or ^{15}N tracers was applied for N_2O emission from nitrification; b) study must reprot gross nitrification rates; c) study must report paired nitrification rates and associated N_2O emission; d) experimental location, design and conditions were given in details for cross-checking.
denitrification database	denitrification, N_2/dinitrogen, nitrogen (N) application, mitigation strategy, management practices, fertilizer type, soil amendment, inhibitor and their combinations	a) soil denitrification rate and N_2 emissions were measured in the topsoil (0-20 cm); b) experimental design or lab incubation conditions were given; c) studies were restricted to terrestrial ecosystems; d) experimental location, design and conditions were given in details for cross-checking.
nitrate leaching database	nitrate/ NO_3^- leaching, nitrogen (N) application, mitigation strategy, management practices, fertilizer type, soil amendment, inhibitor and their combinations	a) Studies should be field experiment only; b) experimental design was given; c) studies were restricted to agroecosystems; d) experimental location, design and conditions were given in details for cross-checking.

3.1.2 Data extraction

Based on the selection criteria, data were extracted and categorised into different sections. For basic information, it included study reference; experiment year; experiment type; experiment duration, study region/country, geographical location (longitude and latitude). Environmental information included climate zones (Köppen Climate Classification), mean annual precipitation (MAP), mean annual temperature (MAT), potential evapotranspiration rate (PET) and aridity index (the ratio of precipitation to evapotranspiration) (AI).

Topsoil physicochemical properties (soil texture; soil pH; water filled pore space (WFPS%); total carbon (%); organic carbon (SOC %); total N (TN %); C/N ratio; clay content (%); bulk density (g/cm^3), mineral N (mg N kg^{-1}), soil initial inorganic N (NO_3^--N, NH_4^+-N) content, etc. were recorded.

Land use; crop; irrigation; tillage; residue management; type of nitrogen fertilizer; application rate of N (kg N ha^{-1}); inhibitor type and rate; experimental treatment were recorded. The amount of N loss, measurement technique and measurement duration were included. WebPlotDigitizer version 4.1 was employed to digitize and extract data from figures (https://apps.automeris.io/wpd/). To decrease the outliers' impact and increase the normality, some data were natural log transformed for statistical analysis. TreeNet® (Salford Systems) was used to conduct SGB models.

3. 2 Machine learning models (Chapters 4 and 6)

3.2.1 Stochastic Gradient Boosting (SGB) Model

In recent decades, supervised machine learning models e.g., ANN (artificial neural networks, Delon et al., 2007; Zhuang et al., 2012), RF (random forest, Yin et al., 2021; Philibert et al., 2013), SVR (support vector regression, Morellos et al., 2016) and BRT (boosted regression tree, Wang et al., 2016b; Schillaci et al., 2017 and Wang et al., 2018), have been widely used and shown to outperform other traditional models in nitrogen and carbon cycling simulation. However, the model performance and interpretability are largely depended on the specific database quality and research purposes. Furthermore, a comprehensive comparison based on a literature search (Table 2 in Chapter 2) demonstrated that the SGB model has the capacity of handling missing value and categorical/continuous data, resisting outliers and overfitting, allowing nonlinear interactions between input variables and determining the importance of input variables. Therefore, considering the quality and quantity of databases based on literature in this

study, the SGB model was adopted to simulate and predict nitrification rate and the ratio of N_2O from nitrification, and nitrate leaching, respectively in Chapters 4 and 6. As a tree-based model, the accuracy of SGB model is enhanced by the sequential error-correcting process in each tree. The SGB model was firstly introduced and improved according to the gradient boosting decision tree (Breiman, 1996; Friedman, 2001, 2002). Different from boosting all the sample data at each iteration, random subsampling is used in SGB model. In this way, the computational complexity is reduced, and the learning speed is improved. Another feature of SGB model is the capacity of handling categorical and numerical variables (Friedman, 2001, 2002; Wang *et al.*, 2017). In SGB model, learn rate (LR), subsample fraction (SF), number of trees (NT), and minimum terminal node (MINCHILD) are user defined parameters. The learning processes of individual trees and NT are controlled by LR, contributing to the final model. To avoid model overfitting, SF is applied during the learning process. MINCHILD are regulated by the size of dataset. To achieve the optimal configurations, multiple combinations of those four parameters were tested (Wang *et al.*, 2016; Yang *et al.*, 2016; Zhang *et al.*, 2021). 10-fold cross-validation (CV) was adopted to evaluate the accuracy, which was particularly effective in small dataset learning without the need to reserve a separate test set (Kohavi, 1995). In 10-fold CV, the whole dataset was partitioned into ten folds, and each fold was treated as a test set individually while other nine folds were treated as the training set. Consequently, the averaged results were used to estimate the model performance.

3.2.2 Algorithm of Stochastic Gradient Boosting Model

Stochastic Gradient Boosting model was proposed in Friedman (2002). This algorithm is based on gradient boosting model with a minor modification. This model includes randomness in each iteration to reduce computational complexity and improve the learning speed of model.

Let $\{y_n, x_n\}\frac{N}{1}$ be the entire training data sample and $\{\pi(n)\}\frac{N}{1}$ be a random permutation of integers $\{1,2,3, \dots, N\}$. Then a random subsample $\tilde{N} < N$ is given by $\{y_{\pi(n)}, x_{\pi(n)}\}\frac{\tilde{N}}{1}$.

1. Initialize $F_0(x) = \arg min_\gamma \sum_{n=1}^{N} \psi(y^n, \gamma)$
2. For each *i*=1, 2, …, *I*, do:

(1) $\{\pi(n)\}\frac{N}{1}$ = rand_perm$\{n\}\frac{N}{1}$

(2) For each $n = 1,2, \dots, \tilde{N}$ do: $\tilde{r}\frac{\pi(n)}{i} = \left[\frac{\Theta\psi(y_{\pi(n)}, F(x_{\pi(n)})}{\Theta F(x_{\pi(n)})}\right]_{F=F_{m-1}}$

(3) Obtain the regression tree by $\left\{x_{\pi(n)}, \check{r}\frac{\pi(n)}{i}\right\}\frac{N}{1}$, then obtain nodes of the *i*th tree $\{R_i^j, j = 1,2, \dots, J\}$

(4) For each $j = 1,2, \dots, J$, $\gamma_i^j = \arg min_\gamma \sum_{x_{\pi(i)} \in R_i^j} \psi(y_{\pi(n)}, F_{i-1}(x_{\pi(n)}) + \gamma)$

(5) Update $F_i(x) = F_{i-1}(x) + v\sum_{j=1}^{J} \gamma_i^j I(x \in R_i^j)$

3. Output $\hat{F}(x) = F_I(x)$

If $\tilde{N} = N$, no randomness is introduced. The fraction of $f = \tilde{N}/N$ controls the randomness in successive iterations. More overall randomness is caused by smaller $f = \tilde{N}/N$.

3.2.3 Indices for model performance evaluation

Different indices were adopted to evaluate the performance of SGB model in this thesis, including regression coefficients of determination (R^2), mean absolute percentage error (MAPE), mean square error (MSE), the Nash–Sutcliffe model efficiency (NSE) (Nash and Sutcliffe, 1970) and root mean square error (RMSE).

$$R^2 = \frac{\sum_{i=1}^{n} {P_i - O_{avg}}^2}{\sum_{i=1}^{n} {O_i - O_{avg}}^2} \quad (1)$$

$$\text{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n} {P_i - O_i}^2} \quad (2)$$

$$\text{NSE} = 1 - \frac{\sum_{i=1}^{n} {P_i - O_i}^2}{\sum_{i=1}^{n} {O_i - O_{avg}}^2} \quad (3)$$

$$MAE = \frac{1}{n}\sum_{i=1}^{n} |O_i - P_i| \quad (4)$$

$$\text{MSE} = \frac{1}{n}\sum_{i=1}^{n} (P_i - O_i)^2 \quad (5)$$

$$\text{MAPE} = \frac{1}{n}\sum_{i=1}^{n} \left|\frac{O_i - P_i}{O_i}\right| \quad (6)$$

where P_i is its predicted value, O_i is the observed value, n is the number of data pairs. O_{avg} and P_{avg} are the averaged observed and its predicted values, respectively.

3.3 Linear mixed-effect models (Chapter 5)

In Chapter 5, LLMs were applied to explore the relationships between soil denitrification rate, soil N_2 emissions and the $N_2O/(N_2O+N_2)$ ratio from denitrification with soil and environmental properties. Linear mixed-effect models (LLMs) are extensions of simple linear models with fixed and random effects and are widely used for analyzing different types of data: longitudinal, clustered and panel data (Peng and Lu, 2012). In LLMs, fixed effects and random effects are used to model intra-cluster correlation between the responses, which are caused by unobserved heterogeneity at the cluster level (Peng and Lu, 2012). Fixed effects model the average trends while random effects model the extent to which these trends vary across levels of different grouping factors (e.g., studies or categories). Random effects are clusters of observations that come from the same study to explain the pattern from a particular group that may differ from the average trend (Brown, 2021). LLMs can handle missing data and unbalanced studies well by

introducing random effects, which is especially useful to address the limitation of literature-based data. Besides, LLMs can handle continuous input variables by providing coefficient estimates (Brown, 2021) to reflect the magnitude. LLMs have been used in examining the relationships between soil mineralization rate and nitrification rate and soil/ environmental properties (Li *et al.*, 2019; Li *et al.*, 2020).

A typical LLM can be presented as:

$$\ln y = \alpha + \beta x + \mathrm{T}_{study} + \varepsilon \quad (7)$$

Where y is soil denitrification rate and should be natural log transformed to satisfy the assumption of normality in the context of the model, soil N_2 emissions or the $N_2O/(N_2O+N_2)$ ratio from denitrification. α, β and ε are the intercept, slope and random error, respectively; x is the soil or environmental factors. T is the random effect factor. 'study' was considered as a random effect to reflect the non-independence among observations within the same experiment. The linear mixed-effect model was performed using *lme4* package (Bates *et al.*, 2014) and *lmerTest* package (Kuznetsova *et al.*, 2017). These analyses were conducted in R (Rstudio, 2020).

3.4 Meta-analysis (Chapter 5)

In Chapter 5, to investigate the effects of mitigation strategies on denitrification and N_2 emissions in the database. Paired observations were used to perform a meta-analysis. Meta-analysis is a series of statistical analyses that are applied to summarize different studies that investigated the same research questions by estimating measures of the overall effect, consistency and homogeneity among the studies, and whether the effect differs significantly from a null expectation (Rosenberg *et al.*, 2004). Traditional narrative reviews are usually subjective and prone to bias and error (Teagarden, 1989). Furthermore, traditional reviews may reach incorrect conclusions by missing important studies (Egger and Smith, 1997). On the contrary, meta-analysis applies objective

formulae, transparent review process. Combining all available data, a well-conducted meta-analysis allows for a more objective appraisal of different studies, which may address the uncertainties from a single study (Egger and Smith, 1997). Meta-analysis has been widely applied in evaluating N processes. For example, Lam *et al.* (2012) conducted a meta-analysis to evaluate the elevated CO_2 on N dynamics in grain crop and legume pastures systems. Xia *et al.* (2017) assessed the effects of different N management on staple grain yield, greenhouse gas and reactive N pollution.

For each mitigation strategy, paired observations were recorded (control and treatment). For N application, zero or the lowest N was set as the control and the other N application rates were set as treatments. For fertilizer type, urea as a widely used fertilizer was set as the control and other fertilizer types were treatments. Fertilizer types were sub-divided into organic fertilizers (animal manure and solid waste), ammonium-based fertilizers, nitrate-based fertilizers and NH_4NO_3 fertilizer. Management practices e.g., soil amendment (biochar, zeolite and lime) or the use of nitrification inhibitor compared to N fertilization only were included in the analysis. No tillage, no irrigation, residue removal, single fertilization and surface application of fertilizers were the controls, and tillage, irrigation, residue retention, split fertilization and incorporation of fertilizers were the corresponding treatments.

Effect sizes in meta-analysis were weighed by the number of replicates in a study (Lam *et al.*, 2012) to avoid extreme weights:

$$\text{Weight} = (n_T \times n_c)/(n_T + n_c) \quad (8)$$

Where n_T and n_c are the number of treatment replicates and that of control replicates.

The response ratio r is represented by the natural log and used as a metric for soil denitrification rate and N_2 emissions in response to management practices (Hedges *et al.*, 1999):

$$\ln r = \ln\left(\frac{\overline{x}^T}{\overline{x}^C}\right) \quad (9)$$

where $\overline{x}^T$ and $\overline{x}^C$ are the mean values of the treatment observation and the control observation, respectively. Results of the meta-analysis are reported as the percentage change of soil denitrification and N_2 emissions under treatment effects ((r-1) ×100). Fixed-effect meta-analysis with 4999 iterations bootstrapping was conducted to generate the mean effect size and the 95% confidence intervals (CIs) by MetaWin version 2.1 (Rosenberg *et al.*, 2000). The effects of mitigation strategies were considered significant if the 95% CIs did not overlap with zero.

3.5 Incubation study (Chapter 6)

Since 1980s, different techniques have been developed to partition N_2O production from different pathways. For example, the acetylene inhibition technique (AIT) was applied to differentiate between N_2O from nitrification and denitrification (Klemedtsson *et al.*, 1988; Inubushi *et al.*, 1996). However, the effectiveness of AIT can be highly affected by soil properties (De Boer and Kowalchuk, 2001). With the development of ^{15}N enrichment techniques, several methods were adopted to determine the contributions of individual N pools to N_2O emissions (Zhang *et al.*, 2015), e.g., source partitioning methods developed by Rütting *et al.* (2010) and (Stevens *et al.*, 1997), the inverse abundance approach by Stange *et al.* (2013) and ^{15}N tracing model by (Muller *et al.*, 2014). Among these methods, the source partitioning method could be applied to calculate the contribution of these pathways to N_2O production based on the dynamics of three N pools: soil ammonium, organic N and soil nitrate pools (Mosier *et al.*, 1986; Rütting *et al.*, 2010), which is a suitable tool to quantify process-based N_2O emission pathways (Muller *et al.*, 2014). To evaluate the contribution of N_2O production pathways of Australian forest soils in subtropical, temperate and arid regions and to investigate the relationships between

dominant N_2O production pathways in forest soils and driving factors, an incubation ^{15}N tracing experiment was carried out in Chapter 6.

3.5.1 Soil samples

Thirteen forest (dominated by Eucalyptus species) soils were sampled across Eastern coastal areas and inland areas of central Australia in May 2019. These soils were located in arid (n= 6), humid subtropical (n= 3) and temperate (n= 4) climates. At each site, topsoil (0-10 cm) was collected by mixing ten soil cores, homogenized thoroughly, and transported on ice to the laboratory. Soils were oven-dried at 40°C after removing roots and leaf residues, ground and sieved through a 2.0 mm mesh. The mean annual temperature (MAT [°C]) and mean annual precipitation (MAP [mm]) of individual sampling locations were extracted from the WorldClim database version 2 (http://www.worldclim.org/) using the '*raster*' and '*sp*' packages in R (Pebesma and Bivand, 2005; Hijmans *et al.*, 2019). Soil pH was measured by a 1: 2.5 (*w:w*) of soil and water ratio with a pH meter (Thermo Scientific Inc., Waltham MA, US). Total nitrogen [TN (%)] and total carbon [TC (%)] were determined by an isotope ratio mass spectrometer (Sercon Hydra, Crewe, UK).

3.5.2 ^{15}N tracing experiment

Soil microcosms were established using 250 ml flasks containing 20 g soil (oven-dry equivalent). Soils were pre-incubated at 25°C with 50% water-filled pore space (WFPS) for three weeks to stabilize the soil microbial communities. After pre-incubation, two labelled treatments i.e., $NH_4{}^{15}NO_3$ (9.7537 atom% excess) and $^{15}NH_4NO_3$ (9.8837 atom% excess) were applied as solution at 60 µg N g^{-1} dry soil (comprising 30 µg NH_4^+-N g^{-1} soil and 30 µg NO_3^--N g^{-1} soil), with three replicates. Soil moisture content in the flasks was adjusted to 55% WFPS upon addition of the N solution. Soils were incubated for 6 days at 25 ^oC in the dark.

After N application, headspace gas samples were collected at days 0, 2 and 6. On each gas sampling day, the flasks were opened and flushed with ambient air and resealed for 6 h before sampling. Gas samples were collected from each flask and injected into two pre-evacuated vials (12 ml) to determine the N_2O concentration by gas chromatograph (7890A; Agilent Technologies, Santa Clara, USA). A separate set of gas samples was injected into a 60 ml evacuated vial for $^{15}N_2O$ analysis by isotope ratio mass spectrometer (IRMS) (Sercon Hydra, Crewe, UK). Soils were destructively sampled for isotopic composition analysis after gas sampling. NH_4^+-N and NO_3^--N content in soils were analysed with 2 M KCl solution with a segmented flow analyzer (Skalar Analytical B. V. Tinstraat, Netherlands) and isotopic composition using a micro-diffusion method as described by Saghir *et al.* (1993) followed by IRMS. After extraction, soils were washed using distilled water, and oven-dried at 60 ^oC and the isotopic composition of the insoluble organic N was determined by IRMS.

3.5.3 Data analysis and calculations

Autotrophic nitrification, heterotrophic nitrification and denitrification were targeted in this study as the pathways of N_2O production. Three pools were considered as possible substrates for N_2O, which differed in their ^{15}N atom fraction, a_n (an autotrophic nitrification pool, assumed to be equivalent to NH_4^+-N), a_h (a heterotrophic nitrification pool, assumed to be equivalent to insoluble organic N), a_d (a denitrification pool, assumed to be equivalent to NO_3^--N) (Stevens *et al.*, 1997; Rütting *et al.*, 2010). The ^{15}N atom fraction of the resulting N_2O can be determined using the following equations (Rütting *et al.*, 2010).

$$a_{N2O} = n_d a_d + n_a a_a + n_h a_h \quad (10)$$

$$n_d + n_a + n_h = 1 \quad (11)$$

Where, a_{N2O} is the ^{15}N abundance of total N_2O produced during the incubation; n_d, n_a and n_h are N_2O emission fractions originating from the NO_3^-, NH_4^+ and insoluble organic N pool via denitrification, autotrophic nitrification and heterotrophic nitrification, respectively; the ^{15}N atom% excess from NO_3^-, NH_4^+ and insoluble organic N in the paired $^{15}NH_4NO_3$ and $NH_4\ ^{15}NO_3$ treatments were used into equations (10) and (11) enabled derivation of n_d, n_a, and n_h, respectively. a_d, a_a and a_h represent the ^{15}N abundance of NO_3^-, $NH_4^+$and insoluble organic N, respectively.

N_2O production rates originating from the NH_4^+ pool (autotrophic nitrification), the NO_3^- pool (denitrification) and the insoluble organic N pool (heterotrophic nitrification) were calculated using equations (12) to (14) as follows:

$$N_2O_d = n_d \times N_2O_t \quad (12)$$

$$N_2O_a = n_a \times N_2O_t \quad (13)$$

$$N_2O_h = n_h \times N_2O_t \quad (14)$$

Where N_2O_t is the total N_2O emission rate from incubated soil.

In summary, the general methods of each study were described above. Based on extensive literature searching, the extracted databases covered a wide range of data under different soil, climate and management conditions across the world. Data-driven models (machine learning models and linear mixed-effects models) can help identify the key variables and reveal their relationships to address the limitation of inconsistent responses of N loss to its key drivers in process-based models. Meta-analysis could provide a comprehensive synthesis to evaluate the mitigation strategies for reducing denitrification and associated gaseous losses. The ^{15}N tracing experiment can differentiate N_2O production pathways in Australian forest soils. By combing different methods, a better understanding of N loss from different pathways can be achieved.

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CHAPTER 4

Nitrification and its N_2O emission in terrestrial ecosystems: a global prediction based on data mining and a machine leaning model

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LETTERS

LETTER

New approach for predicting nitrification and its fraction of N_2O emissions in global terrestrial ecosystems

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Abstract

Nitrification is a major pathway of N_2O production in aerobic soils. Measurements and model simulations of nitrification and associated N_2O emission are challenging. Here we innovatively integrated data mining and machine learning to predict nitrification rate (R_{nit}) and the fraction of nitrification as N_2O emissions ($f_{N_2O_{Nit}}$). Using our global database on R_{nit} and $f_{N_2O_{Nit}}$, we found that the machine-learning based stochastic gradient boosting (SGB) model outperformed three widely used process-based models in estimating R_{nit} and N_2O emission from nitrification. We then applied the SGB technique for global prediction. The potential R_{nit} was driven by long-term mean annual temperature, soil C/N ratio and soil pH, whereas $f_{N_2O_{Nit}}$ by mean annual precipitation, soil clay content, soil pH, soil total N. The global $f_{N_2O_{Nit}}$ varied by over 200 times (0.006%–1.2%), which challenges the common practice of using a constant value in process-based models. This study provides insights into advancing process-based models for projecting N dynamics and greenhouse gas emissions using a machine learning approach.

1. Introduction

Nitrous oxide (N_2O) is a potent greenhouse gas and can cause ozone depletion (Ravishankara *et al* 2009, Robertson and Vitousek 2009). Nitrification, a microbial process that converts ammonium (NH_4^+) into nitrite and subsequently nitrate, is a major pathway of N_2O production, especially in aerobic soils. However, measurements of nitrification rate (R_{nit}) and how much nitrified N is emitted as N_2O are challenging. Many ecological process-based models, such as APSIM (Keating *et al* 2003, Holzworth *et al* 2014), DNDC (Li *et al* 1992, 2000, Zhang *et al* 2002), WNMM (Li et al 2007), and DAYCENT (Parton *et al* 1998, Del Grosso *et al* 2000) have embedded modules to predict R_{nit} and N_2O from nitrification. These models adopt the equations generated from limited empirical data, and nitrification is calculated as a function of soil water content, soil pH, soil temperature and soil NH_4^+ content. The models adopt a 'grey-box' to estimate N_2O emission from nitrification by assuming the fraction of nitrification as N_2O emissions ($f_{N_2O_{Nit}}$) is fixed, and further constraining it by soil water content or soil temperature. The use of a fixed fraction is known to be problematic in predicting N_2O emission because the proportion of N_2O emission from nitrification varies with soil and environmental conditions (Farquharson 2016). In addition, process-based models may not perform well when extrapolating from site-specific to a larger scale, mainly owing to their deficiency in capturing the key processes in response to driving factors, detailed parameterization, limited data availability and their limited capacity in handling complex interacting factors (Giltrap *et al* 2015, Leng and Hall 2020, Saha *et al* 2021).

Because of the nonlinearity between input and output variables and intricate interactions among these input variables (Butterbach-Bahl *et al* 2013), robust prediction of R_{nit} and associated N_2O production is difficult due to uncertainty in existing process-based models and limited measured data to improve those models. To address this issue, we adopted machine learning [stochastic gradient boosting (SGB) modelling] with data technologies and high-performance computing to identify patterns from

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global datasets and predict R_{nit} and $f_{N_2O_{Nit}}$. Models based on machine learning, such as ANN (artificial neural networks), RF (random forest), SVR (support vector regression) and BRT (boosted regression tree) have been recently applied in predicting soil biogeochemical processes. Previous studies showed that machine learning models performed better than traditional models in predicting N_2O and NO emission locally (Villa-Vialaneix *et al* 2012, Philibert *et al* 2013, Saha *et al* 2021) and globally (Delon *et al* 2007, Zhuang *et al* 2012, Perlman *et al* 2014), soil nitrogen (N) loss with biochar application (Liu *et al* 2019), soil organic carbon, and total N (Morellos *et al* 2016, Yang *et al* 2016, Schillaci *et al* 2017). Among the machine-learning based models, BRT (Friedman 2001, 2002, Elith *et al* 2008) relied on SGB, which is especially effective when quantifying nonlinear N transformations that are regulated by variables with complicated interactions, like R_{nit} and associated N_2O emissions. Furthermore, SGB is useful in handling relatively small datasets ($n < 100$) by adjusting the weights of training datasets and tuning learning rate based on the size of the datasets to reduce errors and overfitting (Shepherd *et al* 2003, Andonie 2010, Zhang and Ling 2018). As a variable selection process, SGB also excludes less important variables to achieve better generalization of the dependent variable (Xu *et al* 2014). From the functional perspective, process-based models need to define the function of each input variable. On the contrary, response functions and variable interactions could be handled in SGB models, because they do not pre-set response functions (e.g. linear or exponential) but assume a certain interaction between variables (Leng and Hall 2020). Thus, we hypothesized that these features of SGB models could improve prediction performances of R_{nit} and associated N_2O emissions at a large scale.

In this study, using a comprehensive database compiled from global literature, we first attempted to establish SGB models derived from this database to predict R_{nit} and $f_{N_2O_{Nit}}$, and to compare their performance with existing process-based models. The objectives of the study were: (a) to compare the performance of SGB and process-based models in predicting nitrification rate and associated N_2O production (b) to extend spatially the simulation of R_{nit} and $f_{N_2O_{Nit}}$ using the better performing model.

2. Methods

2.1. Database compilation

Extensive keyword searches of databases (Web of Science (ISI), SCOPUS, CAB Abstracts (ISI), Academic Search Complete (EBSCO) and Google Scholar) and the reference list of cited references were performed. The keywords used in the search were nitrification; N_2O/nitrous oxide emission/pathways; agriculture; cropping; pastures; forest and their combinations. Studies were included if they met the following criteria: (a) ^{15}N tracers or acetylene blockage technique was used to determine N_2O emission produced during nitrification; (b) gross nitrification rates (R_{nit}) were provided; (c) paired nitrification rates and associated N_2O emission were reported; (d) details on experimental location, design and conditions were given to enable cross-checking for duplicate publication; (e) instantaneous measurements were excluded due to high heterogeneity of outcomes. We identified 186 observations from 25 papers published prior to 2018 that satisfied the set criteria for this study. These observations were used to train the SGB models. All studies were conducted under laboratory incubation conditions probably owing to the technical limitations of simultaneous measurements of R_{nit} and $f_{N_2O_{Nit}}$ under field conditions. Data were log transformed to reduce the impact of outliers and improve normality for further analysis.

2.2. Comparing the performance of process-based models and SGB models

The algorithms in APSIM (Holzworth *et al* 2014), Crop-DNDC (Zhang *et al* 2002) and WNMM (Li *et al* 2007) simulate R_{nit} using the same input variables, viz. soil NH_4^+ content, soil pH, soil water content and soil temperature. The responses of nitrification to soil pH, soil water content and soil temperature during nitrification vary between these three models (figure 1). Crop-DNDC covers a wide range of soil water conditions with an increased response up to 80% WFPS. In APSIM and WNMM models, the thresholds of soil water content for nitrification are higher than that in Crop-DNDC. Nitrification increases with soil temperature in WNMM and APSIM models but decreases in Crop-DNDC when soil temperature is over 35 °C. These models estimate N_2O production from nitrification through multiplying the simulated nitrification rate by a fixed $f_{N_2O_{Nit}}$, and/or combining with functions of WFPS and soil temperature (figure 1). Specifically, APSIM simply adopts a simple default $f_{N_2O_{Nit}}$ of 0.002. WNMM considers soil temperature and soil water content for the calculation but sets a threshold $f_{N_2O_{Nit}}$ of 0.002. Crop-DNDC includes soil WFPS and temperature and adopts a constant $f_{N_2O_{Nit}}$ of 0.0006. The compiled database (section 2.1) was used to calculate the nitrification rate and N_2O from nitrification based on the equations (see supplementary information 1 (available online at stacks.iop.org/ERL/16/034053/mmedia)) of WNMM, Crop-DNDC and APSIM.

A SGB model is a tree-based model; each tree is built in a sequential error-correcting process to converge to an accurate model. The SGB model was proposed and modified based on the gradient boosting decision tree (Breiman 1996, Friedman 2001, 2002). SGB model includes random subsampling, which draws a subsample of training data randomly instead

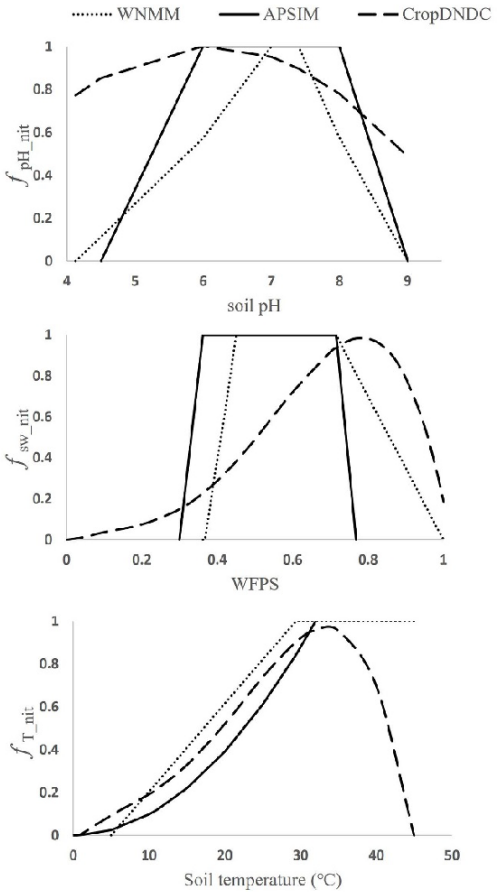


Figure 1. The effects of soil pH, soil temperature and WFPS on nitrification in process-based models (ASPIM, WNMM and CropDNDC).

of boosting all the sample data at each iteration (see supplementary information 3). This modification reduces the computational complexity and improves the model learning speed. The SGB model can also handle both numerical and categorical variables as features (Friedman 2001, 2002, Wang *et al* 2017). To compare with the three process-based models we also adopted the same input variables, i.e. soil NH_4^+ content, soil pH, soil water content and soil temperature to build an SGB model, referred to as SGB1. Four parameters that can be configured in a SGB model are the learn rate (LR), subsample fraction (SF), number of trees (NT), and minimum terminal node (MINCHILD). Learn rate controls the contribution of each tree to the final model. Subsample fraction is involved in the learning process to prevent model from overfitting. The number of trees is based on the learn rate. Minimum terminal nodes are regulated by the size of dataset. To perform the regularization, we tested different combinations of LR (0.001, 0.005, 0.01), SF (0.3, 0.6, 0.8, 0.9), NT (1000, 2000, 3000) and MINCHILD (3, 6, 9) parameters, and found that the optimal configurations for SGB1 were 0.01, 0.8, 3000 and 3 for LC, SF, NT and MINCHILD, respectively (Friedman 2001, Yang *et al* 2016, Wang *et al* 2018) (figure S1). The accuracy of the model was evaluated by 10-fold cross-validation (CV), where the entire dataset was first used for learning purposes, and subsequently partitioned into ten bins. For each of the 10 folds, nine folds were used as a training set and the remaining fold as a test set. The test results from each fold were averaged to estimate the whole model performance. CV is particularly useful for small datasets when one cannot afford to reserve some data for testing (Kohavi 1995) (figure S2).

Regression coefficients of determination (R^2), root mean square error (RMSE) and the Nash–Sutcliffe model efficiency (NSE) (Nash and Sutcliffe 1970) (see supplementary information 2), were used to measure the percentage of variation explained by the model and the model accuracy for SGB1 and the three process-based models.

2.3. Prediction of R_{nit} and $f_{N_2O_{Nit}}$ under different soil properties and climate conditions

We attempted to extend the prediction of R_{nit} and $f_{N_2O_{Nit}}$ to a larger scale by applying SGB technique to our global database. It is unrealistic to perform dynamic prediction of R_{nit} and $f_{N_2O_{Nit}}$ owing to data limitation. Previous studies proved that soil properties (clay content, pH, etc) and climatic conditions (MAT and MAP) can affect microbial community biomass and composition in the soil (Avrahami *et al* 2003, Hu *et al* 2016, Liu *et al* 2017), which are the potential key drivers of the nitrification process and associated N_2O emissions. We therefore performed R_{nit} and $f_{N_2O_{Nit}}$ prediction using long-term edaphic and environmental conditions as input variables.

We followed a series of stepwise procedures (Thompson 1978, Moisen *et al* 2006) to eliminate redundant input variables and optimize the number of variables for this SGB model (referred to as SGB2). This improves the applicability of SGB2 without compromising much prediction capacity. The stepwise variable selection method is efficient in computation and has been widely used in machine learning variable selection (Guyon *et al* 2002, Rakotomamonjy 2003). Since SGB2 was built based on incubation studies conducted under *in vitro* conditions mostly with optimal soil temperature, soil moisture and N availability, the prediction reflects R_{nit} and $f_{N_2O_{Nit}}$ under such conditions.

We further included a set of constraints to regulate the prediction results by SGB2. We first set soil total N and soil organic carbon as trigger conditions, which are the prerequisites for nitrification. Second, desert areas with low precipitation show limited potential for nitrification (Noy-Meir 1974, Skujiņš 1981). In this regard, typical desert areas (mean annual precipitation (MAP) <250 mm) (Marshak 2010) have been excluded from the prediction. To adjust the deviation

between incubation temperature and long-term temperature, we incorporated a function of mean annual temperature (MAT) against nitrification rate to reflect the *in situ* situations globally. The complete model is described as follows:

$$R_{\mathrm{nit}} = M_{\mathrm{SGB2}} \cdot \gamma = \prod_{\alpha \in F} \delta_\alpha \cdot f(T) \cdot M_{\mathrm{SGB2}} \quad (1)$$

$$\gamma = \delta_F \cdot f(T) \quad (2)$$

$$f(T) = \begin{cases} 0 & \text{if } T < 2.8 \\ 0.0023T2 + 0.0032T - 0.006 & \text{if } 2.8 \leqslant T < 8 \\ 0.0006T^{2.2188} & \text{if } 8 \leqslant T < 28 \\ 1 & \text{if } T \geqslant 28 \end{cases} \quad (3)$$

(Malhi and McGill 1982)

$$\delta_F = \prod_{\alpha \in F} \delta_\alpha \quad (4)$$

$$\delta_{\mathrm{SOC}} = \begin{cases} 0 & \text{if SOC} = 0 \\ 1 & \text{if SOC} > 0 \end{cases} \quad (5)$$

$$\delta_{\mathrm{TN}} = \begin{cases} 0 & \text{if TN} = 0 \\ 1 & \text{if TN} > 0 \end{cases} \quad (6)$$

$$\delta_P = \begin{cases} 0 & \text{if } P < 250 \\ 1 & \text{if } P \geqslant 250 \end{cases} \quad (7)$$

Among these equations, M_{SGB} is the original SGB2 prediction, γ is the constraint coefficient of TN, SOC, MAP (P) and MAT (T), $F = \{\mathrm{TN}, \mathrm{SOC}, P\}$ are a set of total N, soil organic carbon and MAP. δ_F is the constraint coefficient of F, δ_α is the constraint coefficient involved in δ_F, where $\delta_\alpha \in \{\delta_{\mathrm{SOC}}, \delta_{\mathrm{TN}}, \delta_P\}$, and $f(T)$ is the function to determine the impact of MAT on R_{nit}. Apart from R^2, RMSE and NSE, additional mean absolute error (MAE) was used to measure the accuracy of prediction for selecting the optimal SGB model when mapping global prediction of R_{nit} and $f_{\mathrm{N_2O_{Nit}}}$.

To project R_{nit} and $f_{\mathrm{N_2O_{Nit}}}$ at a global scale using SGB, potential driving factors of soil and climate data were obtained from different sources and integrated into a spatial GIS database. The soil property database with a spatial resolution of 1 km^2 was obtained from the World Inventory of Soil Emission (WISE) database developed by the International Soil Reference and Information Centre (ISRIC) (www.isric.org) (Batjes 2015). The climate data at 0.5° resolution was collected from Climatic Research Unit (CRU TS v4.01) (www.cru.uea.acuk/data/) from 1911 to 2010 (Harris *et al* 2014). The ESRI ArcGIS software (v. 10.4.1 ESRI) was used to plot the global distribution map. In this study, all SGB models were constructed using TreeNet® (Salford Systems).

3. Results

3.1. Overview of the literature-based dataset

Study sites spanned from 122° W to 152° E and 43° S to 65° N. For most of the incubation experiments, the soil temperature was controlled at 20 °C–25 °C, with a range of 5 °C–45 °C, and average soil moisture of around 45% WFPS. The average R_{nit} in the topsoil (0–20 cm) was 1.4 kg N ha^{-1} d^{-1} and varied widely. The average $f_{\mathrm{N_2O_{Nit}}}$ was 0.46% (0.004%–9.19%) (table 1 and figure S3).

3.2. Comparison of model performance

SGB1 outperformed the other three process-based models with the highest R^2 and NSE value and lowest RMSE in predicting nitrification (0.73, 0.63 and 0.4, respectively) and N_2O emission during nitrification (0.61, 0.54 and 0.61, respectively) (table 2). The R^2 values of R_{nit} for APSIM, Crop-DNDC and WNMM were 0.27, 0.31 and 0.24, respectively. WNMM and Crop-DNDC overestimated nitrification rate by 1.45 kg N ha^{-1} d^{-1} and 3.39 kg N ha^{-1} d^{-1}, respectively, whereas APSIM underestimated it by 1.72 kg N ha^{-1} d^{-1} (figure S4). As for the N_2O emission during nitrification calculated using a fixed ratio and functions of soil water content and soil temperature, all three process-based models did not perform very well (table 2). Crop-DNDC overestimated N_2O emission from nitrification by 1.08 kg N ha^{-1} d^{-1}. APSIM and WNMM underestimated N_2O emission when it was <0.3 g N ha^{-1} d^{-1} but overestimated it when the N_2O emission from nitrification was higher (figure S5).

3.3. Stepwise variable selection of optimal models

Nitrification rate showed a strong relationship with MAT and soil C/N ratio in the model A1 (table 3). The highest R^2 value of 0.8 was achieved when at least six variables were included (models A5–A8). When only MAT, soil C/N ratio and soil pH were included (model A2), the performance reached 0.76 for both R^2 and NSE (table 3), which we considered optimal for further upscale predictions when compromising between the R^2 value and the number of variables included.

When predicting $f_{\mathrm{N_2O_{Nit}}}$, the performance of model B3 was significantly increased with four input variables (MAP, soil clay content, soil pH and soil TN), yielding a relatively higher R^2 value (0.55) and NSE (0.55), and lower MAE (0.26) and RMSE (0.40) than other models (table 4). Therefore, B3 model (table 2, SGB2) was considered optimal for predicting $f_{\mathrm{N_2O_{Nit}}}$.

3.4. Global mapping of the response of R_{nit} and $f_{\mathrm{N_2O_{Nit}}}$ to soil properties and climate conditions

We extend the prediction of R_{nit} spatially using SGB2 (model A2 in table 3) with input variables soil pH,

Table 1. Summary statistics of nitrification, the fraction of associated N_2O emissions and environmental variables from global literature.

	n	Mean	Median	Min	Max	SD
R_{nit} (kg N ha^{-1} d^{-1})	186	1.40	0.29	0.003	27.5	3.9
$f_{N_2O_{Nit}}$ (%)	186	0.46	0.12	0.004	9.20	1.18
MAP (mm)	186	887	700	423	2416	470
MAT (°C)	186	13.5	14.3	−0.6	25.0	5.0
Soil temperature (°C)	181	21.3	22.0	5.0	45.0	5.2
Soil clay content (%)	139	23.2	19.4	4.0	60.6	16.4
Soil pH	186	6.4	6.4	3.7	8.4	1.2
SOC (%)	185	2.4	2.0	1.0	9.5	1.7
Soil TN (%)	114	0.28	0.18	0.09	2.23	0.34
Soil C/N ratio	116	11.7	11.5	2.3	22.2	3.8
Soil NH_4^+–N (mg kg^{-1})	154	18.1	13.0	0.3	116.0	22.1
Soil NO_3^-N (mg kg^{-1})	154	35.9	11.0	1.0	1157.0	120.7
Soil WFPS (%)	174	44.5	42.0	24.0	90.0	13.1
N input (kg N ha^{-1})	186	23.7	8.7	0.0	300.0	48.3

soil C/N ratio and MAT from global soil and climate databases. Nitrification rate varied spatially, spanning a wide range of 0.0001–2.19 kg N ha^{-1} d^{-1} with an average of 0.11 kg N ha^{-1} d^{-1}(figure 2). The highest R_{nit} was observed in subtropical and equatorial areas. Higher nitrification rates generally occur in soils with a lower C/N ratio and a neutral pH (figure S6). $f_{N_2O_{Nit}}$ covered a broad range from 0.006% to 1.24% (figure 3) with an average of 0.13%. Lower $f_{N_2O_{Nit}}$ was noted in tropical and higher latitude regions in the northern hemisphere, with higher $f_{N_2O_{Nit}}$ across the subtropical and around equatorial areas. A large proportion of N_2O from nitrification was found in soils with a lower clay content (figure S7). When soil is neutral pH, more N_2O was released from nitrification compared to both acidic and alkaline soils (figure S7). $f_{N_2O_{Nit}}$ followed the pattern of MAP and soil total N (figure S7).

4. Discussion

4.1. Better performance of SGB models

SGB1 better predicts nitrification and associated N_2O emissions than the three widely used process-based models using the same input variables (soil NH_4^+ content, soil pH, soil water content and soil temperature). There are several possible reasons. First, limited site-specific experimental data were used for deriving process-based model parameters and equations, where less responsive or redundant variables might have been included. On the contrary, the relationships between variables in SGB1 were more representative as their internal interactions were developed from a relatively comprehensive global database.

Second, process-based models may have misused the variable responses for R_{nit} and $f_{N_2O_{Nit}}$ in their equations. For example, the relatively narrow range of the response of soil water to nitrification limited the prediction capacity of the APSIM and WNMM models. If soil water content was beyond of the lower and upper limits set in the equations in these models, nitrification was assumed to stop. In particular we found that APSIM and WNMM estimated that no nitrification occurred below 30% WFPS. However, the measured data from our global database indicated that nitrification did occur under 30% WFPS (Maag and Vinther 1996).

Third, the equations currently used in process-based models cannot account for the interactions of input variables (Butterbach-Bahl *et al* 2013) without comprehensive calibration using more site-specific detailed data. In contrast, SGB1 examines all possible nonlinear relationships and interactions between input variables themselves and output variables (Ryo and Rillig 2017).

4.2. The use of SGB models in spatial prediction globally

The spatial patterns of predicted R_{nit} and $f_{N_2O_{Nit}}$ exhibited a large variation owing to the diverse soil and environmental conditions and their interactions (figures 2 and 3). This result further shows that using a constant $f_{N_2O_{Nit}}$ to estimate the N_2O emission from nitrification by existing process-based models is unsuitable (Chen *et al* 2008, Farquharson 2016).

The relationship between soil properties and R_{nit} has been well studied (Dancer *et al* 1973, Nyborg *et al* 1988, Bengtsson *et al* 2003, Zebarth *et al* 2015). A recent global-scale study reported that soil C/N ratio, soil pH and MAT are the key drivers of R_{nit} (Li *et al* 2020). High soil C/N ratio may stimulate N immobilization, decrease availability of NH_4^+ for nitrification, and subsequently lower nitrification rate (Bengtsson *et al* 2003). Larger microbial populations and activities (including nitrifiers) are generally found in neutral rather than in high or low pH soils (Tabatabai *et al* 1992). MAT can regulate soil nitrification rate directly and indirectly by changing microbial community structure and abundance in the long term (Avrahami *et al* 2003, Hu *et al* 2016). Besides, MAT

Table 2. Comparison of observed and predicted nitrification and associated N_2O emissions by SGB1, APSIM, Crop-DNDC and WNMM models.

	SGB1				APSIM				Crop-DNDC				WNMM			
Output	*n*	R^2	RMSE	NSE	*n*	R^2	RMSE	NSE	*n*	R^2	RMSE	NSE	*n*	R^2	RMSE	NSE
Log_Nit[a]	186	0.73	0.40	0.63	99	0.27	0.71	−0.02	162	0.31	0.68	−1.04	98	0.24	0.66	−0.45
Log_N_2O_Nit[b]	186	0.61	0.54	0.61	99	0.10	0.84	−0.51	162	0.20	0.73	−14.2	98	0.11	0.68	−0.89

[a] Nitrification rate.

[b] N_2O emission from nitrification, RMSE: root mean square error and NSE: the Nash–Sutcliffe model efficiency.

Table 3. Model prediction of R_{nit} by stepwise selection of variables.

Model	Stepwise variable selection	R^2	MAE	RMSE	NSE
A1	MAT, soil C/N ratio	0.73	0.29	0.40	0.73
A2	MAT, soil C/N ratio, soil pH	0.76	0.26	0.37	0.76
A3	MAT, MAP, soil C/N ratio, soil pH	0.76	0.26	0.37	0.76
A4	Land use, MAT, MAP, soil C/N ratio, soil pH	0.79	0.25	0.35	0.79
A5	Land use, MAT, MAP, soil C/N ratio, soil pH, SOC	0.80	0.25	0.34	0.80
A6	Land use, MAT, MAP, soil C/N ratio, soil pH, SOC, soil TN	0.80	0.25	0.34	0.80
A7	Land use, MAT, MAP, soil C/N ratio, soil pH, SOC, soil TN, soil clay content	0.80	0.25	0.34	0.80

Table 4. Model prediction of $f_{N_2O_{Nit}}$ by stepwise selection of variables.

Model	Stepwise variable selection	R^2	MAE	RMSE	NSE
B1	MAP, soil clay content	0.39	0.30	0.48	0.35
B2	MAP, soil clay content, soil pH	0.44	0.30	0.46	0.41
B3	MAP, soil clay content, soil pH, soil TN	0.55	0.26	0.40	0.55
B4	MAP, soil clay content, soil pH, soil TN, soil C/N ratio	0.57	0.26	0.39	0.57
B5	Land use, MAP, soil clay content, soil pH, soil TN, soil C/N ratio	0.58	0.26	0.39	0.58
B6	Land use, MAP, MAT, soil clay content, soil pH, soil TN, soil C/N ratio	0.57	0.26	0.39	0.56
B7	Land use, MAP, MAT, soil clay content, soil pH, SOC, soil TN, soil C/N ratio	0.54	0.27	0.41	0.53

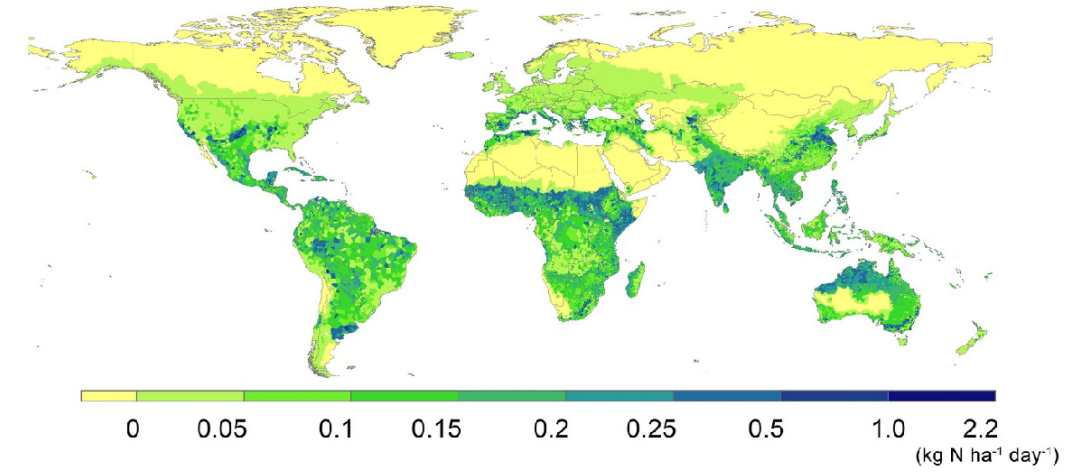


Figure 2. Global distribution of R_{nit} predicted by SGB (A2) model.

reflects the environmental conditions on site and regulates the long-term soil microclimate.

The spatial distribution of $f_{N_2O_{Nit}}$ was consistent with land use distribution (figure S8), which reflects the edaphic and climatic variations across terrestrial systems. The estimated range of $f_{N_2O_{Nit}}$ in humid tropical areas by B3 (table 4) was comparable to the DNDC and DLEM models examined by Inatomi *et al* (2019), but much lower than VISIT (Inatomi *et al* 2010) model estimates. However, $f_{N_2O_{Nit}}$ in semiarid regions was predicted to be lower by DNDC, DELM and VISIT than the B3 model. The differences in model predictions can be explained by three reasons. First, Inatomi *et al* (2019) adopted common protocols and initial and boundary conditions when upscaling the simulation for all three process-based models. Second, for the VISIT model, the parameterization of $f_{N_2O_{Nit}}$ was only dependent on soil pH, which is insufficient to reflect the complicated relationship between $f_{N_2O_{Nit}}$ and soil and environmental factors. Third, soil moisture and temperature in semiarid regions were below the threshold set for nitrification to occur

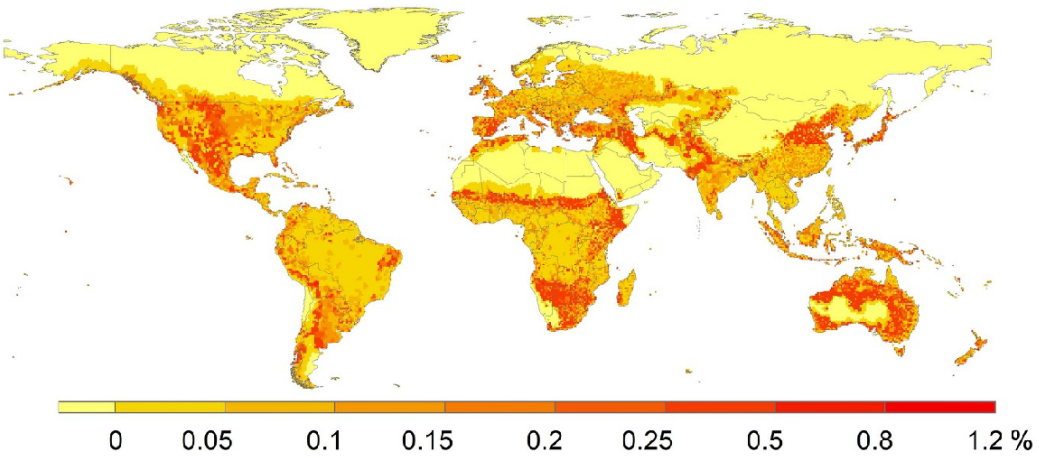


Figure 3. Global distribution of $f_{N_2O_{Nit}}$ predicted by SGB (B3) model.

in process-based models, thereby underestimating nitrification and associated N_2O emissions, whereas our A2 and B3 models cover a wider range of input variables.

In our study, subtropical areas and equatorial areas have higher R_{nit} and $f_{N_2O_{Nit}}$ than other areas. Higher MAT, neutral soil pH and lower soil C/N ratio could increase the microbial populations in soils and nitrifier efficiency, thus increasing nitrification rate (Gilmour 1984, Tabatabai *et al* 1992). The highest R_{nit} and $f_{N_2O_{Nit}}$ were observed in areas with intense human activities, i.e. cropland and grasslands (Ambus 1998). These could be attributed to long-term anthropogenic N input, increasing soil N availability for nitrification and associated N_2O production (Tabatabai *et al* 1992). On the other hand, optimal MAP provided an ideal oxygen level and moderate moisture condition for nitrifiers (Yu and Zhuang 2019). Areas with extensive forest and savannas have the lowest R_{nit} and $f_{N_2O_{Nit}}$ worldwide. This finding indicates that in temperate and tropical forest areas, nitrification is not a major source of N_2O emissions, which is in agreement with previous studies (Stehfest and Bouwman 2006, Werner *et al* 2007, Cheng *et al* 2012, Zhuang *et al* 2012).

Responses and changes of R_{nit} and $f_{N_2O_{Nit}}$ are highly related to long-term environmental, edaphic factors and land use. The fraction of nitrification as N_2O emissions is clearly not a constant; instead it should be adjusted according to edaphic and environmental conditions when used in process-based models or global climate models in projecting N_2O emissions. We are aware of the limited datasets used to develop the SGB2 models for global prediction and did not intend to accurately quantify R_{nit} and $f_{N_2O_{Nit}}$ at any timepoint. Our objective was to demonstrate that SGB2 models can be used to map the global spatial patterns of potential R_{nit} and $f_{N_2O_{Nit}}$ based on a few long-term soil and climate variables that are easily accessible from world databases. These potential R_{nit} and $f_{N_2O_{Nit}}$ values obtained under optimal soil temperature, soil moisture and sufficient N availability, could be used as benchmark for the calibration of process-based models.

Our findings provide important implications for the prediction of the N losses in process-based models. The SGB model is able to derive new parameters that can be incorporated into process-based models for different N loss pathways under various edaphic and environmental conditions. Moreover, SGB could also be directly embedded in a process-based model or replace the existing unsatisfactory modules of a process-based model to capture the complex, dynamic, and nonlinear processes more efficiently and effectively. By integrating with other modules of process-based models, SGB models can be used to develop decision support tools for sustainable N management.

4.3. Limitation

Limitations remained in this study. First, we conducted a comprehensive global literature search, but found a low number of observations. This indicates limited geographical coverage and the difficulties of simultaneously measuring nitrification and associated N_2O emissions both *in situ* and in the laboratory. Second, while SGB models are suitable for handling small databases, the inclusion of more data in training machine learning models would likely improve their performance. Third, although we included constraint coefficients in SGB models to address the issues associated with artificial experimental conditions, this potential data bias can be reduced when more *in situ* data become available.

5. Conclusion

This study is the first attempt to use machine learning to develop SGB models to predict R_{nit} and $f_{N_2O_{Nit}}$ in terrestrial ecosystems. Compared to three widely used process-based models, the SGB models were more accurate in predicting nitrification rate and associated N_2O emissions. The SGB models can be extended to a global level with only a few input variables without compromising accuracy. In particular, nitrification rate was predicted by soil pH, MAT and soil C/N ratio whereas the fraction of nitrification as N_2O emissions was predicted using soil pH, MAP, soil total N and clay content. Large spatial variation in nitrification and its fraction as N_2O emissions was mainly driven by long-term environmental and edaphic factors. The fraction of nitrification as N_2O emissions is clearly not constant; instead it should be adjusted according to edaphic and environmental conditions when used in process-based models or global climate models in projecting N_2O emissions.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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Supplementary Information for

New approach for predicting nitrification and its fraction of N_2O emissions in global terrestrial ecosystems

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This document includes:

1. Supplementary Information Text

1.1 Equations of nitrification and N_2O emission in process-based models

1.1.1 WNMM

1.1.2 Crop-DNDC

1.1.3 APSIM

1.2 Indices for model performance evaluation

1.3 Algorithm of Stochastic Gradient Boosting Model

2. Figures S1 to S8

Figure S1. Mean square errors (MSE) reduction at different levels of the parameters against number of trees. a) Learn rate, b) subsample fraction and c) Minimum terminal node

Figure S2. Mean square errors (MSE) reduction of training and validation datasets for SGB1. a) nitrification rate, b) N_2O emission from nitrification.

Figure S3 Distribution of nitrification, the fraction of associated N_2O emissions from global literature

Figure S4. Scatter plot of the observed against predicted nitrification rate by SGB1, APSIM, crop-DNDC and WNMM models.

Figure S5. Scatter plot of the observed against predicted nitrification associated N_2O emission by SGB1, APSIM, crop-DNDC and WNMM models.

Figure S6. Global soil and climate basic information used in predicting $\boldsymbol{R_{nit}}$. a) Soil pH, b) Mean annual temperature, c) Soil C/N ratio.

Figure S7. Global soil and climate basic information used in predicting $\boldsymbol{f_{N_2O_Nit}}$. a) Soil pH, b) Mean annual precipitation, c) Soil clay content, d Soil total nitrogen.

Figure S8. Global map of land cover–type climatology. Adapted from "A global land cover climatology using MODIS data", Broxton *et al* 2014.

3. SI References

4. Other Supplementary materials for this manuscript include the following:

4.1 Dataset reference list

1. Supplementary Information Text

1.1 Equations of nitrification and N_2O emission in process-based models

1.1.1 WNMM

$$R_{nit} = NH_4[1 - \exp(-k_{nit})]$$

$$K_{nit} = f_{T_nit} f_{sw_nit} f_{pH_nit}$$

$$f_{T_nit} = 0.41\frac{T_{soil} - 5}{10}, T_{soil} > 5℃$$

$$f_{sw_nit} = \begin{cases} \dfrac{SW - WP}{SW25 - WP}, & SW < SW25 \\ 1, & SW25 \le SW \le FC \\ 1 - \dfrac{SW - FC}{PO - FC}, & SW > FC \end{cases}$$

$$f_{pH_nit} = \begin{cases} 0.307pH - 1.269, pH < 7 \\ 1, 7 \le pH \le 7.4 \\ 5.367 - 0.599pH, pH > 7.4 \end{cases}$$

$$f_{\mathrm{N_2O_Nit}} = \frac{N_2O_{nit}}{R_{nit}} = \alpha_{nit} f_T f_{sw_{nit}}$$

Where NH_4 is NH_4^+–N content in the soil layer kg N ha, f_{pH_nit}, f_{T_nit} and f_{sw_nit} are the functions of soil pH, soil temperature °C and water content factors respectively, SW is soil water content v/v, WP is wilting point v/v, SW25=WP+ 0.25FC-WP, FC is field capacity v/v, and α_{nit} is the fraction of nitrification for N_2O emission default value is 0.002 (Li *et al* 2007).

1.1.2 Crop-DNDC

$$R_{nit} = NH_4[1 - \exp(-K_{35} f_{T_nit})] f_{sw_nit} f_{pH_nit}$$

$$f_{pH_nit} = -0.0604pH^2 + 0.7347pH - 1.2314$$

$$f_{sw_nit} = -12.904WFPS^4 + 17.651WFPS^3 - 5.5368WFPS^2 + 0.9975WFPS - 0.0243$$

$$f_{T_{nit}} = -0.0272(0.1T)^4 + 0.1566(0.1T)^3 - 0.2234(0.1T)^2 + 0.03094T - 0.0233$$

$$f_{\mathrm{N_2O_Nit}} = \frac{\mathrm{N_2O_{nit}}}{\mathrm{R_{nit}}} = 0.0006WFPS2.72^{34.6 - 9615/T + 273.15}$$

Where K_{35} has a default value of nitrification rate at 35°C with 25 mg/kg/day, f_{pH_nit}, f_{T_nit} and f_{sw_nit} are the effects of pH, temperature and soil moisture on nitrification respectively (Zhang *et al* 2002).

1.1.3 APSIM

$$\mathrm{R_{nit}} = K_{max}\frac{[\mathrm{NH_4}]}{[\mathrm{NH_4}] + K_{NH_4}}[\mathrm{NH_4}]f_{T_nit}f_{sw_nit}f_{pH_nit}$$

$$f_{T_nit} = \begin{cases} T/32^2, & T > 0; \\ 0, & T \le 0. \end{cases}$$

$$f_{\mathrm{N_2O_Nit}} = \frac{\mathrm{N_2O_{nit}}}{\mathrm{R_{nit}}} = 0.002$$

$[NH_4]$ is the ammonium concentration in the soil mg/kg, K_{max} is the maximum nitrification rate default setting of 40 mg/kg/day, KNH4 is the NH_4^+ concentration for half the maximum response to $[NH_4]$ default setting of 90 mg/kg, f_{pH_nit}, f_{T_nit} and f_{sw_nit} are functions accounting for the limitations imposed by pH, temperature and soil water content, and scaled from 0 to 1. The moisture factor is equal to 1.0, if soil moisture content is above one fourth higher than the lower limit to drained upper limit. It decreases linearly to zero when soil moisture drops to lower limit or increases to saturation water content. N_2O from nitrification is a fixed ratio with 0.002 (Holzworth *et al* 2014).

1.2 Indices for model performance evaluation

$$R^2 = \frac{\sum_{i=1}^{n} P_i - O_{avg}{}^2}{\sum_{i=1}^{n} O_i - O_{avg}{}^2}$$

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n} P_i - O_i{}^2}$$

$$\mathrm{NSE} = 1 - \frac{\sum_{i=1}^{n} P_i - O_i{}^2}{\sum_{i=1}^{n} O_i - O_{avg}{}^2}$$

$$MAE = \frac{1}{n}\sum_{i=1}^{n} |O_i - P_i|$$

Where n is the number of data pairs, O_i is the observed value, P_i is its predicted value. O_{avg} and P_{avg} are the averaged observed and its predicted values, respectively.

1.3 Algorithm of Stochastic Gradient Boosting Model

Stochastic Gradient Boosting model was proposed in Friedman *(2002).* This algorithm is based on gradient boosting model with a minor modification. This model includes randomness in each iteration. This modification could help to reduce computational complexity and improve the learning speed of model.

Let $\{y_n, x_n\}\frac{N}{1}$ be the entire training data sample and $\{\pi(n)\}\frac{N}{1}$ be a random permutation of integers $\{1,2,3, \ldots, N\}$. Then a random subsample $\tilde{N} < N$ is given by $\{y_{\pi(n)}, x_{\pi(n)}\}\frac{\tilde{N}}{1}$.

4. Initialize $F_0(x) = \arg min_\gamma \sum_{n=1}^{N} \psi(y^n, \gamma)$
5. For each i=1, 2, …, I, do:

(6) $\{\pi(n)\}\frac{N}{1}$ = rand_perm$\{n\}\frac{N}{1}$

(7) For each $n = 1,2, \ldots, \tilde{N}$ do: $\tilde{r}\frac{\pi(n)}{i} = \left[\frac{\Theta\psi(y_{\pi(n)}, F(x_{\pi(n)}))}{\Theta F(x_{\pi(n)})}\right]_{F=F_{m-1}}$

(8) Obtain the regression tree by $\left\{x_{\pi(n)}, \tilde{r}\frac{\pi(n)}{i}\right\}\frac{N}{1}$, then obtain nodes of the ith tree $\{R_i^j, j = 1,2, \ldots, J\}$

(9) For each $j = 1,2, \ldots, J$, $\gamma_i^j = \arg min_\gamma \sum_{x_{\pi(i)} \in R_i^j} \psi(y_{\pi(n)}, F_{i-1}(x_{\pi(n)}) + \gamma)$

(10) Update $F_i(x) = F_{i-1}(x) + v\sum_{j=1}^{J} \gamma_i^j I(x \in R_i^j)$

6. Output $\hat{F}(x) = F_I(x)$

If $\tilde{N} = N$, no randomness is introduced. The fraction of $f = \tilde{N}/N$ controls the randomness in successive iterations. More overall randomness is caused by smaller $f = \tilde{N}/N$.

2. Figures S1 to S8

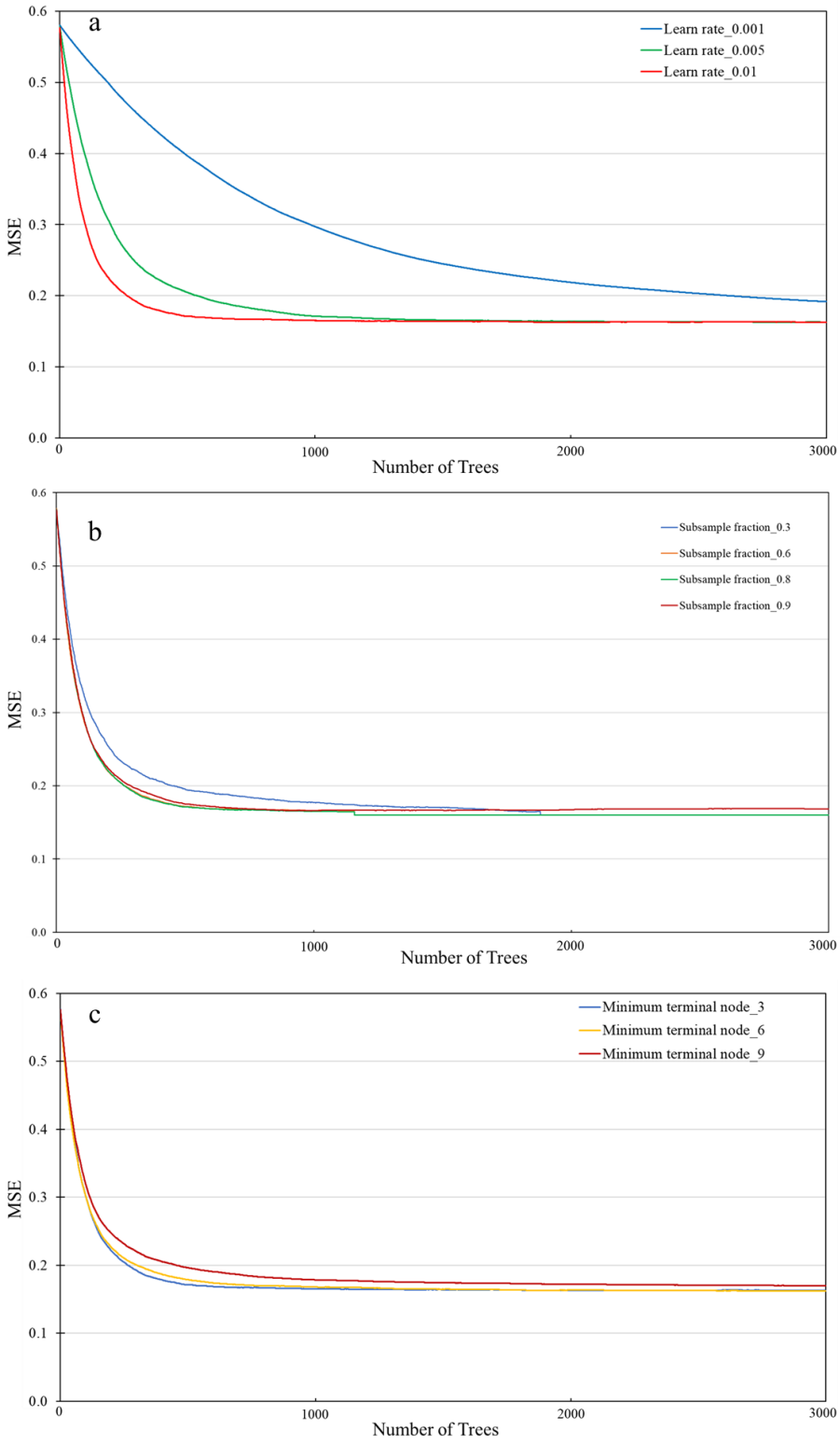


Figure S1. Mean square errors (MSE) reduction at different levels of the parameters against number of trees. a) Learn rate, b) subsample fraction and c) Minimum terminal node

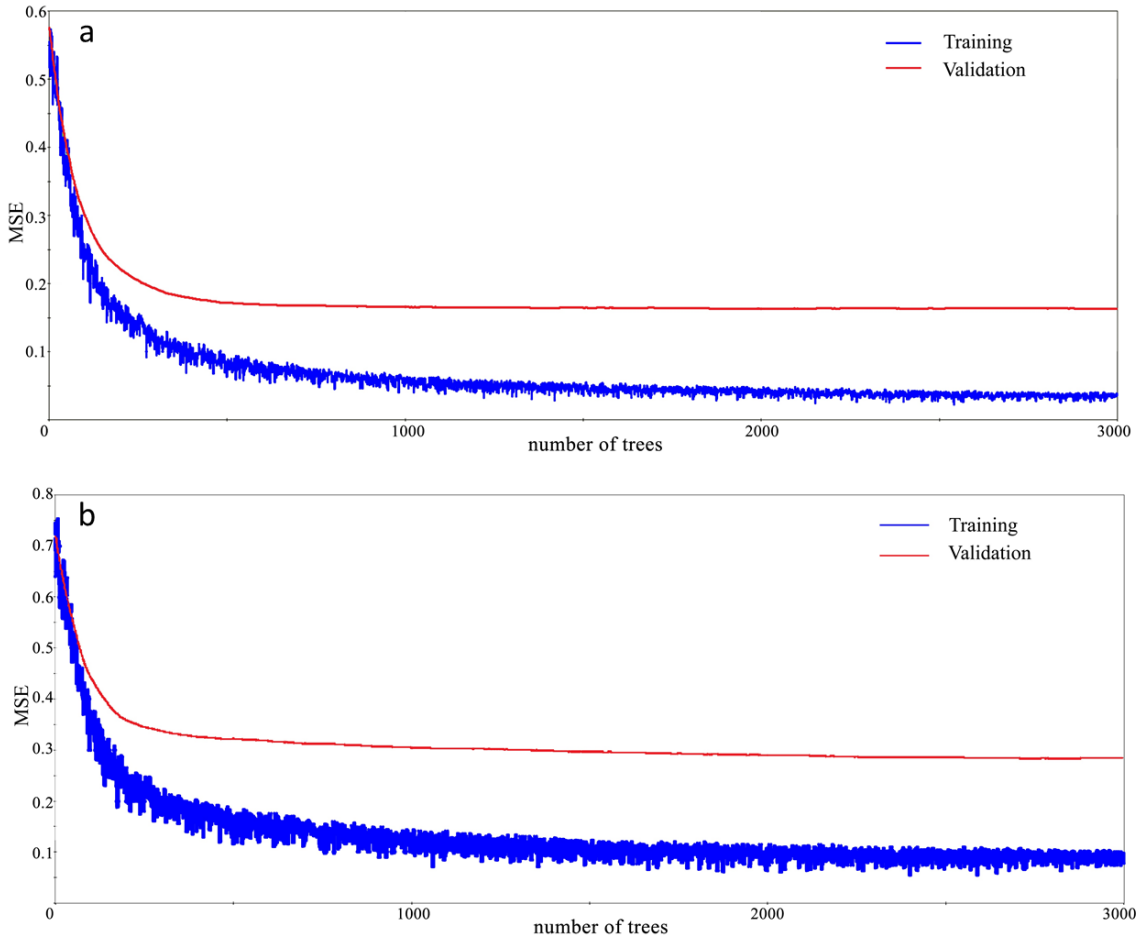


Figure S2. Mean square errors (MSE) reduction of training and validation datasets for SGB1. a) nitrification rate, b) N_2O emission from nitrification.

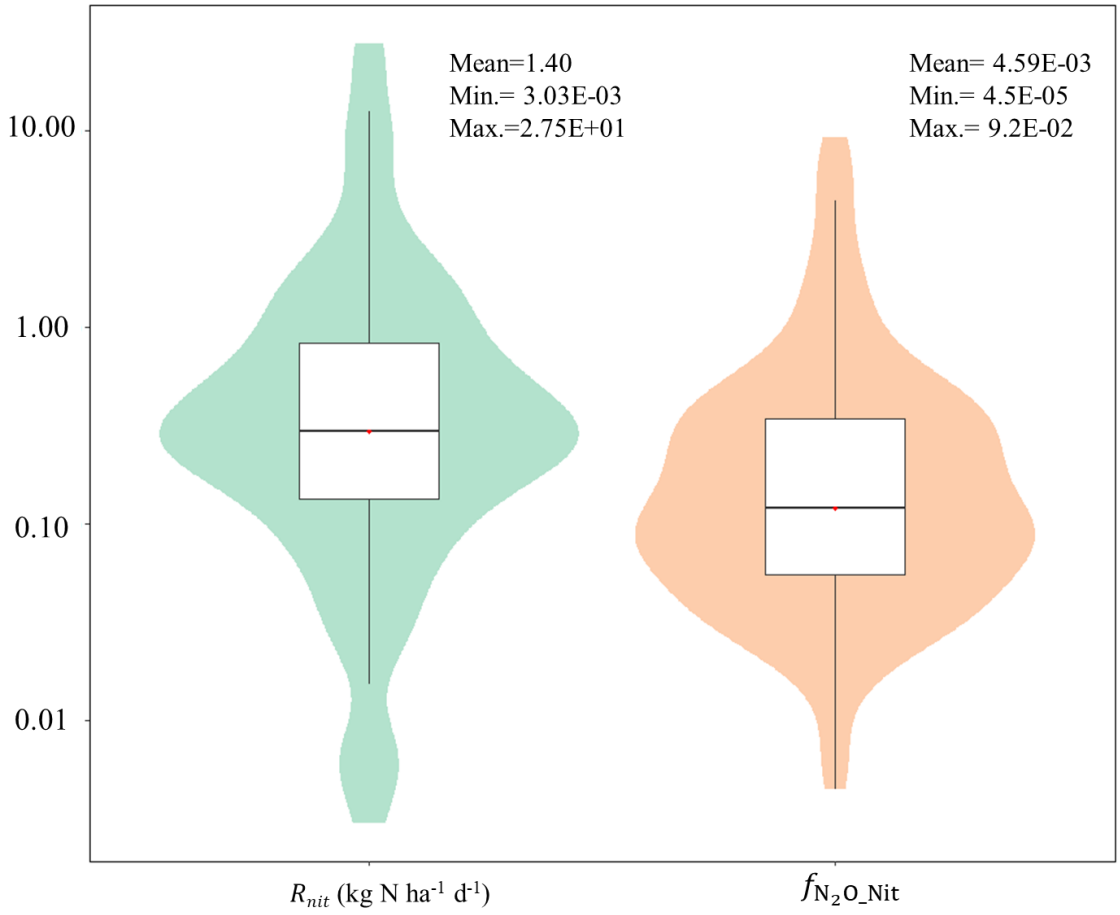
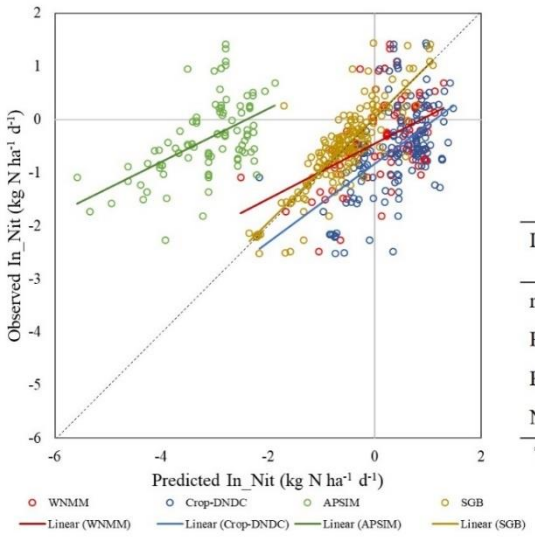


Figure S3. Distribution of nitrification, the fraction of associated N_2O emissions from global literature.

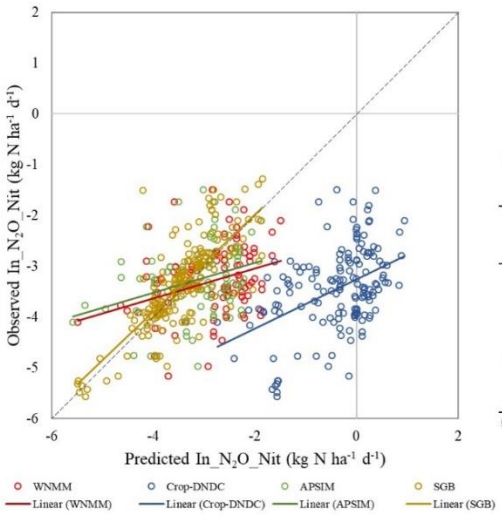


Comparison of observed and predicted nitrification by SGB1, APSIM, Crop-DNDC and WNMM models

Log_Nit*	SGB1	APSIM	Crop-DNDC	WNMM
n	186	99	162	98
R^2	0.73	0.27	0.31	0.24
RMSE	0.40	0.71	0.68	0.66
NSE	0.63	-0.02	-1.04	-0.45

*nitrification rate, RMSE: root mean square error and NSE: the Nash–Sutcliffe model efficiency.

Figure S4. Scatter plot of the observed against predicted nitrification rate by SGB1, APSIM, crop-DNDC and WNMM models



Comparison of observed and predicted nitrification associated N_2O emissions by SGB1, APSIM, Crop-DNDC and WNMM models

Log_Nit[§]	SGB1	APSIM	Crop-DNDC	WNMM
n	186	99	162	98
R^2	0.61	0.10	0.20	0.11
RMSE	0.54	0.84	0.73	0.68
NSE	0.61	-0.51	-14.2	-0.89

[§]N_2O emission from nitrification, RMSE: root mean square error and NSE: the Nash–Sutcliffe model efficiency.

Figure S5. Scatter plot of the observed against predicted nitrification associated N_2O emission by SGB1, APSIM, crop-DNDC and WNMM models.

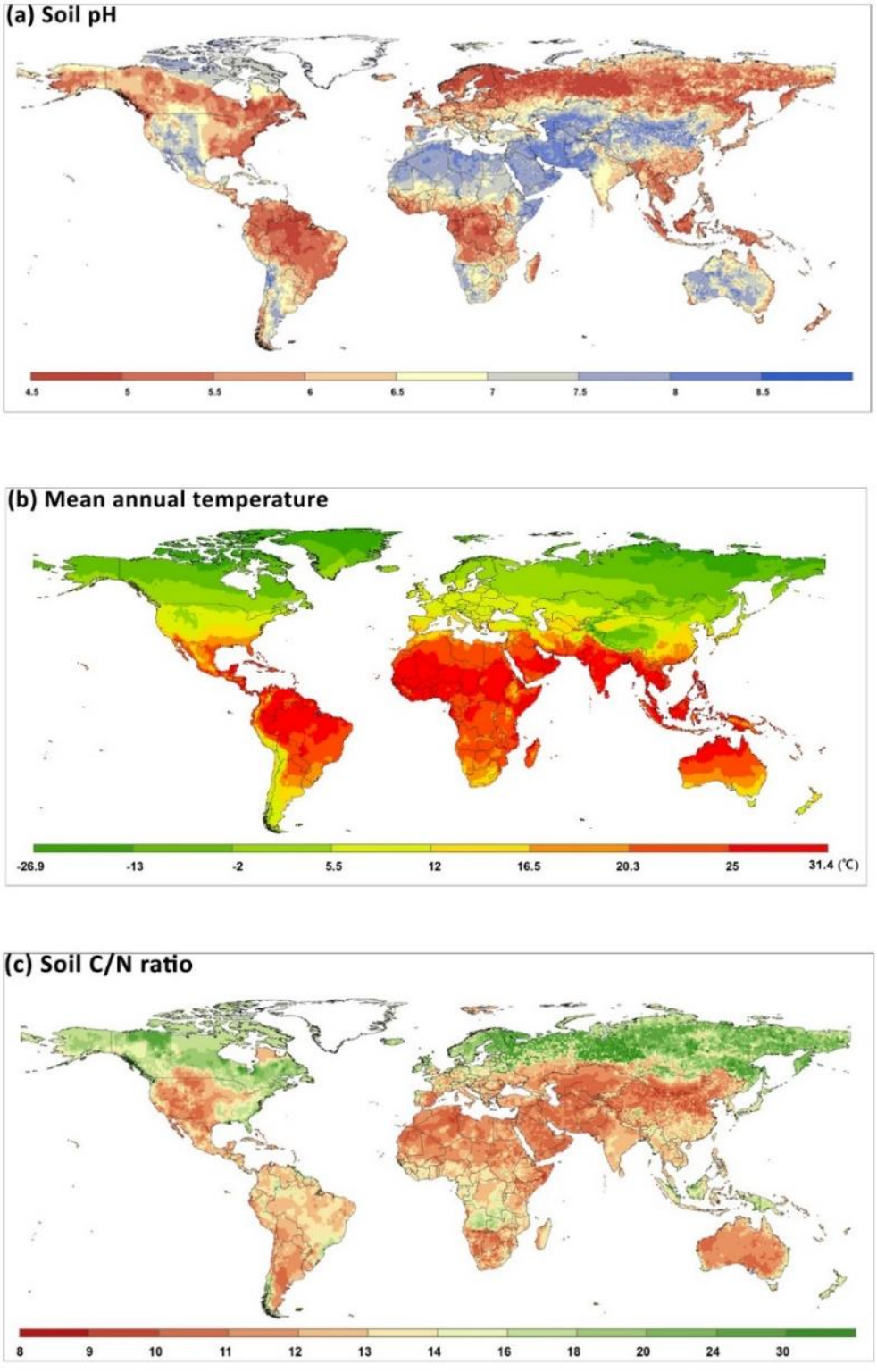


Figure S6. Global soil and climate basic information used in predicting R_{nit}. a) Soil pH, b) Mean annual temperature, c) Soil C/N ratio.

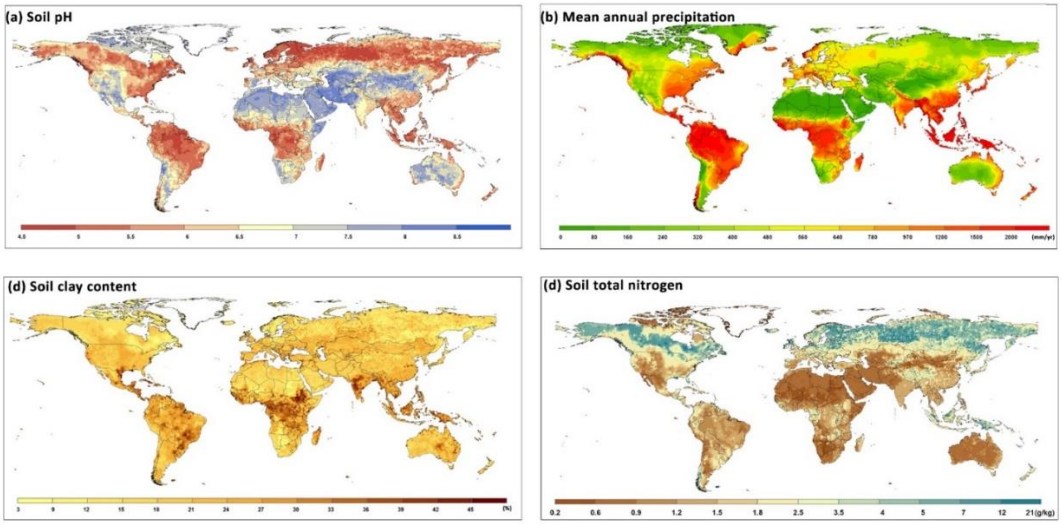


Figure S7. Global soil and climate basic information used in predicting $f_{\mathrm{N_2O_Nit}}$. a) Soil pH, b) Mean annual precipitation, c) Soil clay content, d) Soil total nitrogen.

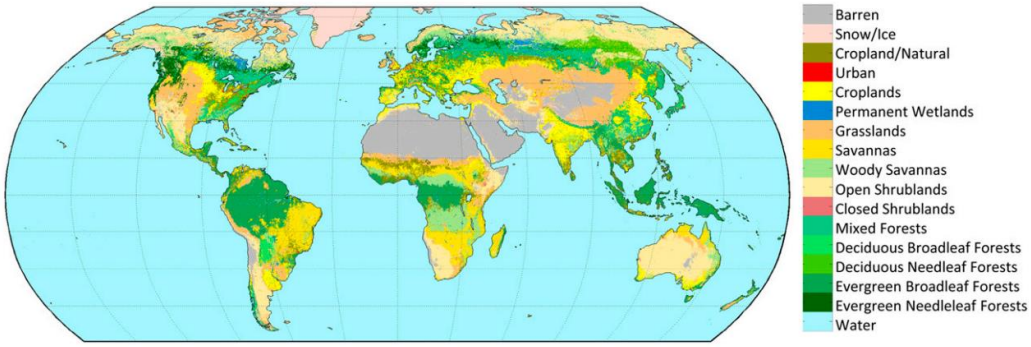


Figure S8. Global map of land cover–type climatology. Adapted from "A global land cover climatology using MODIS data", Broxton *et al* 2014 © American Meteorological Society. Used with permission.

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Chapter 5

Nitrogen loss via denitrification in terrestrial ecosystems: a global synthesis based on data mining.

This chapter is based on a published manuscript entitled:

Pan, B., Xia, L., Lam, S. K., Wang, E., Zhang, Y., Mosier, A., & Chen, D. (2022). A global synthesis of soil denitrification: Driving factors and mitigation strategies. *Agriculture, Ecosystems & Environment*, *327*, 107850.

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A global synthesis of soil denitrification: Driving factors and mitigation strategies

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ABSTRACT

Dinitrogen (N_2) and nitrous oxide (N_2O) produced via denitrification may represent major nitrogen (N) loss in terrestrial ecosystems. A global assessment of soil denitrification rate, $N_2O/(N_2O+N_2)$ ratio, and their driving factors and mitigation strategies is lacking. We conducted a global synthesis using 225 studies (3367 observations) to fill this knowledge gap. We found that daily N loss through soil denitrification varied with ecosystems and averaged 0.25 kg N ha^{-1}. The average emission factor of denitrification (EF_D) was 4.8%. The average $N_2O/(N_2O+N_2)$ ratio from soil denitrification was 0.33. Soil denitrification rate was positively related to soil water-filled pore space (WFPS) ($p < 0.01$), nitrate (NO_3^-) content ($p < 0.05$) and soil temperature ($p < 0.01$), and decreased with higher soil oxygen content ($p < 0.01$). N_2 emissions increased with latitude ($p < 0.05$), WFPS ($p < 0.01$) and soil mineral N ($p < 0.05$) but decreased with soil oxygen content ($p < 0.05$). The $N_2O/(N_2O+N_2)$ ratio increased with soil oxygen content ($p < 0.01$) but decreased with organic carbon (C) ($p < 0.05$), C/N ratio ($p < 0.01$), soil pH ($p < 0.05$) and WFPS ($p < 0.01$). We also found that optimizing N application rates, using ammonium-based fertilizers compared to nitrate-based fertilizers, biochar amendment, and application of nitrification inhibitors could effectively reduce soil denitrification rate and associated N_2 emissions. These findings highlight that N loss via soil denitrification and N_2 emissions cannot be neglected, and that mitigation strategies should be adopted to reduce N loss and improve N use efficiency. Our study presents a comprehensive data synthesis for large-scale estimations of denitrification and the refinement of relevant parameters used in the submodels of denitrification in process-based models.

1. Introduction

Denitrification is a microbial process by which nitrate (NO_3^-) is gradually reduced to nitrous oxide (N_2O) and dinitrogen (N_2) (Zaman et al., 2012); it is a major pathway of nitrogen (N) loss in terrestrial ecosystems. N_2O emissions contribute to global warming and stratospheric ozone depletion whereas N_2 losses decrease nitrogen use efficiency (NUE) in croplands and grasslands (Bouwman et al., 2013). The N loss through soil denitrification from terrestrial ecosystems was estimated to be 105–185 Tg N yr^{-1} (Tiedje, 1988) and 166 Tg N yr^{-1} (Scheer et al., 2020), with around 66–87 Tg N yr^{-1} in global croplands (Hofstra and Bouwman, 2005; Seitzinger et al., 2006) and 5.6 Tg N yr^{-1} in temperate grasslands (Saggar et al., 2013). N_2 represents a substantial N loss accounting for up to 85% of total denitrification (year 2000) (Bouwman et al., 2013) but it is often overlooked and poorly studied as N_2 is non-reactive (Almaraz et al., 2020). The quantification of N_2 emissions from terrestrial ecosystems remains challenging because of the high background atmospheric N_2 concentration (Zaman et al., 2012; Saggar et al., 2013). Many empirical and process-based models were developed to estimate N_2O and N_2 emissions from denitrification based on a fixed N_2O to N_2 ratio (NOE model) (Hénault et al., 2005) or combined with functions of soil properties (e.g., APSIM and DayCent model) (Del Grosso et al., 2000; Keating et al., 2003; Stehfest and Muller, 2004). However, the N_2O and N_2 emissions from soil denitrification showed a large spatial and temporal heterogeneity at a global scale (Butterbach-Bahl and Dannenmann, 2011). Therefore, a better understanding

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of the $N_2O/(N_2O+N_2)$ ratio and its driving factors could improve the estimation of denitrification in earth system models (Almaraz et al., 2020).

Soil denitrification and the emissions of its end-products (N_2O and N_2) are regulated by soil and environmental factors. For example, soil NO_3^- availability affects the denitrification rate (Abbasi and Adams, 1999; Senbayram et al., 2019). Higher carbon (C) availability could increase the denitrification rate but decrease the N_2O/N_2 ratio (Clough et al., 1998; Gillam et al., 2008). The effect of soil pH on denitrification is controversial. It is generally accepted that acidic soil has a lower soil denitrification rate (Sun et al., 2012) and higher $N_2O/(N_2O+N_2)$ ratio (Zaman et al., 2007; Zaman and Nguyen, 2010). Soil pH can also indirectly affect denitrification because nitrification is inhibited in acidic soils and subsequently less NO_3^- is available for denitrification (Tiedje, 1988). On the contrary, Šimek et al. (2000) found no significant correlation between soil pH and denitrifying enzyme activity. Soil oxygen concentration, water content and rainfall are other important factors as anaerobic conditions favor denitrification (De Klein and Van Logtestijn, 1996; Ellis et al., 1998; Rudaz et al., 1999; Hefting et al., 2003; Saggar et al., 2004). Generally, soil temperature is positively related to soil denitrification rate and associated N_2O emissions (Knowles, 1982; Ryden, 1983). However, some studies demonstrated that N_2O emissions may decrease with increasing temperature, because elevated temperature may enhance anaerobiosis and decrease the N_2O/N_2 ratio (Butterbach-Bahl and Dannenmann, 2011). These inconsistencies highlight the need of a comprehensive assessment to identify the important drivers of soil denitrification rate and its end-products.

Mitigation strategies have been widely investigated and adopted to lower soil denitrification and increase NUE. Optimizing N application rate could reduce the excess N that is subjected to denitrification (Morley et al., 2014; Lai and Denton, 2018; Senbayram et al., 2019). The application of soil amendments e.g., lime (McMillan et al., 2016) and biochar (Cayuela et al., 2013) generally raised soil pH and increased the activities of N_2O reductase that might subsequently decrease N_2O emissions from denitrification. Reduced crop residue retention decreases organic C input for soil denitrifiers (Chintala et al., 2015; Sun et al., 2018) but may reduce the potential of C sequestration and affect crop yield (Xia et al., 2018). Avoiding excessive irrigation could effectively maintain oxygen supply in the soil and lower denitrification (Huang et al., 2015). The application of nitrification inhibitors can reduce the NO_3^- availability for denitrification by inhibiting nitrification without impairing crop yields (McGeough et al., 2012; Wang et al., 2017). A systematic synthesis of mitigation strategies for denitrification and N_2 is vital for improving global fertilizer NUE and environmental quality.

In this study, we compiled a comprehensive database based on peer-reviewed studies worldwide. We attempted to (1) summarize and quantify soil denitrification rate, N_2 emissions and the $N_2O/(N_2O+N_2)$ ratio across different land use systems and climatic zones; (2) quantify the relationships between soil/environmental factors and soil denitrification rate, N_2 emissions and the $N_2O/(N_2O+N_2)$ ratio; and (3) evaluate the effects of mitigation strategies on soil denitrification rate and N_2 production.

2. Data and Methods

2.1. Database compilation

An extensive keyword search was conducted from Web of Science (ISI), CAB Abstracts (ISI) and Google Scholar between 1978 and 2019. The keywords used for searching include the combinations of denitrification, N_2/dinitrogen, nitrogen (N) application, mitigation strategy, management practices, fertilizer type, soil amendment and inhibitor, $N_2O/(N_2O+N_2)$ ratio. The database was also cross-checked with the studies included in recent key review papers of denitrification by Wang et al. (2018) and Almaraz et al. (2020). These papers did not examine the aspects of land use types or mitigation strategies of soil denitrification and associated gaseous losses. Literature included in our database fulfilled the following selection criteria: (a) soil denitrification rate and its N_2O or N_2 emissions in the topsoil (0–20 cm) were specified; (b) experimental design or lab incubation conditions were given; (c) studies were restricted to terrestrial ecosystems. After data collection, a total of 3367 observations (1599 observations for soil denitrification rate, 860 observations for soil N_2 emissions and 908 observations for the $N_2O/(N_2O+N_2)$ ratio from soil denitrification) were included in the database.

In the database, geographic coordinates (latitude and longitude) of the study sites, environmental conditions (mean annual temperature [MAT] and mean annual precipitation [MAP]), soil physicochemical properties (e.g., soil texture, clay content, soil water filled pore space (WFPS), soil temperature under incubation condition, soil pH, soil total nitrogen (TN) and organic carbon (SOC) content, soil C/N ratio, NH_4^+ and NO_3^- concentrations at the beginning of the experiment, etc.), and management practices (N application rate and farming practices) were extracted. This database was used for conducting linear mixed-effect models (Section 3.2) and a meta-analysis (Section 3.3). To decrease the outliers' impact and increase the normality, some data were natural log transformed when necessary for statistical analysis.

The database covered the dominant vegetation types (e.g., upland fields, paddy fields, grasslands, forests and wetlands). Overall, these sites spanned from 123.27°W to 176.07°E and from 42.97°S to 60.62°N (Fig. 1). These sites were classified into four major climatic zones according to Köppen Climate Classification (tropical, dry, temperate, and continental climates).

2.2. Data analyses

We calculated the emission factor of denitrification (EF_D) from field experiments conducted for at least a cropping season using the following equation:

$$EF_D(\%) = \left(\frac{\text{cumulative denitrification N loss } (N_2O + N_2)_{\text{treatment}} - \text{cumulative denitrification N loss}(N_2O + N_2)_{\text{control}} \left(\text{kg N ha}^{-1}\right)}{\text{N Fertilizer/manure } \textit{applied} \left(\text{kg N ha}^{-1}\right)} \right) * 100 \quad (1)$$

We then used linear mixed-effect models to explore the relationships between soil denitrification rate, soil N_2 emissions and the $N_2O/(N_2O+N_2)$ ratio from denitrification with soil and environmental properties:

$$\ln y = \alpha + \beta x + \mathrm{T}_{study} + \varepsilon \quad (2)$$

Where y is soil denitrification rate and should be natural log transformed to satisfy the assumption of normality in the context of the model, soil N_2 emissions or the $N_2O/(N_2O+N_2)$ ratio from denitrification. α, β and ε are

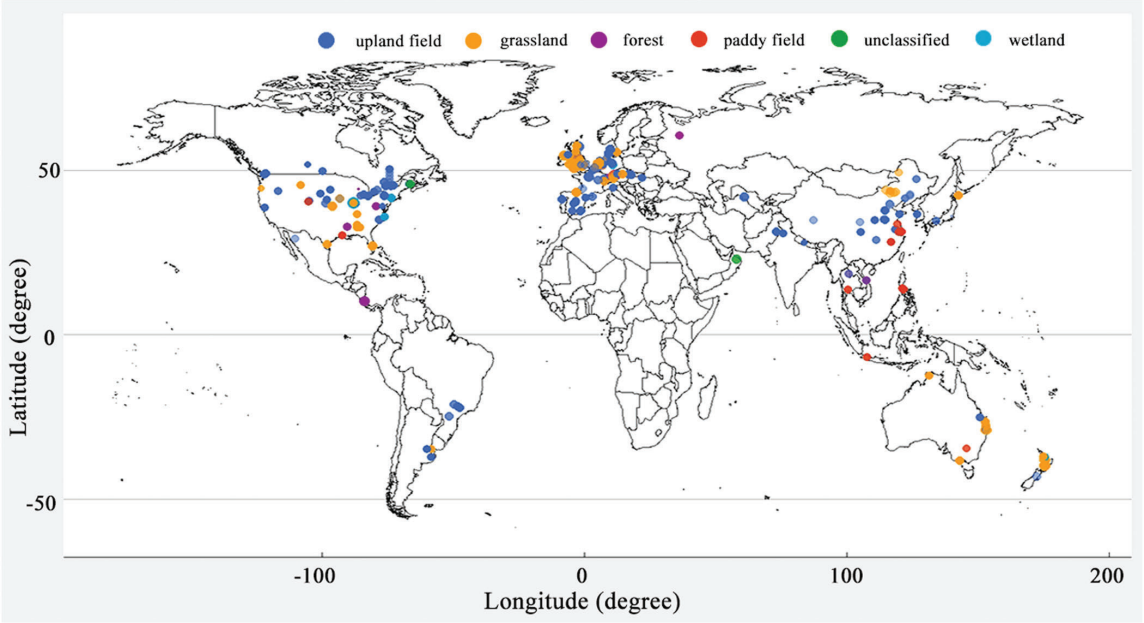


Fig. 1. Location of study sites included in the database.

the intercept, slope and random error, respectively; x is the soil WFPS, soil O_2 concentration, SOC, C/N ratio, NO_3^- concentration, soil temperature, soil mineral N content, soil pH, N application rate or latitude. T is the random effect factor. We considered 'study' as a random effect to reflect the non-independence among observations within the same experiment. The linear mixed-effect model was performed using *lme4* package (Bates et al., 2014) and *lmerTest* package (Kuznetsova et al., 2017). The average and median soil denitrification rate, EF_D and $N_2O/(N_2O+N_2)$ ratio from denitrification were calculated. Tukey HSD test was performed across land uses and climatic zones by *agricolae* package. (Mendiburu, 2014). These analyses were conducted in R (RStudio, 2020).

We also used paired observations to perform a meta-analysis to examine the effects of mitigation strategies on denitrification and N_2 emissions using our database. For N application, we used zero or the lowest N as the control and the other N application rates as treatments. For fertilizer type, we considered urea, a widely used fertilizer, as the control and other fertilizer types as treatments. Fertilizer types were subdivided into organic fertilizers (animal manure and solid waste), ammonium-based fertilizers, nitrate-based fertilizers and NH_4NO_3 fertilizer. For management practices, soil amendment (biochar, zeolite and lime) or the use of nitrification inhibitor was compared to N fertilization only. We also considered no tillage, no irrigation, residue removal, single fertilization and surface application of fertilizers as controls, and compared them with their corresponding treatments: tillage, irrigation, residue retention, split fertilization and incorporation of fertilizers.

Effect sizes in meta-analysis were weighed by the number of replicates in a study (Lam et al., 2012) to avoid extreme weights:

$$Weight = (n_c \times n_T)/(n_c + n_T) \quad (3)$$

Where n_c and n_T are the number of replicates of the control and treatment, respectively.

The response ratio r is represented by the natural log and used as a metric for soil denitrification rate and N_2 emissions in response to management practices (Hedges et al., 1999):

$$\ln \quad r = \ln\left(\frac{\overline{x}^T}{\overline{x}^C}\right) \quad (4)$$

where $\overline{x}^T$ and $\overline{x}^C$ are the mean values of the treatment observation and the control observation, respectively. Results of meta-analysis are reported as the percentage change of soil denitrification and N_2 emissions under treatment effects ($(r\text{-}1) \times 100$). Fixed-effect meta-analysis with 4999 iterations bootstrapping was conducted to generate the mean effect size and the 95% confidence intervals (CIs) by MetaWin version 2.1 (Rosenberg et al., 2000). The effects of mitigation strategies were considered significant if the 95% CIs did not overlap with zero.

3. Results

3.1. Summary of soil denitrification and N_2 emissions studies

The summarized data from 648 field observations suggested a mean daily loss of 0.25 kg N ha^{-1} (median value of 0.06 kg N ha^{-1}) through soil denitrification (Fig. 2). Soil denitrification rate was the highest in wetlands with an average of 0.89 kg N ha^{-1} d^{-1} followed by paddy fields (0.62 kg N ha^{-1} d^{-1}), while upland fields had a lower rate of 0.11 kg N ha^{-1} d^{-1}. In terms of climatic zones, the highest denitrification rate with an average of 0.83 kg N ha^{-1} d^{-1} was found in tropical areas while the lowest rate (0.11 kg N ha^{-1} d^{-1}) in continental areas. The overall EF_D was 4.8% (median value of 2.6%) (Fig. 3). Paddy fields had the highest EF_D with an average of 27%, which was significantly higher than grassland and upland fields (4.7% and 4.1%, respectively). Across climatic zones, temperate regions had the highest mean EF_D (5.2%), followed by continental and dry areas (3.2% and 1.9%, respectively).

Overall, the mean $N_2O/(N_2O+N_2)$ ratio was 0.33 (Fig. 4). The $N_2O/(N_2O+N_2)$ ratio across different ecosystems followed the order of grassland (0.37) > wetland (0.36) > upland fields (0.35) > paddy field (0.23) >forest (0.12), and that across climatic regions the order of continental (0.38) > temperate (0.35) > dry (0.25) > tropical (0.20).

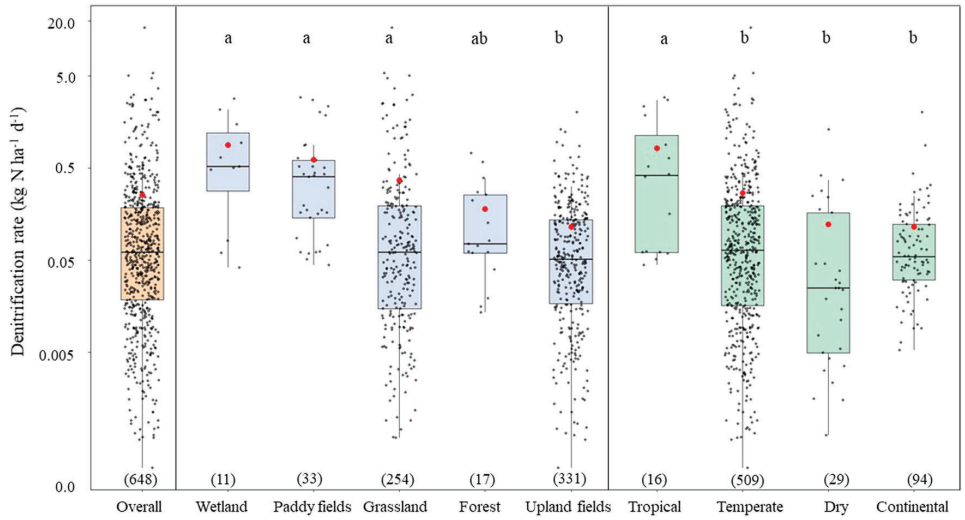


Fig. 2. Denitrification rate across land uses and climatic zones from field experiments. Values in parentheses represent the number of observations. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$.

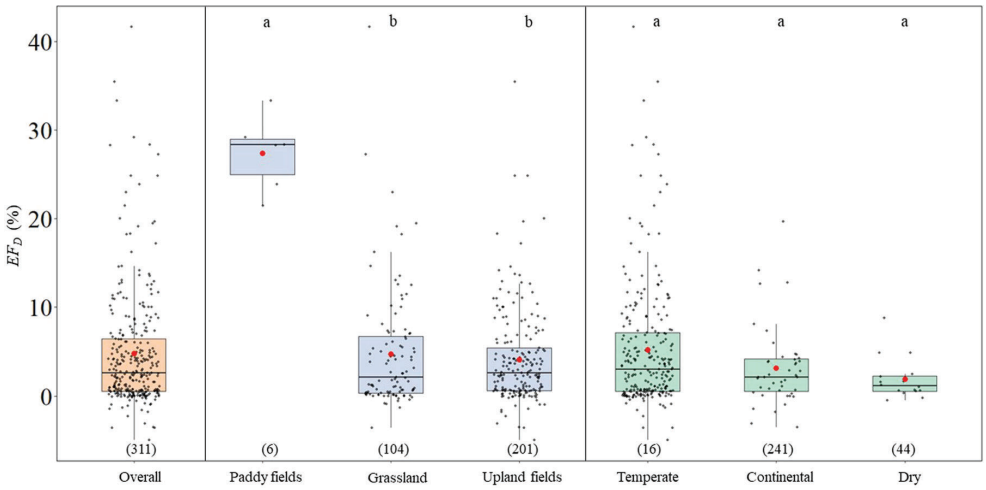


Fig. 3. EF_D across land uses and climatic zones from field experiments. Values in parentheses represent the number of observations. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$.

3.2. Bivariate relationships of soil denitrification rate, N_2 emissions and $N_2O/(N_2O+N_2)$ with soil and environmental properties

Soil denitrification rate was significantly increased with soil WFPS ($p < 0.001$; Fig. 5a) but decreased with increasing O_2 concentration ($p < 0.001$; Fig. 5b). Soil denitrification rate was positively related to soil NO_3^- content ($p < 0.05$; Fig. 5c) and mineral N content ($p < 0.05$; Fig. 5d). The relationship between soil denitrification and soil pH was not significant (Fig. 5e). Soil temperature was positively correlated to soil denitrification rate ($p < 0.001$, Fig. 5f).

Soil N_2 emissions from denitrification significantly increased from low to high latitude ($p < 0.05$; Fig. 6a). Soil mineral N and WFPS were positively related to soil N_2 emissions ($p < 0.05$ and $p < 0.001$, respectively; Fig. 6b, c). N_2 emissions decreased significantly with increased O_2 concentration ($p < 0.05$; Fig. 6d).

The $N_2O/(N_2O+N_2)$ ratio was negatively related to soil pH ($p < 0.05$; Fig. 7a) and WFPS ($p < 0.01$, Fig. 7b). The soil C/N ratio and organic C contents were also negatively related to the $N_2O/(N_2O+N_2)$ ratio ($p < 0.05$; Fig. 7d, f). However, the $N_2O/(N_2O+N_2)$ ratio was positively related to soil O_2 concentration ($p < 0.01$; Fig. 7c).

3.3. The effect of management practices on soil denitrification rate and N_2 production

Increasing N application rate significantly increased soil denitrification rate by 219% (Fig. 8a) and N_2 emissions by 204% (Fig. 8b). Under the same N application rate, compared to urea, other types of fertilizer led to higher soil denitrification rate by 77% (Fig. 8a). Among these fertilizer types, the application of NO_3^--based fertilizer produced the highest increase in soil denitrification rate (199%) (Fig. 8a), whereas

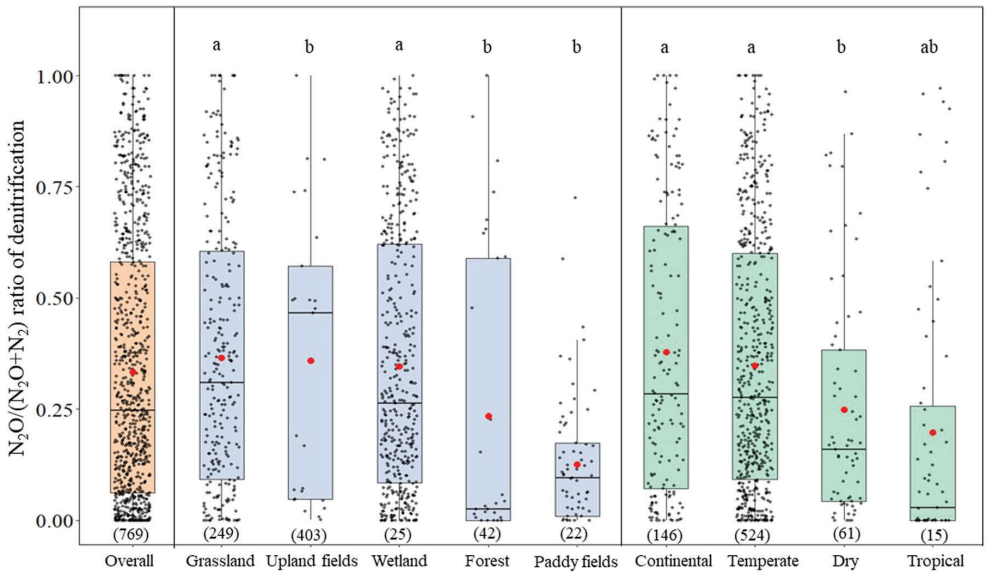


Fig. 4. The $N_2O/(N_2O+N_2)$ ratio of denitrification across climatic zones and land use types. Values in parentheses represent the number of observations. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$.

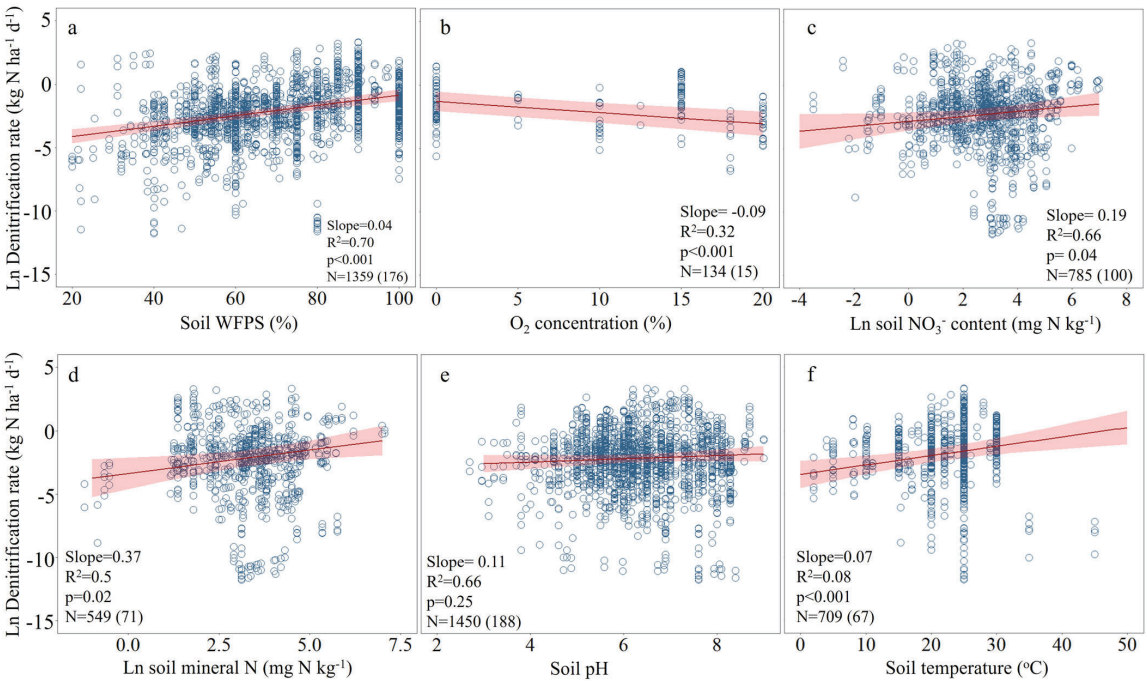


Fig. 5. Relationships between soil denitrification rate and a) soil WFPS, b) soil O_2 concentration, c) soil NO_3^- content, d) soil mineral N, e) soil pH and f) soil temperature. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.

applying NH_4-based fertilizer significantly reduced N_2 emissions by 56% (Fig. 8b). Lime application significantly increased soil denitrification rate and N_2 emissions by 52% and 48%, respectively (Fig. 8a, b), whereas biochar application reduced soil denitrification rate by 38% but had no significant effect on N_2 emissions. The application of zeolite had no significant effects on soil denitrification or N_2 production. Residue retention and irrigation significantly increased soil denitrification rate (144% and 75%, respectively) and N_2 emissions (226% and 102%,

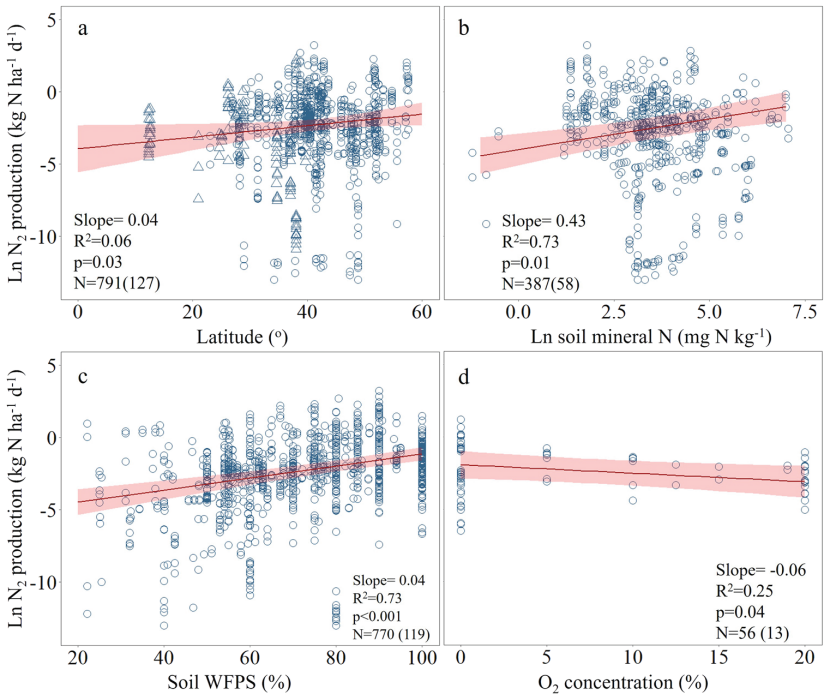


Fig. 6. Relationships between soil N_2 emissions and a) latitude (circles: the Northern Hemisphere; triangles: Southern Hemisphere), b) soil mineral N, c) soil WFPS and d) soil O_2 concentration. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.

respectively) (Fig. 8a, b). Tillage, split application and fertilizer incorporation had no significant effects on soil denitrification rate or N_2 emissions (Fig. 8a, b). Overall, nitrification inhibitor application significantly reduced soil denitrification rate and N_2 emissions by 41% and 30%, respectively (Fig. 8a, b). 3,4-Dimethylpyrazole phosphate (DMPP) was the most effective inhibitor in reducing denitrification rate (71%) and N_2 emissions (68%). Dicyandiamide (DCD) reduced soil denitrification rate but had no significant effect on N_2 emissions. 2-Chloro-6-(trichloromethyl) pyridine (Nitrapyrin) decreased N_2 emissions by 15%.

4. Discussion

4.1. Denitrification is a key N loss pathway globally

Denitrification occurs in a wide range of terrestrial ecosystems. Typically, denitrification requires an anoxic environment with readily available soil NO_3^- and organic C, so fertilized and/or irrigated paddy fields and grasslands located in humid areas had higher denitrification rates (Fig. 2). Moreover, anaerobic microsites within soil aggregates can produce hot-spots and hot moments of denitrification activity, resulting in significant denitrification from apparently well-drained soils (Scheer et al., 2020), such as in the croplands and grasslands in dry climatic areas. The average EF_D calculated in our study was 4.8% and could be up to 41.7% in paddy fields and grasslands in temperate areas (Fig. 3). This indicated that better management strategies should be adopted because N loss via denitrification could decrease NUE and plant productivity.

The $N_2O/(N_2O+N_2)$ ratio varied greatly across land use types and climatic zones with a mean value of 0.33, which implies that N_2 emissions dominated the end-products of denitrification. The $N_2O/(N_2O+N_2)$ ratio in denitrification was used to estimate N_2 emissions in process-based models. These models generally assume the ratio is a fixed value (Hénault et al., 2005) or determined by a function based on soil properties such as soil water condition and soil NO_3^- content (Del Grosso et al., 2000; Stehfest and Muller, 2004; Li et al., 2007). In this regard, the wide range of $N_2O/(N_2O+N_2)$ ratio observed in this study suggests that the estimation of N_2 emissions in process-based models is highly uncertain. In fact, the $N_2O/(N_2O+N_2)$ ratio adopted in process-based models was derived from limited datasets and the relationships between N_2 emission and its key drivers might not have been well captured in these models.

4.2. Key factors of soil denitrification rate, N_2 emissions and $N_2O/(N_2O+N_2)$ ratio

We found that N loss via denitrification could be substantial and varied greatly with edaphic and environmental conditions. These results confirm the traditional conceptual frameworks laid out by Tiedje (1988) and Robertson (1989). Meanwhile, a better understanding of the driving factors of denitrification and its end products is vital for developing effective management strategies that mitigate N_2O and N_2 losses and improve NUE.

Soil NO_3^- is the primary N substrate for soil denitrifying bacteria. We found that soil NO_3^- content was positively related to denitrification rate, which has been widely reported (Zaman et al., 2007; Senbayram et al., 2012). According to our database, when soil NO_3^- content reached a high level (80 mg N kg^{-1}), a higher $N_2O/(N_2O+N_2)$ ratio (>0.5) was observed in highly fertilized soils such as croplands and grasslands. This could be attributed to the suppression of the activity of N_2O reductase, the enzyme responsible for reducing N_2O to N_2 (Senbayram et al., 2019), resulting in incomplete denitrification and higher $N_2O/(N_2O+N_2)$ ratio (Blackmer and Bremner, 1978; Stevens and Laughlin, 1998).

With decreased WFPS, the synthesis of denitrifying enzymes and N_2O reductase is relatively inhibited; thus, denitrification is reduced (Hwang

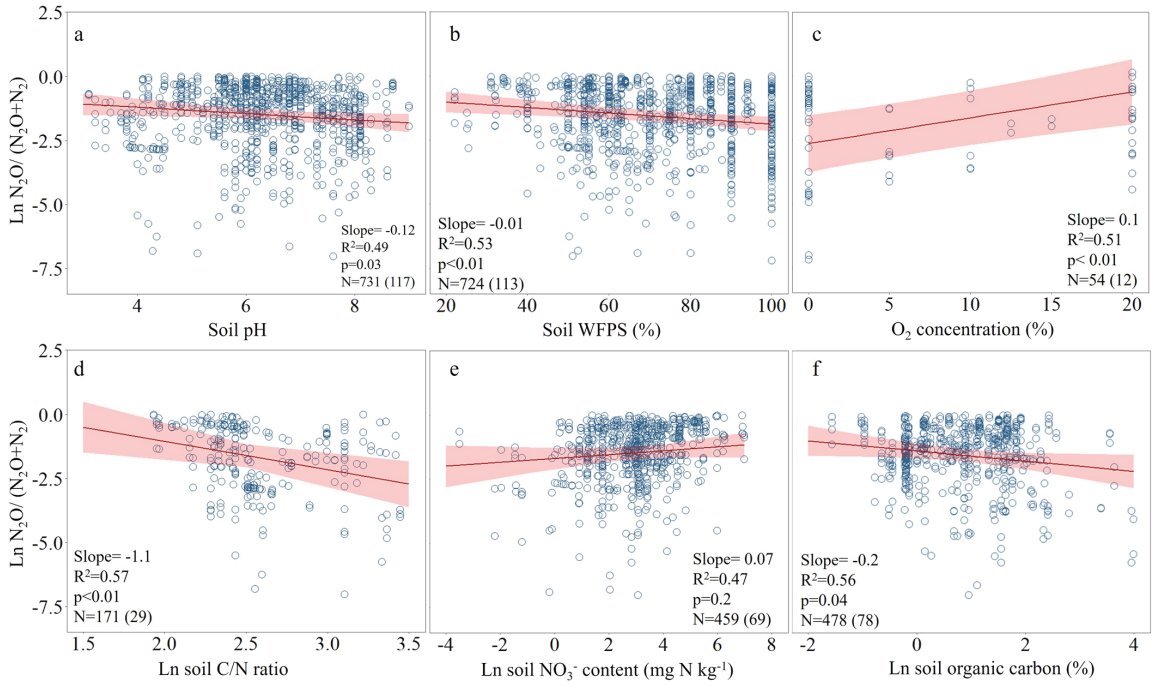


Fig. 7. Relationships between soil $N_2O/(N_2O+N_2)$ ratio and a) soil pH, b) soil WFPS, c) soil O_2 concentration, d) soil C/N ratio, e) soil NO_3^- content and f) soil organic C. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.

and Hanaki, 2000). Our database indicated that when WFPS was over 72%, more N_2 than N_2O was emitted through denitrification, thereby decreasing $N_2O/(N_2O+N_2)$ ratio. The activity of N_2O reductase was decreased with increased soil O_2 concentration (Friedl et al., 2016). At high soil water content, the limited O_2 availability in the soil resulted in anaerobic conditions. Subsequently, N_2O reductase capacity was upregulated, leading to a lower $N_2O/(N_2O+N_2)$ ratio (Fig. 7c).

We found that denitrification occurred over a wide range of soil temperatures (2–45 °C) and was positively related to soil temperature (Fig. 5f). Soil respiration increased with soil temperature, resulting in O_2 depletion and anaerobic conditions. This promoted the activity of denitrifiers and denitrification (Lai and Denton, 2018).

Soil pH regulates denitrification and its end products (Dannenmann et al., 2008; Sun et al., 2012). N_2O reductase was less active under low pH (Liu et al., 2010). At a reduced soil pH, denitrification may liberate more N_2O and subsequently increase the ratio of $N_2O/(N_2O+N_2)$ (Simek and Cooper, 2002; Cuhel et al., 2010; Qu et al., 2014). However, the relationship between denitrification and soil pH was not significant in this study (Fig. 5e). This might be attributed to the confounding effects of other factors such as the application of N fertilizers and organic amendments under aerobic conditions in upland fields and grasslands. Under anaerobic conditions (paddy fields and wetlands), soil pH was positively correlated with denitrification (Fig. S4).

We found that for a soil with a higher C/N ratio (>14), more N_2O was reduced to N_2, decreasing the $N_2O/(N_2O+N_2)$ ratio from denitrification (Fig. 7d). This is because higher soil C/N ratio promoted denitrifying population and the diffusion rate of soil NO_3^- into denitrifying microsites (Saggar et al., 2013). Moreover, soil organic C was highly related to the ratio of N_2O to total denitrification (Fig. 7f). More available C could promote the growth of soil microorganisms and increase N_2O reductase in the soil, which could reduce the $N_2O/(N_2O+N_2)$ ratio from denitrification. However, the effect of soil organic C on $N_2O/(N_2O+N_2)$ can be outweighed by other factors, such as soil NO_3^- content (Weier et al., 1993).

4.3. Management practices to mitigate soil denitrification and N_2 emissions

Our meta-analysis showed that optimizing N application rate to match plant N demand and avoiding excessive NO_3^- in the soil can effectively reduce denitrification and N_2 emissions (Fig. 8, Fig. S1d). The type of N fertilizers can also impact denitrification and N_2 emissions. Compared to urea, NO_3^- based fertilizers and organic fertilizers significantly increased denitrification by 77% (Fig. 8) under the same application rate. Nitrate-based fertilizers provide more NO_3^- compared to urea, which is the key substrate for denitrification. We found that soil NO_3^- content was significantly correlated with soil denitrification rate and N_2 emissions (Fig. 5c). Our analysis also indicated that the application of urea/ammonium-based fertilizers could reduce N_2 emissions compared to nitrate-based or organic fertilizers. When nitrate-based fertilizers are applied, more NO_3^- is available for denitrification and subsequent gaseous losses (Wang et al., 2018). Organic fertilizers like livestock manure, increased denitrification by providing the energy source (readily available C) for denitrifiers and aggravate anaerobic conditions through its decomposition (Fig. 9) (Estavillo et al., 1994; Mogge et al., 1998; Barton et al., 1999).

Managing crop residue properly is important to regulate denitrification and N_2 emissions without impairing soil fertility and crop yields. Our results showed that residue retention could increase denitrification and N_2 emissions by 144% and 226%, respectively (Fig. 8), because it can increase C availability for denitrifiers and promote subsequent denitrification (Garcia-Montiel et al., 2003). In addition, crop residue decomposition consumes O_2 thereby inducing higher denitrification and N_2 emissions (Clough et al., 1998; Gillam et al., 2008) (Fig. 9). It is worth

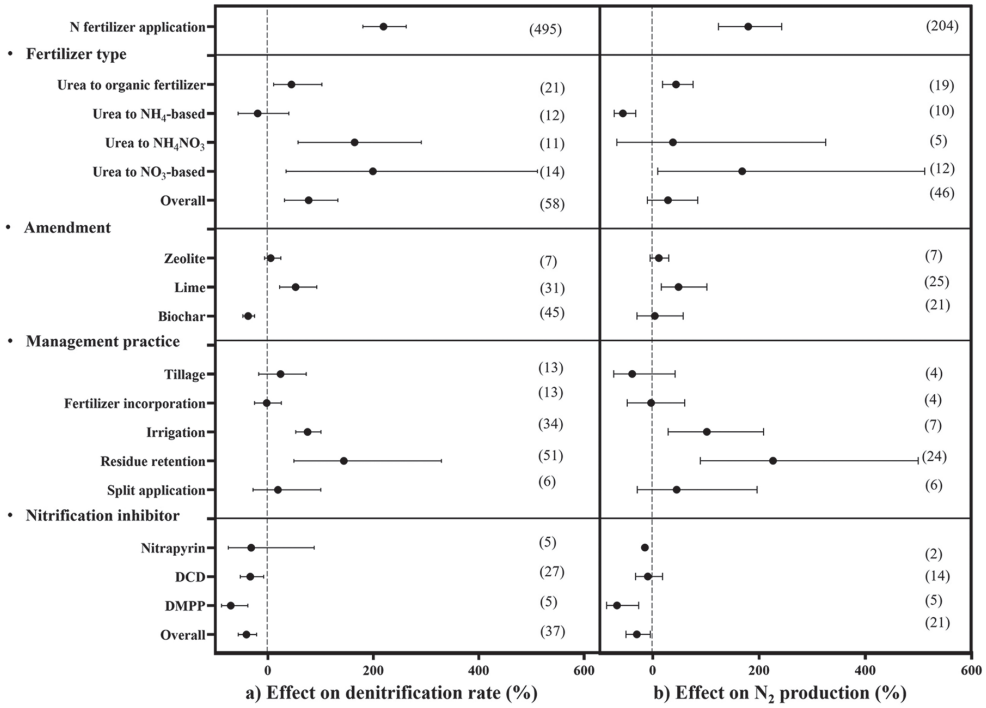


Fig. 8. Effect of N application, fertilizer type, amendment, management practice and nitrification inhibitor on a) denitrification rate and b) N_2 production. Means and 95% confidence intervals are depicted. Numbers of experimental observations are in parentheses. DCD: dicyandiamide, DMPP: 3,4-Dimethylpyrazole phosphate, Nitrapyrin: 2-chloro-6-(trichloromethyl) pyridine.

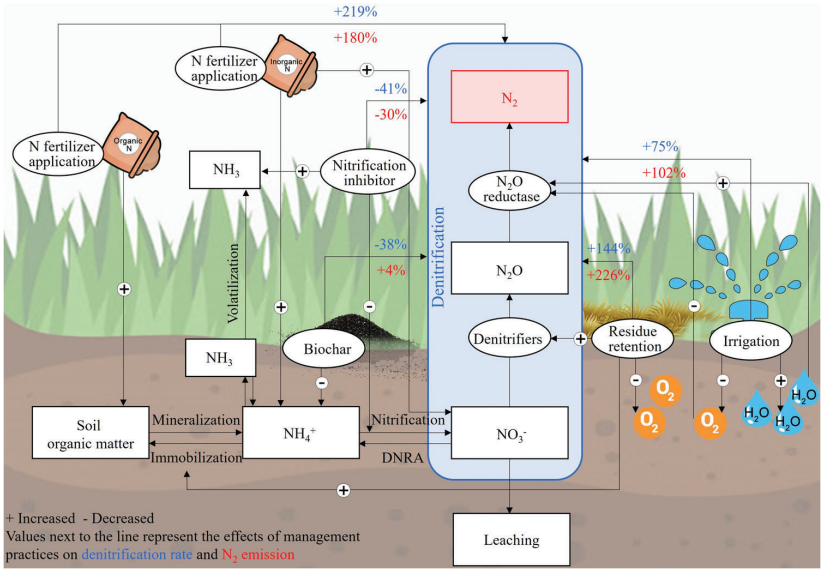


Fig. 9. The effects of management practices on soil denitrification rate, N_2 emission and associated soil N transformations. Values presented were obtained from the meta-analysis in this study. '+ ' and '–'denote increased and decreased responses to management practices. DNRA: dissimilatory nitrate reduction to ammonium.

nothing that the effect of residue retention on denitrification and N_2 emissions is highly related to residue quality. Incorporating crop residues with low C/N ratios would promote denitrification by providing sufficient N for soil microbial growth and proliferation (Xia et al., 2018). However, the application of crop residue with higher C/N ratios (e.g., wheat straw and switchgrass) could reduce denitrification by promoting

soil microbial N immobilization (Sun et al., 2018).

Irrigation could temporarily increase soil water content and develop an anaerobic zone that favors denitrification especially for fine-textured soils (De Klein and Van Logtestijn, 1996; Zaman et al., 2012; Saggar et al., 2013) (Fig. 9). In this regard, advanced irrigation techniques (e.g., drip irrigation) should be adopted to reduce denitrification and associated N_2 emissions (Barakat et al., 2016).

Biochar application effectively reduced soil denitrification rate by 38% (Fig. 8a), because it promoted the chemical affinity of inorganic N substrates (Cayuela et al., 2013; Chintala et al., 2015). However, N_2 emissions overall were not significantly affected by biochar application (Fig. 8b), likely because biochars have diverse characteristics e.g., pH and C/N ratio, and may exert contrasting effects on N_2 emissions (Malghani et al., 2018). Lime is often applied in pastures and wetlands to decrease total N_2O emissions (Saggar et al., 2013) but may increase soil denitrification rate and N_2 emissions (Simek and Cooper, 2002). Liming can also increase soil pH and subsequently enhance the activity of N_2O reductase, which reduces N_2O to N_2 in acidic soils.

Nitrification inhibitors delay the conversion of NH_4^+ to NO_3^- via nitrification (Zaman et al., 2012) (Fig. 9). This delay maintains the applied N fertilizer/animal manure in the form of NH_4^+ limiting NO_3^- availability for denitrification and decreases N_2O and N_2 emissions (McGeough et al., 2012; Maris et al., 2015; Friedl et al., 2017). However, nitrification inhibitors may increase N loss via NH_3 volatilization (Pan et al., 2016; Lam et al., 2017, 2018; Xia et al., 2017). Therefore, additional knowledge-based N managements (e.g., fertilizer deep placement) should be adopted simultaneously with nitrification inhibitors to reduce the overall N losses (Xia et al., 2020).

Many studies have focused on mitigation strategies to reduce potent greenhouse gas N_2O emissions and how to facilitate N_2O reduction to N_2. However, from the perspective of N use efficiency, N loss via N_2 should be minimized. Management practices that can reduce both denitrification and N_2 emissions should be recommended considering the trade-off between N_2O mitigation and N use efficiency improvement. Our meta-analysis suggests that reduced N application rate, use of ammonium-based fertilizers, reduced residue retention, biochar amendment and nitrification inhibitors, and improved irrigation techniques such as drip irrigation could effectively reduce denitrification and N_2 emissions.

5. Uncertainty

We are aware that the number of observations included in the database varies largely across land uses; more relevant studies should be conducted in different ecosystems. Moreover, the acetylene inhibition technique (AIT) has been widely used but may underestimate denitrification rate (Wang and Yan, 2016) under both field and laboratory conditions (Fig. S5) mainly owing to the inhibition of nitrification (Groffman et al., 2006; Qin et al., 2013). Previous studies demonstrated that AIT may underestimate denitrification by 4–10 times compared to ^{15}N tracer method or gas-flow-soil-core techniques when applied to the same soils (Watts and Seitzinger, 2000; Lindau et al., 2011; Sgouridis et al., 2016). Although ^{15}N tracer method or gas flow core techniques could reliably estimate denitrification rate (Groffman et al., 2006), they are not widely adopted, as evidenced by the number of observations in our database (^{15}N-gas flux method: n = 268; gas-flow (helium or Ar) method: n = 131; AIT: n = 1200). Such bias in the observation number among different methods might have caused uncertainties in quantifying soil denitrification rate in this study. Besides, some studies that used Nitrogen-Free Air Recirculation Method (N-FARM) to measure the denitrification rate (Burgin and Groffman, 2012) may overestimate the N_2O from denitrification because it cannot distinguish the N_2O produced from denitrification and that from nitrification. Therefore, more studies using advanced techniques should be conducted. We are also aware that NO emissions are a product of denitrification and some studies applied stable isotope models (Houlton et al., 2006) to estimate NO emission from denitrification. We only identified N_2O and N_2 emissions from denitrification owing to the limited studies on $NO/N_2O/N_2$ ratio.

6. Conclusions

We compiled a global database on denitrification rate and its N_2O and N_2 emissions. We found that N loss through denitrification varied greatly across land uses and climatic regions. The average EF_D was 4.8%. The wide range of $N_2O/(N_2O+N_2)$ demonstrated that the adoption of a fixed ratio in some process-based models for estimating N_2 emissions from denitrification is not suitable. We found that N_2 loss accounted for 67% of total denitrification. N loss as N_2, although harmless to the environment, deserves more attention from the perspective of improving N use efficiency. Denitrification and its N_2O and N_2 emissions are highly dependent on soil properties, e.g., soil mineral N, soil water, temperature and oxygen conditions. We further examined the potential of mitigation strategies in decreasing denitrification and N_2 emissions. Specifically, optimized N input, the use of ammonium-based fertilizers, biochar amendment, use of nitrification inhibitors, and advanced irrigation technique could effectively decrease denitrification and N_2 emissions.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.agee.2021.107850.

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Supporting Information for

A global synthesis of soil denitrification: driving factors and mitigation strategies

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1. Figure S1. Relationships between soil denitrification rate and a) latitude (circles: the Northern Hemisphere; triangles: Southern Hemisphere.), b) SOC, c) soil C/N ratio and d) N application rate. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.
2. Figure S2. Relationships between soil N_2 production and a) soil NO_3^- content, b) soil pH, c) soil C/N ratio, d) soil organic carbon e) N application rate and f) soil temperature. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.

3. Figure S3. Relationships between soil N_2O /(N_2O+N_2) ratio and a) soil mineral N, b) latitude (circles: the Northern Hemisphere; triangles: Southern Hemisphere) and c) N application rate and d) soil temperature. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.
4. Figure S4. Relationships between soil denitrification rate and soil pH in different land use types. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.
5. Figure S5. Measurement of denitrification rate between AIT and ^{15}N-gas flux method. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$.
6. Reference list of included studies.

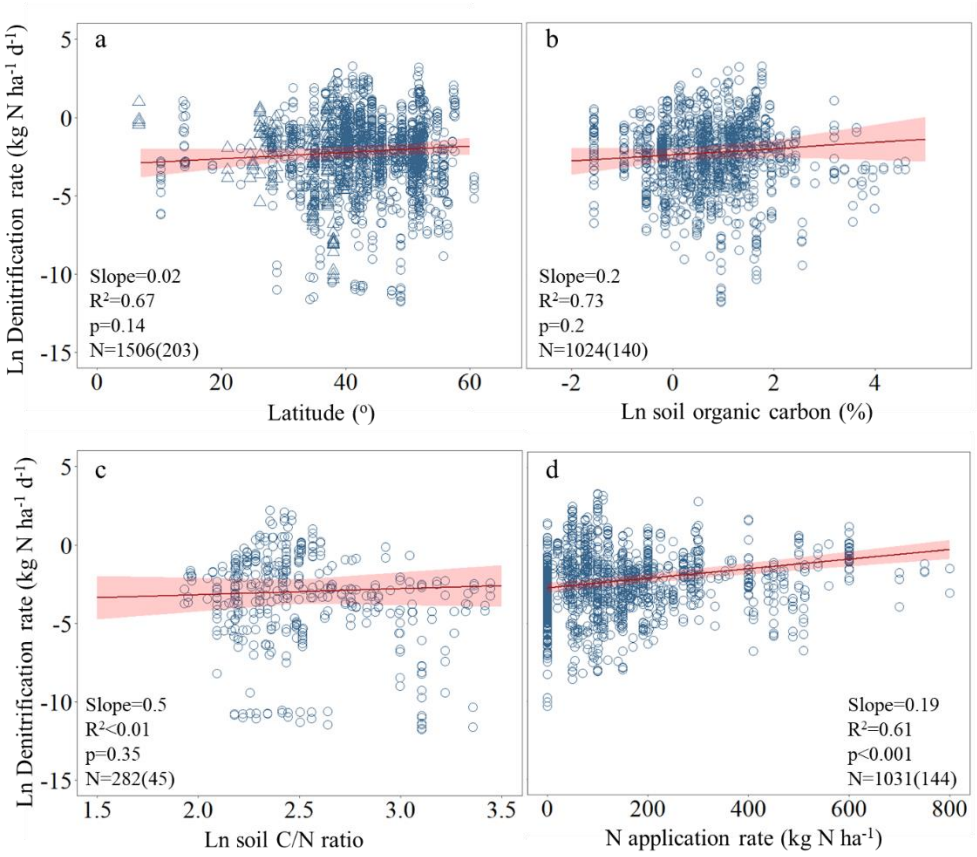


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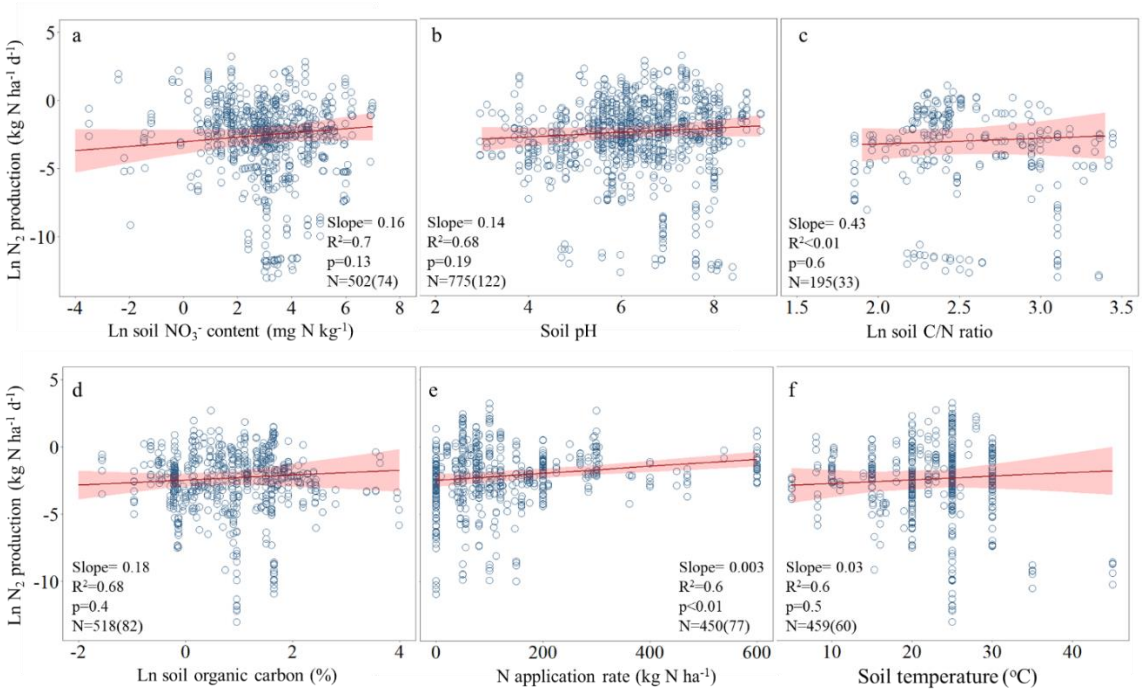


Figure S2. Relationships between soil N_2 production and a) soil NO_3^- content, b) soil pH, c) soil C/N ratio, d) soil organic carbon e) N application rate and f) soil temperature. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.

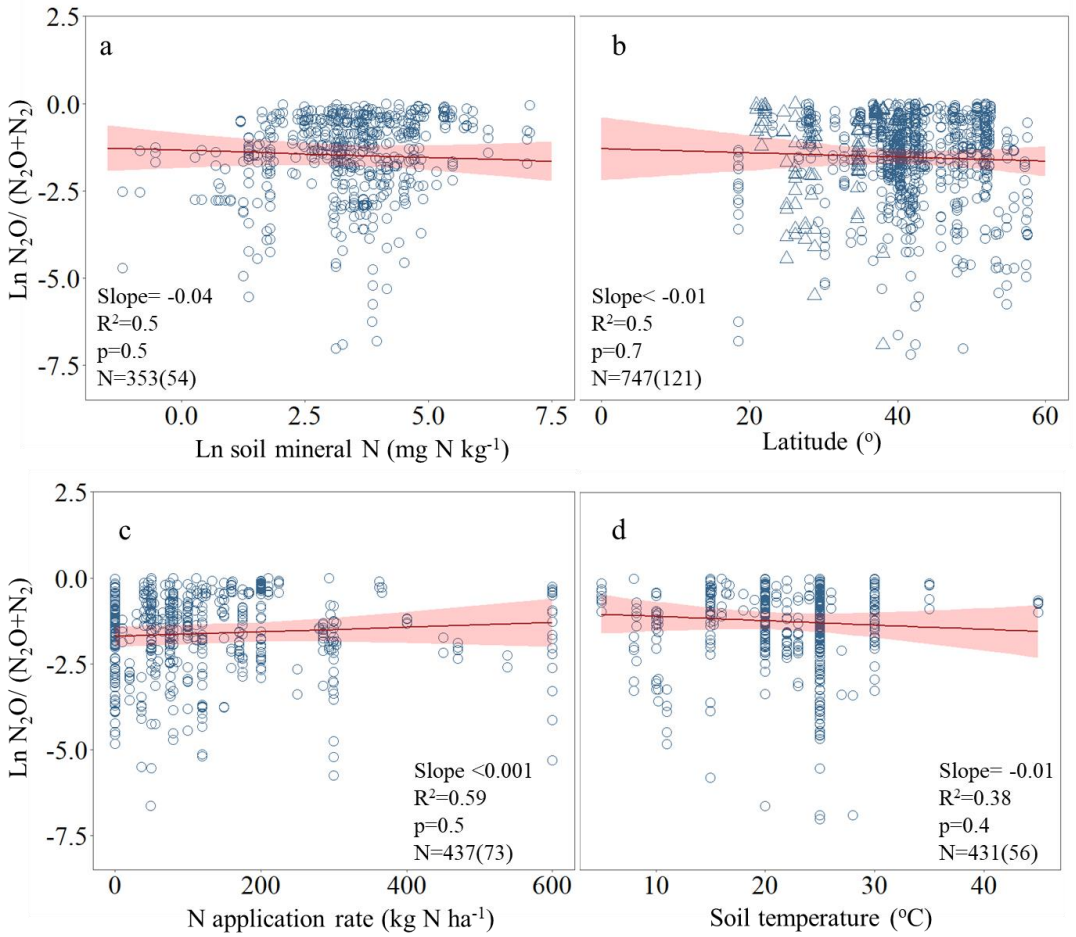


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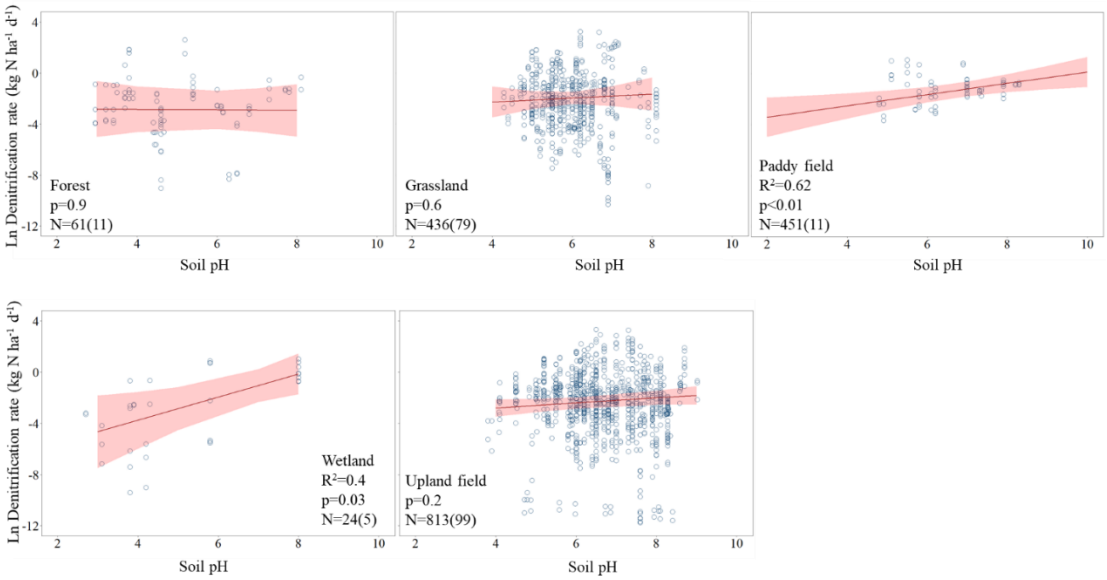


Figure S4. Relationships between soil denitrification rate and soil pH in different land use types. The red lines indicate the slopes from the linear mixed effects models and the red shadings represent the 95% confidence intervals. N in the figure represents the number of observations and the values in parentheses the number of studies.

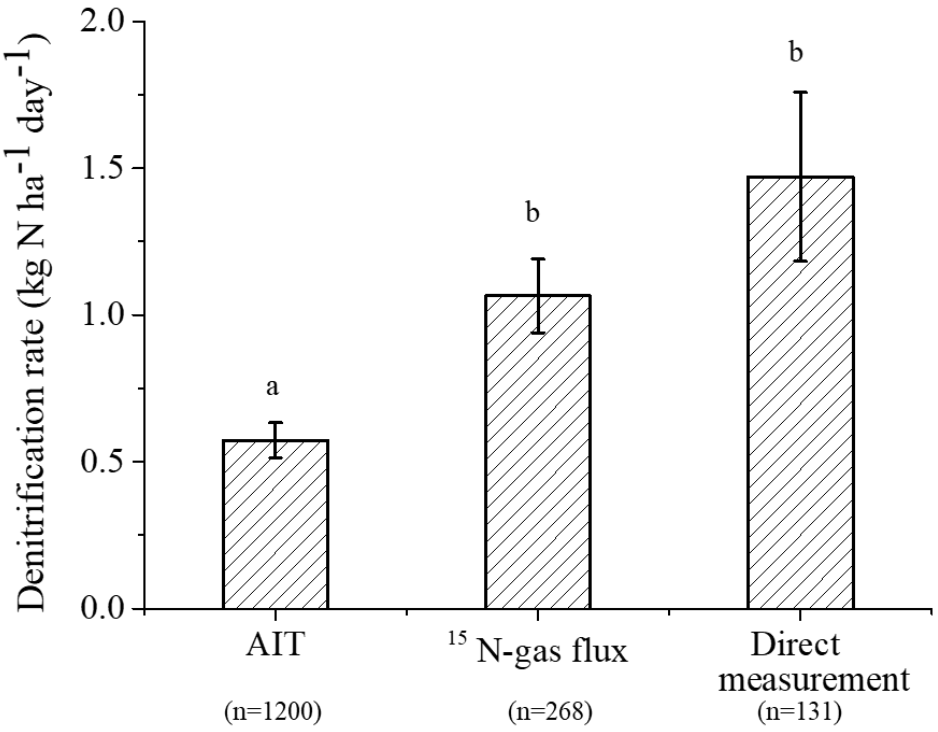


Figure S5. Measurement of denitrification rate between AIT, ^{15}N-gas flux and direct measurement (gas-flow helium/Ar techniques) method. Different letters on the top indicate that the mean values are significantly different at $p < 0.05$.

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Supporting Table S1 Studies used in the meta-analysis to examine the effects of mitigation strategies on soil denitrification rate

study	replication	replication	weight	variance	fertilizer type	amnedment/inhibitor		denitrification rate	management practice	lnRR
	control	treatment			treatment	treatment	CK	treatment	category	
Arah et al 1991	2	2	1	1			0.132	0.397	N_fertilizer_application	1.104
Arah et al 1991	2	2	1	1			0.05	0.044	N_fertilizer_application	-0.129
Arah et al 1991	2	2	1	1			0.05	0.023	N_fertilizer_application	-0.759
Arah et al 1991	2	2	1	1			0.05	0.089	N_fertilizer_application	0.577
Arah et al 1991	2	2	1	1			0.009	0.015	N_fertilizer_application	0.493
Arah et al 1991	2	2	1	1			0.008	0.015	N_fertilizer_application	0.632
Arah et al 1991	2	2	1	1			0.018	0.047	N_fertilizer_application	0.939
Arah et al 1991	2	2	1	1			0.023	0.046	N_fertilizer_application	0.708
Arcara et al 1999	3	3	1.5	0.667			0.007	0.016	N_fertilizer_application	0.781
Arcara et al 1999	3	3	1.5	0.667			0.007	0.026	N_fertilizer_application	1.254
Arcara et al 1999	3	3	1.5	0.667			0.007	0.015	N_fertilizer_application	0.724
Aulakh et al 1984	2	2	1	1			0.035	0.037	residue_retention	0.061
Aulakh et al 1984	2	2	1	1			0.035	0.047	N_fertilizer_application	0.291
Aulakh et al 1984	2	2	1	1			0.047	0.08	residue_retention	0.527
Aulakh et al 1984	2	2	1	1			0.08	0.108	residue_retention	0.306
Aulakh et al 1984	2	2	1	1			0.08	0.131	N_fertilizer_application	0.494
Aulakh et al 1984	2	2	1	1			0.131	0.247	residue_retention	0.639
Aulakh et al 2001	3	3	1.5	0.667			0.144	0.501	N_fertilizer_application	1.248
Aulakh et al 2001	3	3	1.5	0.667			0.144	0.603	N_fertilizer_application	1.433
Aulakh et al 2001	3	3	1.5	0.667			0.144	0.427	N_fertilizer_application	1.088
Aulakh et al 2001	3	3	1.5	0.667			0.144	0.196	residue_retention	0.308
Aulakh et al 2001	3	3	1.5	0.667			0.501	0.516	residue_retention	0.03
Aulakh et al 2001	3	3	1.5	0.667			0.427	0.44	residue_retention	0.031
Aulakh et al 2001	3	3	1.5	0.667			0.024	0.028	N_fertilizer_application	0.161
Aulakh et al 2001	3	3	1.5	0.667			0.024	0.028	N_fertilizer_application	0.161
Aulakh et al 2001	3	3	1.5	0.667			0.024	0.039	residue_retention	0.486
Burton et al 2008	4	4	2	0.5			0.005	0.026	N_fertilizer_application	1.604
Burton et al 2008	4	4	2	0.5			0.029	0.036	N_fertilizer_application	0.21
Cao et al 2006	3	3	1.5	0.667			0.213	0.378	N_fertilizer_application	0.575
Cao et al 2006	3	3	1.5	0.667			0.213	0.431	N_fertilizer_application	0.706
Cao et al 2006	3	3	1.5	0.667			0.213	0.492	N_fertilizer_application	0.839
Ciarlo et al 2008	3	3	1.5	0.667			0.001	0.001	N_fertilizer_application	0.223
Ciarlo et al 2008	3	3	1.5	0.667			0.001	0.002	N_fertilizer_application	0.56
Ciarlo et al 2008	3	3	1.5	0.667			0.001	0.004	split_application	1.184
Ciarlo et al 2008	3	3	1.5	0.667			0.002	0.004	split_application	0.751
Ciarlo et al 2008	3	3	1.5	0.667			0.051	0.382	N_fertilizer_application	2.008
Ciarlo et al 2008	3	3	1.5	0.667			0.051	0.17	N_fertilizer_application	1.201
Ciarlo et al 2008	3	3	1.5	0.667			0.382	0.17	split_application	-0.808
Ciarlo et al 2008	3	3	1.5	0.667			0.063	0.074	split_application	0.169
Ciarlo et al 2008	3	3	1.5	0.667			0.054	0.789	N_fertilizer_application	2.685
Ciarlo et al 2008	3	3	1.5	0.667			0.054	0.582	N_fertilizer_application	2.381
Ciarlo et al 2008	3	3	1.5	0.667			0.789	0.582	split_application	-0.304
Ciarlo et al 2008	3	3	1.5	0.667			0.056	0.058	split_application	0.044
Ciarlo et al 2008	3	3	1.5	0.667	NO3-based		0.002	0.001	fertilizer	-0.336
Ciarlo et al 2008	3	3	1.5	0.667	NO3-based		0.004	0.004	fertilizer	0.096
Ciarlo et al 2008	3	3	1.5	0.667	NO3-based		0.063	0.382	fertilizer	1.805
Ciarlo et al 2008	3	3	1.5	0.667	NO3-based		0.074	0.17	fertilizer	0.828

Ciarlo et al 2008	3	3	1.5	0.667	NO3-based	0.056	0.789	fertilizer	2.654
Ciarlo et al 2008	3	3	1.5	0.667	NO3-based	0.058	0.582	fertilizer	2.305
De Vries et al 2011	6	6	3	0.333		0.121	0.311	N_fertilizer_application	0.94
De Vries et al 2011	6	6	3	0.333		0.068	0.218	N_fertilizer_application	1.166
Duxbury and McConnaug	3	3	1.5	0.667		0.019	0.024	N_fertilizer_application	0.223
Duxbury and McConnaug	3	3	1.5	0.667		0.019	0.028	N_fertilizer_application	0.405
Duxbury and McConnaug	3	3	1.5	0.667	NO3-based	0.028	0.024	fertilizer	-0.182
Blackmer and Bremner 1	3	3	1.5	0.667		0.222	0.778	N_fertilizer_application	1.253
Blackmer and Bremner 1	3	3	1.5	0.667		0.222	1.333	N_fertilizer_application	1.792
Blackmer and Bremner 1	3	3	1.5	0.667		0.222	1.444	N_fertilizer_application	1.872
Blackmer and Bremner 1	3	3	1.5	0.667		0.222	1.456	N_fertilizer_application	1.879
Blackmer and Bremner 1	3	3	1.5	0.667		0.222	1.422	N_fertilizer_application	1.856
Blackmer and Bremner 1	3	3	1.5	0.667		0.078	0.622	N_fertilizer_application	2.079
Blackmer and Bremner 1	3	3	1.5	0.667		0.078	0.822	N_fertilizer_application	2.358
Blackmer and Bremner 1	3	3	1.5	0.667		0.078	0.844	N_fertilizer_application	2.385
Blackmer and Bremner 1	3	3	1.5	0.667		0.078	0.789	N_fertilizer_application	2.317
Blackmer and Bremner 1	3	3	1.5	0.667		0.078	0.722	N_fertilizer_application	2.228
Buresh et al 1991	3	3	1.5	0.667		2.741	0.887	fertilzer_incorporation	-1.128
Buresh et al 1991	3	3	1.5	0.667		2.915	1.871	fertilzer_incorporation	-0.443
Fangueiro et al 2008	5	5	2.5	0.4		0.003	0.007	N_fertilizer_application	0.972
Fangueiro et al 2008	5	5	2.5	0.4		0.003	0.007	N_fertilizer_application	0.918
Fangueiro et al 2008	5	5	2.5	0.4		0.003	0.006	N_fertilizer_application	0.905
Fangueiro et al 2008	5	5	2.5	0.4		0.003	0.008	N_fertilizer_application	1.072
Fangueiro et al 2008	5	5	2.5	0.4		0.003	0.005	N_fertilizer_application	0.653
Lai and Denton 2018	4	4	2	0.5		0	0	N_fertilizer_application	1.737
Lai and Denton 2018	4	4	2	0.5		0	0	N_fertilizer_application	1.744
Lai and Denton 2018	4	4	2	0.5		0	0	N_fertilizer_application	1.929
Lai and Denton 2018	4	4	2	0.5		0	0	N_fertilizer_application	1.738
Lai and Denton 2018	4	4	2	0.5		0	0	N_fertilizer_application	2.233
Lai and Denton 2018	4	4	2	0.5		0	0	N_fertilizer_application	2.343
Lai and Denton 2018	4	4	2	0.5		0	0	N_fertilizer_application	2.262
Lai and Denton 2018	4	4	2	0.5		0	0.001	N_fertilizer_application	2.692
Lai and Denton 2018	4	4	2	0.5		0	0.001	N_fertilizer_application	3.259
Loick et al 2017	4	4	2	0.5		0.38	2.23	N_fertilizer_application	1.77
Loick et al 2017	4	4	2	0.5		0.38	1.771	N_fertilizer_application	1.539
Loick et al 2017	12	12	6	0.167		0.08	0.73	N_fertilizer_application	2.211
Loick et al 2017	12	12	6	0.167		0.08	2.09	N_fertilizer_application	3.263
Qin SP et al 2017 temper	4	4	2	0.5		0.246	0.419	N_fertilizer_application	0.534
Senbayram, et al 2019	3	3	1.5	0.667		0.12	0.431	N_fertilizer_application	1.281
Senbayram, et al 2019	3	3	1.5	0.667		0.099	0.434	N_fertilizer_application	1.477
Senbayram, et al 2019	3	3	1.5	0.667		0.103	0.174	N_fertilizer_application	0.527
Senbayram, et al 2019	3	3	1.5	0.667		0.103	0.036	N_fertilizer_application	-1.04
Senbayram, et al 2019	3	3	1.5	0.667		0.024	0.123	N_fertilizer_application	1.616
Senbayram, et al 2019	3	3	1.5	0.667		0.024	0.037	N_fertilizer_application	0.411
Senbayram, et al 2018	3	3	1.5	0.667		0.175	0.289	N_fertilizer_application	0.502
Senbayram, et al 2018	3	3	1.5	0.667		0.289	0.367	residue_retention	0.238
Senbayram, et al 2018	3	3	1.5	0.667		0.289	0.348	residue_retention	0.186
Senbayram, et al 2018	3	3	1.5	0.667		0.289	0.322	residue_retention	0.106
Wallenstein et al 2006	3	3	1.5	0.667		0.023	0.02	N_fertilizer_application	-0.145
Wallenstein et al 2006	3	3	1.5	0.667		0.397	0.417	N_fertilizer_application	0.05

Wallenstein et al 2006	3	3	1.5	0.667		0.025	0.019	N_fertilizer_application	-0.283
Wu D et al 2018	3	3	1.5	0.667		2.555	2.776	N_fertilizer_application	0.083
Wu D et al 2018	3	3	1.5	0.667		2.776	6.634	residue_retention	0.871
Cai G et al 2002	4	4	2	0.5		0.643	0.304	fertilzer_incorporation	-0.75
Cai G et al 2002	4	4	2	0.5		0.268	0.643	N_fertilizer_application	0.875
Cai G et al 2002	4	4	2	0.5		0.427	0.409	irrigation	-0.043
Cai G et al 2002	4	4	2	0.5		0.355	0.427	N_fertilizer_application	0.187
Carter et al 2007	4	4	2	0.5		0.015	0.099	N_fertilizer_application	1.894
Chintala et al 2014	3	3	1.5	0.667	biochar	0.209	0.1	amendment	-0.731
Chintala et al 2014	3	3	1.5	0.667	biochar	0.209	0.085	amendment	-0.898
Chintala et al 2014	3	3	1.5	0.667	biochar	0.209	0.1	amendment	-0.731
Chintala et al 2014	3	3	1.5	0.667	biochar	0.209	0.088	amendment	-0.868
Chintala et al 2014	3	3	1.5	0.667		0.209	0.209	residue_retention	0
Chintala et al 2014	3	3	1.5	0.667		0.209	0.191	residue_retention	-0.09
Chintala et al 2014	3	3	1.5	0.667		0.209	0.206	residue_retention	-0.012
Chintala et al 2014	3	3	1.5	0.667	biochar	0.791	0.507	amendment	-0.444
Chintala et al 2014	3	3	1.5	0.667	biochar	0.791	0.384	amendment	-0.723
Chintala et al 2014	3	3	1.5	0.667	biochar	0.791	0.399	amendment	-0.683
Chintala et al 2014	3	3	1.5	0.667	biochar	0.791	0.373	amendment	-0.75
Chintala et al 2014	3	3	1.5	0.667		0.791	0.662	residue_retention	-0.178
Chintala et al 2014	3	3	1.5	0.667		0.791	0.561	residue_retention	-0.342
Chintala et al 2014	3	3	1.5	0.667		0.791	0.718	residue_retention	-0.096
Chintala et al 2014	3	3	1.5	0.667	biochar	2.271	1.004	amendment	-0.816
Chintala et al 2014	3	3	1.5	0.667	biochar	2.271	0.891	amendment	-0.936
Chintala et al 2014	3	3	1.5	0.667	biochar	2.271	1.087	amendment	-0.737
Chintala et al 2014	3	3	1.5	0.667	biochar	2.271	0.85	amendment	-0.983
Chintala et al 2014	3	3	1.5	0.667		2.271	2.529	residue_retention	0.107
Chintala et al 2014	3	3	1.5	0.667		2.271	2.382	residue_retention	0.048
Chintala et al 2014	3	3	1.5	0.667		2.271	2.498	residue_retention	0.095
Chintala et al 2014	3	3	1.5	0.667	biochar	0.247	0.197	amendment	-0.229
Chintala et al 2014	3	3	1.5	0.667	biochar	0.247	0.197	amendment	-0.229
Chintala et al 2014	3	3	1.5	0.667	biochar	0.247	0.168	amendment	-0.389
Chintala et al 2014	3	3	1.5	0.667	biochar	0.247	0.173	amendment	-0.358
Chintala et al 2014	3	3	1.5	0.667		0.247	0.223	residue_retention	-0.102
Chintala et al 2014	3	3	1.5	0.667		0.247	0.202	residue_retention	-0.202
Chintala et al 2014	3	3	1.5	0.667		0.247	0.234	residue_retention	-0.055
Chintala et al 2014	3	3	1.5	0.667	biochar	0.899	0.622	amendment	-0.368
Chintala et al 2014	3	3	1.5	0.667	biochar	0.899	0.449	amendment	-0.693
Chintala et al 2014	3	3	1.5	0.667	biochar	0.899	0.463	amendment	-0.664
Chintala et al 2014	3	3	1.5	0.667	biochar	0.899	0.505	amendment	-0.576
Chintala et al 2014	3	3	1.5	0.667		0.899	0.827	residue_retention	-0.083
Chintala et al 2014	3	3	1.5	0.667		0.899	0.88	residue_retention	-0.021
Chintala et al 2014	3	3	1.5	0.667		0.899	0.912	residue_retention	0.015
Chintala et al 2014	3	3	1.5	0.667	biochar	2.646	2.042	amendment	-0.259
Chintala et al 2014	3	3	1.5	0.667	biochar	2.646	1.678	amendment	-0.455
Chintala et al 2014	3	3	1.5	0.667	biochar	2.646	1.955	amendment	-0.303
Chintala et al 2014	3	3	1.5	0.667	biochar	2.646	1.569	amendment	-0.523
Chintala et al 2014	3	3	1.5	0.667		2.646	2.816	residue_retention	0.062
Chintala et al 2014	3	3	1.5	0.667		2.646	2.848	residue_retention	0.074
Chintala et al 2014	3	3	1.5	0.667		2.646	2.755	residue_retention	0.04

Cayuela et al 2013	4	4	2	0.5	biochar	3.188	2.956	amendment	-0.076
Cayuela et al 2013	4	4	2	0.5	biochar	0.021	0.036	amendment	0.522
Cayuela et al 2013	4	4	2	0.5	biochar	1.256	0.616	amendment	-0.712
Cayuela et al 2013	4	4	2	0.5	biochar	0.088	0.221	amendment	0.925
Cayuela et al 2013	4	4	2	0.5	biochar	0.288	0.144	amendment	-0.695
Cayuela et al 2013	4	4	2	0.5	biochar	0.492	0.315	amendment	-0.446
Cayuela et al 2013	4	4	2	0.5	biochar	0.637	0.184	amendment	-1.241
Cayuela et al 2013	4	4	2	0.5	biochar	0.506	0.115	amendment	-1.477
Cayuela et al 2013	4	4	2	0.5	biochar	0.015	0.024	amendment	0.468
Cayuela et al 2013	4	4	2	0.5	biochar	0.436	0.342	amendment	-0.242
Cayuela et al 2013	4	4	2	0.5	biochar	0.565	0.181	amendment	-1.138
Cayuela et al 2013	4	4	2	0.5	biochar	0.321	0.174	amendment	-0.612
Cayuela et al 2013	4	4	2	0.5	biochar	1.534	0.798	amendment	-0.653
Cayuela et al 2013	4	4	2	0.5	biochar	3.424	0.672	amendment	-1.628
Cayuela et al 2013	4	4	2	0.5	biochar	0.097	0.021	amendment	-1.531
Dendooven et al 1996	4	4	2	0.5		0.313	0.386	N_fertilizer_application	0.209
Dendooven et al 1996	4	4	2	0.5		0.313	0.419	residue_retention	0.292
Dendooven et al 1998	5	5	2.5	0.4		0.172	0.177	fertilzer_incorporation	0.029
Dendooven et al 1998	5	5	2.5	0.4		0.087	0.172	N_fertilizer_application	0.682
Dendooven et al 1999	3	3	1.5	0.667		0.226	0.288	N_fertilizer_application	0.243
Guo XB et al 2013	4	4	2	0.5		1.164	1.445	N_fertilizer_application	0.216
McGeough et al 2012	4	4	2	0.5	DCD	1.043	0.712	inhibitor	-0.382
McGeough et al 2012	4	4	2	0.5	DCD	1.347	1.067	inhibitor	-0.232
Miller et al 2008	4	4	2	0.5		0.081	0.143	N_fertilizer_application	0.573
Miller et al 2008	4	4	2	0.5		0.081	0.205	N_fertilizer_application	0.934
Miller et al 2008	4	4	2	0.5		0.081	0.187	N_fertilizer_application	0.841
Miller et al 2008	4	4	2	0.5		0.055	0.103	N_fertilizer_application	0.624
Miller et al 2008	4	4	2	0.5		0.055	0.22	N_fertilizer_application	1.386
Miller et al 2008	4	4	2	0.5		0.055	0.488	N_fertilizer_application	2.182
Miller et al 2008	4	4	2	0.5		0.044	0.095	N_fertilizer_application	0.773
Miller et al 2008	4	4	2	0.5		0.044	0.187	N_fertilizer_application	1.447
Miller et al 2008	4	4	2	0.5		0.044	0.326	N_fertilizer_application	2.004
Parkin et al 1985	4	4	2	0.5		0.009	0.102	N_fertilizer_application	2.433
Parkin et al 1985	4	4	2	0.5		0.022	0.246	N_fertilizer_application	2.409
Radersma & Smit 2011	3	3	1.5	0.667		0.432	2.01	residue_retention	1.537
Rolston et al 1982	3	3	1.5	0.667		0.043	0.186	irrigation	1.462
Rolston et al 1982	3	3	1.5	0.667		0.043	0.064	irrigation	0.393
Rolston et al 1982	3	3	1.5	0.667		0.165	0.392	irrigation	0.869
Rolston et al 1982	3	3	1.5	0.667		0.165	0.484	irrigation	1.08
Rolston et al 1982	3	3	1.5	0.667		0.061	0.195	irrigation	1.159
Rolston et al 1982	3	3	1.5	0.667		0.061	0.068	irrigation	0.103
Tenuta and Sparling 2011	3	3	1.5	0.667		0.058	0.04	N_fertilizer_application	-0.378
Tenuta and Sparling 2011	3	3	1.5	0.667		0.064	0.123	N_fertilizer_application	0.65
Weier et al 1996	3	3	1.5	0.667		0.26	0.095	tillage	-1.01
Weier et al 1996	3	3	1.5	0.667		0.123	0.198	tillage	0.477
Egginton and Smith 1986	3	3	1.5	0.667		0.018	0.174	N_fertilizer_application	2.262
Egginton and Smith 1986	3	3	1.5	0.667		0.018	0.02	N_fertilizer_application	0.087
Ellis et al 1998	3	3	1.5	0.667		0.011	0.005	N_fertilizer_application	-0.811
Ellis et al 1998	3	3	1.5	0.667		0.032	0.038	fertilzer_incorporation	0.167
Ellis et al 1998	3	3	1.5	0.667		0.06	0.112	N_fertilizer_application	0.624

Ellis et al 1998	3	3	1.5	0.667	0.152	0.146	fertilzer_incorporation	-0.04
Estavillo et al 2002	4	4	2	0.5	0.122	0.226	N_fertilizer_application	0.62
Estavillo et al 2002	4	4	2	0.5	0.286	0.424	N_fertilizer_application	0.395
Estavillo et al 2002	4	4	2	0.5	0.326	0.532	N_fertilizer_application	0.49
Estavillo et al 2002	4	4	2	0.5	0.122	0.286	tillage	0.855
Estavillo et al 2002	4	4	2	0.5	0.122	0.326	tillage	0.987
Estavillo et al 2002	4	4	2	0.5	0.226	0.424	tillage	0.63
Estavillo et al 2002	4	4	2	0.5	0.226	0.532	tillage	0.857
Estavillo et al 1994	6	6	3	0.333	0.009	0.067	N_fertilizer_application	2.005
Estavillo et al 1994	6	6	3	0.333	0.009	0.1	N_fertilizer_application	2.406
Estavillo et al 1994	6	6	3	0.333	0.009	0.016	N_fertilizer_application	0.581
Estavillo et al 1994	6	6	3	0.333	0.009	0.022	N_fertilizer_application	0.91
Estavillo et al 1994	6	6	3	0.333	0.013	0.05	N_fertilizer_application	1.375
Estavillo et al 1994	6	6	3	0.333	0.013	0.059	N_fertilizer_application	1.551
Estavillo et al 1994	6	6	3	0.333	0.013	0.035	N_fertilizer_application	1.008
Estavillo et al 1994	6	6	3	0.333	0.013	0.06	N_fertilizer_application	1.565
Fan et al 1997	2	2	1	1	0.015	0.027	N_fertilizer_application	0.56
Fan et al 1997	2	2	1	1	0.015	0.03	N_fertilizer_application	0.66
Fan et al 1997	2	2	1	1	0.023	0.029	N_fertilizer_application	0.231
Fan et al 1997	2	2	1	1	0.023	0.028	N_fertilizer_application	0.205
Fan et al 1997	2	2	1	1	0.02	0.019	N_fertilizer_application	-0.086
Fan et al 1997	2	2	1	1	0.02	0.022	N_fertilizer_application	0.091
Fan et al 1997	2	2	1	1	0.039	0.068	N_fertilizer_application	0.565
Fan et al 1997	2	2	1	1	0.039	0.082	N_fertilizer_application	0.748
Fan et al 1997	2	2	1	1	0.044	0.06	N_fertilizer_application	0.311
Fan et al 1997	2	2	1	1	0.044	0.048	N_fertilizer_application	0.087
Fan et al 1997	2	2	1	1	0.044	0.041	N_fertilizer_application	-0.073
Fan et al 1997	2	2	1	1	0.044	0.032	N_fertilizer_application	-0.319
Fan et al 1997	4	4	2	0.5	0.014	0.042	N_fertilizer_application	1.101
Fan et al 1997	4	4	2	0.5	0.014	0.048	N_fertilizer_application	1.228
Fan et al 1997	4	4	2	0.5	0.014	0.09	N_fertilizer_application	1.867
Fan et al 1997	4	4	2	0.5	0.025	0.055	N_fertilizer_application	0.785
Fan et al 1997	4	4	2	0.5	0.025	0.123	N_fertilizer_application	1.589
Fan et al 1997	4	4	2	0.5	0.025	0.154	N_fertilizer_application	1.813
Goulding et al 1993	4	4	2	0.5	0.008	0.241	N_fertilizer_application	3.384
Goulding et al 1993	4	4	2	0.5	0.041	0.163	N_fertilizer_application	1.386
Huang et al 2015	3	3	1.5	0.667	0.076	0.091	irrigation	0.182
Huang et al 2015	3	3	1.5	0.667	0.076	0.09	irrigation	0.172
Huang et al 2015	3	3	1.5	0.667	0.016	0.067	N_fertilizer_application	1.415
Huang et al 2015	3	3	1.5	0.667	0.016	0.094	N_fertilizer_application	1.762
Huang et al 2015	3	3	1.5	0.667	0.016	0.126	N_fertilizer_application	2.05
Huang et al 2015	3	3	1.5	0.667	0.016	0.15	N_fertilizer_application	2.223
Huang et al 2015	3	3	1.5	0.667	0.003	0.004	irrigation	0.271
Huang et al 2015	3	3	1.5	0.667	0.003	0.005	irrigation	0.414
Huang et al 2015	3	3	1.5	0.667	0.003	0.005	irrigation	0.405
Huang et al 2015	3	3	1.5	0.667	0.001	0.004	N_fertilizer_application	1.721
Huang et al 2015	3	3	1.5	0.667	0.001	0.005	N_fertilizer_application	1.876
Huang et al 2015	3	3	1.5	0.667	0.001	0.006	N_fertilizer_application	2.116
Huang et al 2015	3	3	1.5	0.667	0.001	0.007	N_fertilizer_application	2.308
Huang et al 2015	3	3	1.5	0.667	0.041	0.054	irrigation	0.283

Huang et al 2015	3	3	1.5	0.667		0.041	0.055	irrigation	0.304
Huang et al 2015	3	3	1.5	0.667		0.041	0.062	irrigation	0.42
Huang et al 2015	3	3	1.5	0.667		0.009	0.04	N_fertilizer_application	1.481
Huang et al 2015	3	3	1.5	0.667		0.009	0.062	N_fertilizer_application	1.926
Huang et al 2015	3	3	1.5	0.667		0.009	0.066	N_fertilizer_application	1.984
Huang et al 2015	3	3	1.5	0.667		0.009	0.076	N_fertilizer_application	2.123
Huang et al 2015	3	3	1.5	0.667		0.005	0.006	irrigation	0.195
Huang et al 2015	3	3	1.5	0.667		0.005	0.007	irrigation	0.427
Huang et al 2015	3	3	1.5	0.667		0.005	0.007	irrigation	0.475
Huang et al 2015	3	3	1.5	0.667		0.001	0.004	N_fertilizer_application	1.548
Huang et al 2015	3	3	1.5	0.667		0.001	0.007	N_fertilizer_application	2.152
Huang et al 2015	3	3	1.5	0.667		0.001	0.008	N_fertilizer_application	2.257
Huang et al 2015	3	3	1.5	0.667		0.001	0.009	N_fertilizer_application	2.366
Huang et al 2015	3	3	1.5	0.667		0.083	0.094	irrigation	0.121
Huang et al 2015	3	3	1.5	0.667		0.083	0.103	irrigation	0.212
Huang et al 2015	3	3	1.5	0.667		0.083	0.108	irrigation	0.256
Huang et al 2015	3	3	1.5	0.667		0.013	0.067	N_fertilizer_application	1.615
Huang et al 2015	3	3	1.5	0.667		0.013	0.108	N_fertilizer_application	2.082
Huang et al 2015	3	3	1.5	0.667		0.013	0.124	N_fertilizer_application	2.22
Huang et al 2015	3	3	1.5	0.667		0.013	0.138	N_fertilizer_application	2.328
Jarvis et al 1991	3	3	1.5	0.667		0.009	0.009	N_fertilizer_application	0
Jarvis et al 1991	3	3	1.5	0.667		0.009	0.013	N_fertilizer_application	0.329
Jarvis et al 1991	3	3	1.5	0.667		0.009	0.015	N_fertilizer_application	0.491
Jarvis et al 1991	3	3	1.5	0.667		0.009	0.047	N_fertilizer_application	1.643
Jordan 1989	10	10	5	0.2	NH4NO3	0.089	0.188	fertilizer	0.752
Jordan 1989	10	10	5	0.2	NH4NO3	0.074	0.305	fertilizer	1.417
Jordan 1989	10	10	5	0.2		0.002	0.017	N_fertilizer_application	2.037
Jordan 1989	10	10	5	0.2		0.002	0.052	N_fertilizer_application	3.178
Jordan 1989	10	10	5	0.2		0.002	0.07	N_fertilizer_application	3.481
Jordan 1989	10	10	5	0.2		0.002	0.027	N_fertilizer_application	2.512
Jordan 1989	10	10	5	0.2	NH4NO3	0.027	0.07	fertilizer	0.969
Jordan 1989	10	10	5	0.2		0.002	0.009	N_fertilizer_application	1.329
Jordan 1989	10	10	5	0.2		0.002	0.027	N_fertilizer_application	2.398
Jordan 1989	10	10	5	0.2		0.002	0.041	N_fertilizer_application	2.82
Jordan 1989	10	10	5	0.2		0.002	0.008	N_fertilizer_application	1.237
Jordan 1989	10	10	5	0.2		0.002	0.024	N_fertilizer_application	2.291
Jordan 1989	10	10	5	0.2	NH4NO3	0.008	0.041	fertilizer	1.583
Jordan 1989	10	10	5	0.2	NH4-based	0.008	0.024	fertilizer	1.055
Kaluli et al 1999	3	3	1.5	0.667		0.051	0.081	N_fertilizer_application	0.474
Kaluli et al 1999	3	3	1.5	0.667		0.051	0.136	N_fertilizer_application	0.984
Kirkham and Wilkins 199	3	3	1.5	0.667		0.09	0.089	N_fertilizer_application	-0.011
Kirkham and Wilkins 199	3	3	1.5	0.667		0.09	0.168	N_fertilizer_application	0.624
Kirkham and Wilkins 199	3	3	1.5	0.667		0.06	0.078	N_fertilizer_application	0.253
Kirkham and Wilkins 199	3	3	1.5	0.667		0.06	0.137	N_fertilizer_application	0.824
Koops et al 1996	4	4	2	0.5		0.071	0.134	N_fertilizer_application	0.634
Koops et al 1996	4	4	2	0.5		0.096	0.195	N_fertilizer_application	0.707
Koops et al 1996	4	4	2	0.5		0.074	0.088	N_fertilizer_application	0.17
Koops et al 1996	4	4	2	0.5		0.036	0.047	N_fertilizer_application	0.268
Liang and MacKenzie 199	4	4	2	0.5		0.035	0.065	N_fertilizer_application	0.626
Liang and MacKenzie 199	4	4	2	0.5		0.035	0.089	N_fertilizer_application	0.931

Liang and MacKenzie 199	4	4	2	0.5		0.035	0.121	N_fertilizer_application	1.242
Liang and MacKenzie 199	4	4	2	0.5		0.158	0.202	N_fertilizer_application	0.244
Liang and MacKenzie 199	4	4	2	0.5		0.158	0.273	N_fertilizer_application	0.545
Liang and MacKenzie 199	4	4	2	0.5		0.158	0.29	N_fertilizer_application	0.603
Liang and MacKenzie 199	4	4	2	0.5		0.186	0.202	N_fertilizer_application	0.085
Liang and MacKenzie 199	4	4	2	0.5		0.186	0.137	N_fertilizer_application	-0.307
Liang and MacKenzie 199	4	4	2	0.5		0.186	0.153	N_fertilizer_application	-0.194
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.049	0.101	N_fertilizer_application	0.729
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.049	0.147	N_fertilizer_application	1.105
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.049	0.142	N_fertilizer_application	1.074
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.049	0.127	N_fertilizer_application	0.963
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.049	0.143	N_fertilizer_application	1.078
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.036	0.11	N_fertilizer_application	1.126
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.036	0.076	N_fertilizer_application	0.761
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.036	0.099	N_fertilizer_application	1.021
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.036	0.082	N_fertilizer_application	0.831
Lopez-Fernandez et al 20	3	3	1.5	0.667		0.036	0.121	N_fertilizer_application	1.222
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.101	0.147	fertilizer	0.377
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.101	0.142	fertilizer	0.346
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.101	0.127	fertilizer	0.234
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.101	0.143	fertilizer	0.349
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.11	0.076	fertilizer	-0.365
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.11	0.099	fertilizer	-0.106
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.11	0.082	fertilizer	-0.295
Lopez-Fernandez et al 20	3	3	1.5	0.667	organic fertilizer	0.11	0.121	fertilizer	0.096
Lubbers et al 2013	5	5	2.5	0.4		0.002	0.001	N_fertilizer_application	-0.48
Lubbers et al 2013	5	5	2.5	0.4		0.002	0.003	N_fertilizer_application	0.12
Lubbers et al 2013	5	5	2.5	0.4		0.036	0.065	N_fertilizer_application	0.582
Lubbers et al 2013	5	5	2.5	0.4		0.036	0.196	N_fertilizer_application	1.681
Maag et al 1995	4	4	2	0.5		0.002	0.016	N_fertilizer_application	2.071
Maag et al 1995	4	4	2	0.5		0.002	0.025	N_fertilizer_application	2.499
Maag et al 1995	4	4	2	0.5		0.022	0.034	N_fertilizer_application	0.405
Maag et al 1995	4	4	2	0.5		0.022	0.022	N_fertilizer_application	0
Maag et al 1995	4	4	2	0.5		0.008	0.024	N_fertilizer_application	1.076
Maag et al 1995	4	4	2	0.5		0.008	0.133	N_fertilizer_application	2.808
Maag et al 1995	4	4	2	0.5		0.001	0.001	N_fertilizer_application	-0.381
Maag et al 1995	4	4	2	0.5		0.001	0.001	N_fertilizer_application	0.198
Maag et al 1995	4	4	2	0.5		0.001	0.001	N_fertilizer_application	-0.536
Mahmood et al 2005	4	4	2	0.5		0.015	0.025	N_fertilizer_application	0.525
Mahmood et al 2005	4	4	2	0.5		0.015	0.038	N_fertilizer_application	0.957
Mahmood et al 2005	4	4	2	0.5		0.015	0.025	N_fertilizer_application	0.523
Mahmood et al 2005	4	4	2	0.5		0.015	0.031	N_fertilizer_application	0.738
Mahmood et al 2005	4	4	2	0.5		0.176	0.248	N_fertilizer_application	0.341
Mahmood et al 2005	4	4	2	0.5		0.176	0.367	N_fertilizer_application	0.735
Mahmood et al 2005	4	4	2	0.5		0.176	0.283	N_fertilizer_application	0.475
Mahmood et al 2005	4	4	2	0.5		0.176	0.24	N_fertilizer_application	0.311
Maris et al 2015	3	3	1.5	0.667		0.002	0.004	N_fertilizer_application	0.609
Maris et al 2015	3	3	1.5	0.667		0.003	0.005	N_fertilizer_application	0.457
Maris et al 2015	3	3	1.5	0.667		0.005	0.011	N_fertilizer_application	0.71
Maris et al 2015	3	3	1.5	0.667		0.002	0.004	N_fertilizer_application	0.806

Maris et al 2015	3	3	1.5	0.667		DMPP	0.004	0.002	inhibitor	-1.041
Maris et al 2015	3	3	1.5	0.667		DMPP	0.011	0.001	inhibitor	-2.874
Marshall et al 1999	3	3	1.5	0.667			0.012	0.018	N_fertilizer_application	0.456
Marshall et al 1999	3	3	1.5	0.667			0.042	0.033	N_fertilizer_application	-0.251
Marshall et al 1999	3	3	1.5	0.667			0.012	0.021	N_fertilizer_application	0.57
Marshall et al 1999	3	3	1.5	0.667			0.002	0.012	N_fertilizer_application	2.012
Marshall et al 1999	3	3	1.5	0.667			0.013	0.015	N_fertilizer_application	0.125
Marshall et al 1999	3	3	1.5	0.667			0.012	0.005	N_fertilizer_application	-0.941
Meijide et al 2007	3	3	1.5	0.667			0.061	0.179	N_fertilizer_application	1.077
Meijide et al 2007	3	3	1.5	0.667			0.061	0.182	N_fertilizer_application	1.093
Meijide et al 2007	3	3	1.5	0.667			0.061	0.121	N_fertilizer_application	0.684
Meijide et al 2007	3	3	1.5	0.667			0.061	0.238	N_fertilizer_application	1.361
Meijide et al 2007	3	3	1.5	0.667			0.061	0.32	N_fertilizer_application	1.656
Meijide et al 2007	3	3	1.5	0.667			0.061	0.22	N_fertilizer_application	1.282
Meijide et al 2007	3	3	1.5	0.667	organic fertilizer		0.22	0.179	fertilizer	-0.205
Meijide et al 2007	3	3	1.5	0.667		DCD	0.179	0.121	inhibitor	-0.393
Meijide et al 2009	3	3	1.5	0.667			0.025	0.057	N_fertilizer_application	0.81
Meijide et al 2009	3	3	1.5	0.667			0.025	0.063	N_fertilizer_application	0.916
Meijide et al 2009	3	3	1.5	0.667			0.025	0.016	N_fertilizer_application	-0.459
Meijide et al 2009	3	3	1.5	0.667			0.025	0.015	N_fertilizer_application	-0.538
Meijide et al 2009	3	3	1.5	0.667			0.025	0.003	N_fertilizer_application	-2.05
Meijide et al 2009	3	3	1.5	0.667	organic fertilizer		0.003	0.057	fertilizer	2.86
Menendez et al 2008	3	3	1.5	0.667			0.008	0.009	N_fertilizer_application	0.084
Menendez et al 2008	3	3	1.5	0.667			0.035	0.023	N_fertilizer_application	-0.422
Misselbrook et al 1996	3	3	1.5	0.667			0.009	0.04	N_fertilizer_application	1.474
Misselbrook et al 1996	3	3	1.5	0.667			0.009	0.033	N_fertilizer_application	1.295
Misselbrook et al 1996	3	3	1.5	0.667			0.033	0.04	fertilzer_incorporation	0.179
Pain et al 1990	3	3	1.5	0.667			0.029	0.003	N_fertilizer_application	-2.303
Pain et al 1990	3	3	1.5	0.667			0.029	0.017	N_fertilizer_application	-0.545
Pain et al 1990	3	3	1.5	0.667			0.001	0.014	N_fertilizer_application	3.178
Pain et al 1990	3	3	1.5	0.667			0.001	0.009	N_fertilizer_application	2.708
Pain et al 1990	3	3	1.5	0.667			0.001	0.001	N_fertilizer_application	0.693
Pain et al 1990	3	3	1.5	0.667			0.001	0.012	N_fertilizer_application	2.996
Pain et al 1990	3	3	1.5	0.667			0.009	0.191	N_fertilizer_application	3.088
Pain et al 1990	3	3	1.5	0.667			0.009	0.058	N_fertilizer_application	1.887
Pain et al 1990	3	3	1.5	0.667			0.009	0.285	N_fertilizer_application	3.488
Pain et al 1990	3	3	1.5	0.667			0.009	0.041	N_fertilizer_application	1.54
Pain et al 1990	3	3	1.5	0.667			0.009	0.227	N_fertilizer_application	3.261
Pain et al 1990	3	3	1.5	0.667			0.009	0.2	N_fertilizer_application	3.068
Pain et al 1990	3	3	1.5	0.667			0.009	0.149	N_fertilizer_application	2.773
Pain et al 1990	3	3	1.5	0.667			0.009	0.074	N_fertilizer_application	2.072
Pain et al 1990	3	3	1.5	0.667			0.009	0.048	N_fertilizer_application	1.646
Pain et al 1990	3	3	1.5	0.667		DCD	0.003	0.028	inhibitor	2.262
Pain et al 1990	3	3	1.5	0.667		DCD	0.017	0.02	inhibitor	0.188
Pain et al 1990	3	3	1.5	0.667		Nitrapyrin	0.017	0.004	inhibitor	-1.421
Pain et al 1990	3	3	1.5	0.667		DCD	0.014	0.009	inhibitor	-0.47
Pain et al 1990	3	3	1.5	0.667		DCD	0.014	0.001	inhibitor	-2.485
Pain et al 1990	3	3	1.5	0.667		DCD	0.014	0.012	inhibitor	-0.182
Pain et al 1990	3	3	1.5	0.667		DCD	0.191	0.058	inhibitor	-1.201
Pain et al 1990	3	3	1.5	0.667		Nitrapyrin	0.285	0.227	inhibitor	-0.228

Pain et al 1990	3	3	1.5	0.667		DCD	0.285	0.041	inhibitor	-1.948
Pain et al 1990	3	3	1.5	0.667		DCD	0.2	0.149	inhibitor	-0.295
Pain et al 1990	3	3	1.5	0.667		DCD	0.2	0.074	inhibitor	-0.996
Pain et al 1990	3	3	1.5	0.667		DCD	0.2	0.048	inhibitor	-1.422
Palma et al 1997	3	3	1.5	0.667			0.002	0.004	N_fertilizer_application	0.435
Palma et al 1997	3	3	1.5	0.667			0.004	0.005	fertilzer_incorporation	0.187
Palma et al 1997	3	3	1.5	0.667			0.002	0.002	N_fertilizer_application	0.194
Palma et al 1997	3	3	1.5	0.667			0.002	0.003	fertilzer_incorporation	0.345
Paul and Zebarth 1997	4	4	2	0.5			0.219	0.658	N_fertilizer_application	1.099
Paul and Zebarth 1997	4	4	2	0.5			0.019	0.106	N_fertilizer_application	1.735
Paul and Zebarth 1997	4	4	2	0.5			0.151	0.397	N_fertilizer_application	0.969
Paul and Zebarth 1997	4	4	2	0.5			0.044	0.125	N_fertilizer_application	1.05
Paul and Zebarth 1997b	4	4	2	0.5			0.201	0.872	N_fertilizer_application	1.466
Paul and Zebarth 1997b	4	4	2	0.5			0.201	0.598	N_fertilizer_application	1.089
Paul and Zebarth 1997b	4	4	2	0.5			0.201	0.419	N_fertilizer_application	0.734
Paul and Zebarth 1997b	4	4	2	0.5			0.201	0.218	N_fertilizer_application	0.08
Paul and Zebarth 1997b	4	4	2	0.5			0.207	0.531	N_fertilizer_application	0.943
Paul and Zebarth 1997b	4	4	2	0.5			0.207	0.151	N_fertilizer_application	-0.315
Paul and Zebarth 1997b	4	4	2	0.5			0.207	0.38	N_fertilizer_application	0.609
Paul and Zebarth 1997b	4	4	2	0.5			0.207	0.19	N_fertilizer_application	-0.085
Phoenix et al 2003	3	3	1.5	0.667			0.001	0.001	N_fertilizer_application	-0.185
Phoenix et al 2003	3	3	1.5	0.667			0.033	0.019	N_fertilizer_application	-0.547
Rozas et al 2001	3	3	1.5	0.667			0.062	0.136	N_fertilizer_application	0.792
Rozas et al 2001	3	3	1.5	0.667			0.062	0.189	N_fertilizer_application	1.119
Rozas et al 2001	3	3	1.5	0.667			0.034	0.054	N_fertilizer_application	0.444
Rozas et al 2001	3	3	1.5	0.667			0.034	0.078	N_fertilizer_application	0.816
Rozas et al 2001	3	3	1.5	0.667			0.131	0.254	N_fertilizer_application	0.663
Rozas et al 2001	3	3	1.5	0.667			0.131	0.316	N_fertilizer_application	0.882
Rozas et al 2001	3	3	1.5	0.667			0.032	0.049	N_fertilizer_application	0.445
Rozas et al 2001	3	3	1.5	0.667			0.032	0.033	N_fertilizer_application	0.055
Ryden 1983	3	3	1.5	0.667			0.004	0.03	N_fertilizer_application	1.937
Ryden 1983	3	3	1.5	0.667			0.004	0.08	N_fertilizer_application	2.901
Sanchez et al 2001	3	3	1.5	0.667			0.016	0.033	N_fertilizer_application	0.706
Sanchez et al 2001	3	3	1.5	0.667			0.016	0.056	N_fertilizer_application	1.244
Sanchez et al 2001	3	3	1.5	0.667	organic fertilizer		0.056	0.033	fertilizer	-0.538
Sanchez-Martin et al 200	3	3	1.5	0.667			0.053	0.055	N_fertilizer_application	0.02
Sanchez-Martin et al 200	3	3	1.5	0.667			0.122	0.188	N_fertilizer_application	0.426
Sanchez-Martin et al 201	3	3	1.5	0.667			0.051	0.122	N_fertilizer_application	0.872
Sanchez-Martin et al 201	3	3	1.5	0.667			0.088	0.174	N_fertilizer_application	0.682
Schwarz et al 1994	4	4	2	0.5			0.001	0.015	N_fertilizer_application	2.734
Schwarz et al 1994	4	4	2	0.5			0.001	0.003	N_fertilizer_application	1.209
Schwarz et al 1994	4	4	2	0.5			0.001	0.003	N_fertilizer_application	1.099
Schwarz et al 1994	4	4	2	0.5			0.002	0.007	N_fertilizer_application	1.216
Schwarz et al 1994	4	4	2	0.5			0.002	0.003	N_fertilizer_application	0.501
Schwarz et al 1994	4	4	2	0.5			0.002	0.004	N_fertilizer_application	0.615
Tenuta and Beauchamp 2	3	3	1.5	0.667			0.013	0.017	N_fertilizer_application	0.305
Tenuta and Beauchamp 2	3	3	1.5	0.667			0.013	0.01	N_fertilizer_application	-0.197
Tenuta and Beauchamp 2	3	3	1.5	0.667			0.013	0.006	N_fertilizer_application	-0.693
Tenuta and Beauchamp 2	3	3	1.5	0.667			0.013	0.009	N_fertilizer_application	-0.336
Tenuta and Beauchamp 2	3	3	1.5	0.667	NH4-based		0.017	0.01	fertilizer	-0.502

Tenuta and Beauchamp 2	3	3	1.5	0.667	NH4NO3		0.017	0.006	fertilizer	-0.999
Tenuta and Beauchamp 2	3	3	1.5	0.667	NO3-based		0.017	0.009	fertilizer	-0.642
Thompson and Meisinger	2	2	1	1			0.128	0.21	fertilzer_incorporation	0.496
Thompson and Meisinger	2	2	1	1			0.033	0.066	tillage	0.679
Thompson and Meisinger	2	2	1	1			0.033	0.128	N_fertilizer_application	1.342
Thompson and Meisinger	2	2	1	1			0.033	0.21	N_fertilizer_application	1.838
Ullah et al 2005	3	3	1.5	0.667			0.065	0.118	N_fertilizer_application	0.596
Ullah et al 2005	3	3	1.5	0.667			0.358	0.245	N_fertilizer_application	-0.379
Ullah et al 2005	3	3	1.5	0.667			0.185	0.787	N_fertilizer_application	1.449
Ullah et al 2005	3	3	1.5	0.667			0.324	0.49	N_fertilizer_application	0.413
Vallejo et al 2005	3	3	1.5	0.667			0.084	0.203	N_fertilizer_application	0.881
Vallejo et al 2005	3	3	1.5	0.667			0.084	0.27	N_fertilizer_application	1.162
Vallejo et al 2005	3	3	1.5	0.667			0.084	0.266	N_fertilizer_application	1.15
Vallejo et al 2005	3	3	1.5	0.667			0.203	0.27	fertilzer_incorporation	0.282
Vallejo et al 2005	3	3	1.5	0.667		DCD	0.27	0.266	inhibitor	-0.012
Vallejo et al 2006	3	3	1.5	0.667			0.052	0.186	N_fertilizer_application	1.27
Vallejo et al 2006	3	3	1.5	0.667			0.052	0.142	N_fertilizer_application	0.998
Vallejo et al 2006	3	3	1.5	0.667			0.052	0.186	N_fertilizer_application	1.267
Vallejo et al 2006	3	3	1.5	0.667			0.052	0.141	N_fertilizer_application	0.993
Vallejo et al 2006	3	3	1.5	0.667			0.052	0.218	N_fertilizer_application	1.426
Vallejo et al 2006	3	3	1.5	0.667			0.052	0.156	N_fertilizer_application	1.09
Vallejo et al 2006	3	3	1.5	0.667		DCD	0.186	0.186	inhibitor	-0.004
Vallejo et al 2006	3	3	1.5	0.667	organic fertilizer		0.156	0.186	fertilizer	0.18
Vallejo et al 2006	3	3	1.5	0.667	organic fertilizer		0.156	0.142	fertilizer	-0.092
Zaman et al 2010	3	3	1.5	0.667		lime	0.003	0.007	amendment	0.745
Zaman et al 2010	3	3	1.5	0.667		zeolite	0.003	0.006	amendment	0.52
Zaman et al 2010	3	3	1.5	0.667		lime	0.077	0.076	amendment	-0.013
Zaman et al 2010	3	3	1.5	0.667		zeolite	0.077	0.081	amendment	0.052
Zaman et al 2010	3	3	1.5	0.667		lime	0.162	0.165	amendment	0.015
Zaman et al 2010	3	3	1.5	0.667		zeolite	0.162	0.143	amendment	-0.13
Zaman et al 2010	3	3	1.5	0.667			0.003	0.077	N_fertilizer_application	3.106
Zaman et al 2010	3	3	1.5	0.667			0.003	0.162	N_fertilizer_application	3.855
Zaman et al 2008	3	3	1.5	0.667		zeolite	0.202	0.196	amendment	-0.029
Zaman et al 2008	3	3	1.5	0.667		lime	0.202	0.262	amendment	0.26
Zaman et al 2008	3	3	1.5	0.667		zeolite	0.584	0.632	amendment	0.079
Zaman et al 2008	3	3	1.5	0.667		lime	0.584	0.658	amendment	0.12
Zaman et al 2008	3	3	1.5	0.667			0.196	0.632	N_fertilizer_application	1.168
Zaman et al 2008	3	3	1.5	0.667			0.262	0.658	N_fertilizer_application	0.921
Zaman et al 2008	3	3	1.5	0.667			0.202	0.584	N_fertilizer_application	1.06
Zaman et al 2008	3	3	1.5	0.667		zeolite	0.005	0.004	amendment	-0.096
Zaman et al 2008	3	3	1.5	0.667		lime	0.005	0.004	amendment	-0.205
Zaman et al 2008	3	3	1.5	0.667		zeolite	2.102	2.023	amendment	-0.038
Zaman et al 2008	3	3	1.5	0.667		lime	2.102	2.437	amendment	0.148
Zaman et al 2008	3	3	1.5	0.667			0.005	2.102	N_fertilizer_application	6.077
Zaman et al 2008	3	3	1.5	0.667			0.004	2.437	N_fertilizer_application	6.43
Zaman et al 2008	3	3	1.5	0.667			0.004	2.023	N_fertilizer_application	6.135
Monaghan and Barraclou	1	1	0.5	2		Nitrapyrin	8.633	14.212	inhibitor	0.498
Dambreville et al 2006	8	8	4	0.25			0.138	0.007	N_fertilizer_application	-3.055
Dambreville et al 2006	8	8	4	0.25			0.117	0.003	N_fertilizer_application	-3.664
de Klein and van Logtestij	2	2	1	1			0.357	5	N_fertilizer_application	2.639

de Klein and van Logtestij	2	2	1	1			0.143	5.357	N_fertilizer_application	3.624
Dong et al 2014	3	3	1.5	0.667			0.007	0.046	N_fertilizer_application	1.903
Dong et al 2014	3	3	1.5	0.667			0.007	0.06	N_fertilizer_application	2.157
Dong et al 2014	3	3	1.5	0.667			0.007	0.042	N_fertilizer_application	1.794
Dong et al 2014	3	3	1.5	0.667			0.007	0.054	N_fertilizer_application	2.058
Dong et al 2014	3	3	1.5	0.667			0.007	0.036	N_fertilizer_application	1.664
Dong et al 2014	3	3	1.5	0.667			0.042	0.036	residue_retention	-0.13
Elmi et al 2003	3	3	1.5	0.667			0.01	0.009	tillage	-0.105
Elmi et al 2003	3	3	1.5	0.667			0.01	0.014	tillage	0.329
Elmi et al 2003	3	3	1.5	0.667			0.055	0.053	tillage	-0.046
Elmi et al 2003	3	3	1.5	0.667			0.055	0.037	tillage	-0.41
Friedl et al 2017	4	4	2	0.5		DMPP	1.051	0.289	inhibitor	-1.291
Friedl et al 2017	4	4	2	0.5		DMPP	1.387	0.357	inhibitor	-1.357
Friedl et al 2017	4	4	2	0.5		DMPP	0.196	0.208	inhibitor	0.059
Gillam et al 2008	4	4	2	0.5			0.407	0.397	N_fertilizer_application	-0.024
Gillam et al 2008	4	4	2	0.5			0.407	0.379	N_fertilizer_application	-0.07
Gillam et al 2008	4	4	2	0.5			0.407	0.39	N_fertilizer_application	-0.042
Gillam et al 2008	4	4	2	0.5			0.035	0.046	N_fertilizer_application	0.27
Gillam et al 2008	4	4	2	0.5			0.035	0.034	N_fertilizer_application	-0.035
Gillam et al 2008	4	4	2	0.5			0.449	0.476	N_fertilizer_application	0.06
Gillam et al 2008	4	4	2	0.5			0.449	0.371	N_fertilizer_application	-0.191
Gillam et al 2008	4	4	2	0.5			0.013	0.834	residue_retention	4.146
Gillam et al 2008	4	4	2	0.5			0.013	0.232	residue_retention	2.865
Gillam et al 2008	4	4	2	0.5			0.1	0.877	residue_retention	2.176
Gillam et al 2008	4	4	2	0.5			0.1	0.1	residue_retention	0
Gillam et al 2008	4	4	2	0.5			0.013	0.1	N_fertilizer_application	2.021
Gillam et al 2008	4	4	2	0.5			0.155	0.12	N_fertilizer_application	-0.255
Gillam et al 2008	4	4	2	0.5			0.282	0.313	N_fertilizer_application	0.105
Gillam et al 2008	4	4	2	0.5			0.718	0.468	N_fertilizer_application	-0.427
Gillam et al 2008	4	4	2	0.5			0.952	0.762	N_fertilizer_application	-0.222
Gillam et al 2008	4	4	2	0.5			0.834	0.877	N_fertilizer_application	0.051
Gillam et al 2008	4	4	2	0.5			0.232	0.1	N_fertilizer_application	-0.844
Gillam et al 2008	4	4	2	0.5			0.001	0.002	N_fertilizer_application	0.693
Gillam et al 2008	4	4	2	0.5			0.002	0.004	N_fertilizer_application	0.405
Gillam et al 2008	4	4	2	0.5			0.079	0.019	N_fertilizer_application	-1.417
Gillam et al 2008	4	4	2	0.5			0.089	0.065	N_fertilizer_application	-0.315
Gillam et al 2008	4	4	2	0.5			0.844	0.458	N_fertilizer_application	-0.61
Gillam et al 2008	4	4	2	0.5			1.459	1.703	N_fertilizer_application	0.154
Koops van Beusichem Oe	4	4	2	0.5			0.074	0.112	N_fertilizer_application	0.418
Koops van Beusichem Oe	4	4	2	0.5			0.063	0.121	N_fertilizer_application	0.649
Krause et al 2017	3	3	1.5	0.667			0.197	0.256	N_fertilizer_application	0.264
Krause et al 2017	3	3	1.5	0.667			0.197	0.179	N_fertilizer_application	-0.094
Li x et al 2014	3	3	1.5	0.667			0.173	0.304	fertilzer_incorporation	0.564
Lindau et al 1990	3	3	1.5	0.667	NH4NO3		0.072	0.173	fertilizer	0.873
Lindau et al 1990	3	3	1.5	0.667	NO3-based		0.072	0.877	fertilizer	2.495
Lindau et al 1990	3	3	1.5	0.667	NH4NO3		0.086	0.163	fertilizer	0.636
Lindau et al 1990	3	3	1.5	0.667	NO3-based		0.086	2.028	fertilizer	3.158
Liu xj et al 2007	3	3	1.5	0.667			1.843	1.875	tillage	0.017
Liu xj et al 2007	3	3	1.5	0.667			0.631	0.201	tillage	-1.142
Malghani et al 2018	3	3	1.5	0.667		biochar	0.063	0.091	amendment	0.362

Malghani et al 2018	3	3	1.5	0.667	biochar	0.063	0.103	amendment	0.481
Malghani et al 2018	3	3	1.5	0.667	biochar	0.063	0.081	amendment	0.248
Malghani et al 2018	3	3	1.5	0.667	biochar	0.063	0.112	amendment	0.564
McMillan et al 2016	3	3	1.5	0.667		0.115	0.367	N_fertilizer_application	1.161
McMillan et al 2016	3	3	1.5	0.667		0.139	0.442	N_fertilizer_application	1.156
McMillan et al 2016	3	3	1.5	0.667		0.162	0.448	N_fertilizer_application	1.018
McMillan et al 2016	3	3	1.5	0.667		0.181	1.1	N_fertilizer_application	1.803
McMillan et al 2016	3	3	1.5	0.667		0.147	1.266	N_fertilizer_application	2.15
McMillan et al 2016	3	3	1.5	0.667		0.217	1.151	N_fertilizer_application	1.669
McMillan et al 2016	3	3	1.5	0.667		0.272	0.581	N_fertilizer_application	0.76
McMillan et al 2016	3	3	1.5	0.667		0.208	0.493	N_fertilizer_application	0.862
McMillan et al 2016	3	3	1.5	0.667		0.205	0.513	N_fertilizer_application	0.918
McMillan et al 2016	3	3	1.5	0.667		0.288	0.851	N_fertilizer_application	1.083
McMillan et al 2016	3	3	1.5	0.667		0.308	0.923	N_fertilizer_application	1.097
McMillan et al 2016	3	3	1.5	0.667		0.317	0.88	N_fertilizer_application	1.021
McMillan et al 2016	3	3	1.5	0.667	lime	0.115	0.139	amendment	0.19
McMillan et al 2016	3	3	1.5	0.667	lime	0.115	0.162	amendment	0.341
McMillan et al 2016	3	3	1.5	0.667	lime	0.367	0.442	amendment	0.186
McMillan et al 2016	3	3	1.5	0.667	lime	0.367	0.448	amendment	0.199
McMillan et al 2016	3	3	1.5	0.667	lime	0.181	0.147	amendment	-0.207
McMillan et al 2016	3	3	1.5	0.667	lime	0.181	0.217	amendment	0.179
McMillan et al 2016	3	3	1.5	0.667	lime	1.1	1.266	amendment	0.141
McMillan et al 2016	3	3	1.5	0.667	lime	1.1	1.151	amendment	0.045
McMillan et al 2016	3	3	1.5	0.667	lime	0.272	0.208	amendment	-0.267
McMillan et al 2016	3	3	1.5	0.667	lime	0.272	0.205	amendment	-0.282
McMillan et al 2016	3	3	1.5	0.667	lime	0.581	0.493	amendment	-0.165
McMillan et al 2016	3	3	1.5	0.667	lime	0.581	0.513	amendment	-0.124
McMillan et al 2016	3	3	1.5	0.667	lime	0.288	0.308	amendment	0.067
McMillan et al 2016	3	3	1.5	0.667	lime	0.288	0.317	amendment	0.095
McMillan et al 2016	3	3	1.5	0.667	lime	0.851	0.923	amendment	0.081
McMillan et al 2016	3	3	1.5	0.667	lime	0.851	0.88	amendment	0.033
Merino et al 2001	4	4	2	0.5		0.066	0.259	N_fertilizer_application	1.365
Merino et al 2001	4	4	2	0.5		0.066	0.23	N_fertilizer_application	1.245
Merino et al 2001	4	4	2	0.5		0.066	0.368	N_fertilizer_application	1.716
Merino et al 2001	4	4	2	0.5		0.066	0.29	N_fertilizer_application	1.476
Merino et al 2001	4	4	2	0.5		0.066	0.167	N_fertilizer_application	0.927
Merino et al 2001	4	4	2	0.5	DCD	0.259	0.23	inhibitor	-0.12
Merino et al 2001	4	4	2	0.5	DCD	0.368	0.29	inhibitor	-0.24
Thompson 1989	4	4	2	0.5		0.023	0.473	N_fertilizer_application	3.01
Thompson 1989	4	4	2	0.5		0.023	0.068	N_fertilizer_application	1.07
Thompson 1989	4	4	2	0.5		0.023	0.121	N_fertilizer_application	1.65
Thompson 1989	4	4	2	0.5		0.023	0.087	N_fertilizer_application	1.312
Thompson 1989	4	4	2	0.5		0.023	0.081	N_fertilizer_application	1.245
Thompson 1989	4	4	2	0.5		0.023	0.081	N_fertilizer_application	1.245
Thompson 1989	4	4	2	0.5	DCD	0.473	0.068	inhibitor	-1.94
Thompson 1989	4	4	2	0.5	DCD	0.121	0.087	inhibitor	-0.338
Thompson 1989	4	4	2	0.5	DCD	0.081	0.081	inhibitor	0
Morkved et al 2006	3	3	1.5	0.667		0.089	0.418	N_fertilizer_application	1.55
Morkved et al 2006	3	3	1.5	0.667		0.277	0.34	N_fertilizer_application	0.205
Morkved et al 2006	3	3	1.5	0.667		0.042	0.222	N_fertilizer_application	1.663

Morkved et al 2006	3	3	1.5	0.667			0.16	0.18	N_fertilizer_application	0.116
Mukumbuta et al 2018	4	4	2	0.5		lime	0.034	0.047	amendment	0.306
Mukumbuta et al 2018	4	4	2	0.5		lime	0.034	0.089	amendment	0.953
Mukumbuta et al 2018	4	4	2	0.5		lime	0.329	0.88	amendment	0.985
Mukumbuta et al 2018	4	4	2	0.5		lime	0.329	1.057	amendment	1.168
Mukumbuta et al 2018	4	4	2	0.5		lime	0.175	0.409	amendment	0.852
Mukumbuta et al 2018	4	4	2	0.5		lime	0.175	0.722	amendment	1.42
Mukumbuta et al 2018	4	4	2	0.5		lime	0.937	6.22	amendment	1.893
Mukumbuta et al 2018	4	4	2	0.5		lime	0.937	6.066	amendment	1.868
Mukumbuta et al 2018	4	4	2	0.5			0.034	0.329	N_fertilizer_application	2.257
Mukumbuta et al 2018	4	4	2	0.5			0.047	0.88	N_fertilizer_application	2.936
Mukumbuta et al 2018	4	4	2	0.5			0.089	1.057	N_fertilizer_application	2.472
Mukumbuta et al 2018	4	4	2	0.5			0.175	0.937	N_fertilizer_application	1.68
Mukumbuta et al 2018	4	4	2	0.5			0.409	6.22	N_fertilizer_application	2.721
Mukumbuta et al 2018	4	4	2	0.5			0.722	6.066	N_fertilizer_application	2.128
Schuster and Conrad 199	3	3	1.5	0.667			0	0	N_fertilizer_application	0.678
Schuster and Conrad 199	3	3	1.5	0.667			0	0	N_fertilizer_application	1.95
Schuster and Conrad 199	3	3	1.5	0.667			0	0	N_fertilizer_application	-0.092
Schuster and Conrad 199	3	3	1.5	0.667	NH4-based		0	0	fertilizer	-1.272
Schuster and Conrad 199	3	3	1.5	0.667	NO3-based		0	0	fertilizer	-2.042
Sun zy et al 2018	3	3	1.5	0.667			0.007	4.377	residue_retention	6.438
Sun zy et al 2018	3	3	1.5	0.667			0.007	4.522	residue_retention	6.471
Sun zy et al 2018	3	3	1.5	0.667			0.007	4.822	residue_retention	6.535
Sun zy et al 2018	3	3	1.5	0.667			0.007	5.085	residue_retention	6.588
Vallejo et al 2003	3	3	1.5	0.667			0.062	0.13	N_fertilizer_application	0.731
Vallejo et al 2003	3	3	1.5	0.667			0.062	0.237	N_fertilizer_application	1.333
Vallejo et al 2003	3	3	1.5	0.667			0.062	0.167	N_fertilizer_application	0.986
Vallejo et al 2003	3	3	1.5	0.667			0.062	0.201	N_fertilizer_application	1.17
Vallejo et al 2003	3	3	1.5	0.667	organic fertilizer		0.13	0.237	fertilizer	0.603
Vallejo et al 2003	3	3	1.5	0.667	organic fertilizer		0.13	0.167	fertilizer	0.255
Vallejo et al 2003	3	3	1.5	0.667			0.167	0.201	fertilzer_incorporation	0.184
Vallejo et al 2001	3	3	1.5	0.667			0.179	0.3	N_fertilizer_application	0.518
Vallejo et al 2001	3	3	1.5	0.667			0.179	0.209	N_fertilizer_application	0.157
Vallejo et al 2001	3	3	1.5	0.667			0.179	0.404	N_fertilizer_application	0.816
Vallejo et al 2001	3	3	1.5	0.667			1.064	1.358	N_fertilizer_application	0.244
Vallejo et al 2001	3	3	1.5	0.667			1.064	1.75	N_fertilizer_application	0.498
Vallejo et al 2001	3	3	1.5	0.667			1.064	2.809	N_fertilizer_application	0.971
Vallejo et al 2001	3	3	1.5	0.667			0.242	0.131	N_fertilizer_application	-0.61
Vallejo et al 2001	3	3	1.5	0.667			0.242	0.222	N_fertilizer_application	-0.083
Vallejo et al 2001	3	3	1.5	0.667			1.304	1.264	N_fertilizer_application	-0.03
Vallejo et al 2001	3	3	1.5	0.667			1.304	1.559	N_fertilizer_application	0.179
Vallejo et al 2001	3	3	1.5	0.667			0.171	0.128	N_fertilizer_application	-0.288
Vallejo et al 2001	3	3	1.5	0.667			0.171	0.444	N_fertilizer_application	0.956
Vallejo et al 2001	3	3	1.5	0.667			0.5	0.521	N_fertilizer_application	0.041
Vallejo et al 2001	3	3	1.5	0.667			0.5	1.021	N_fertilizer_application	0.714
Vallejo et al 2001	3	3	1.5	0.667			0.5	1.479	N_fertilizer_application	1.085
Vallejo et al 2001	3	3	1.5	0.667		DCD	0.3	0.209	inhibitor	-0.361
Vallejo et al 2001	3	3	1.5	0.667	organic fertilizer		0.3	0.404	fertilizer	0.298
Vallejo et al 2001	3	3	1.5	0.667		DCD	1.358	1.75	inhibitor	0.254
Vallejo et al 2001	3	3	1.5	0.667	organic fertilizer		1.358	2.809	fertilizer	0.727

Vallejo et al 2001	3	3	1.5	0.667		DCD	0.131	0.198	inhibitor	0.413
Vallejo et al 2001	3	3	1.5	0.667	organic fertilizer		0.131	0.222	fertilizer	0.527
Vallejo et al 2001	3	3	1.5	0.667		DCD	1.264	1.049	inhibitor	-0.187
Vallejo et al 2001	3	3	1.5	0.667	organic fertilizer		1.264	1.559	fertilizer	0.209
Vallejo et al 2001	3	3	1.5	0.667		DCD	0.128	0.103	inhibitor	-0.223
Vallejo et al 2001	3	3	1.5	0.667	organic fertilizer		0.128	0.444	fertilizer	1.243
Vallejo et al 2001	3	3	1.5	0.667		DCD	0.521	1.021	inhibitor	0.673
Vallejo et al 2001	3	3	1.5	0.667	organic fertilizer		0.521	1.479	fertilizer	1.044
Velthof et al 1997	4	4	2	0.5	NH4NO3		0.048	0.211	fertilizer	1.474
Velthof et al 1997	4	4	2	0.5	NO3-based		0.048	0.267	fertilizer	1.71
Velthof et al 1997	4	4	2	0.5	NH4NO3		0.031	0.403	fertilizer	2.551
Velthof et al 1997	4	4	2	0.5	NO3-based		0.031	0.354	fertilizer	2.422
Velthof et al 1997	4	4	2	0.5	NH4-based		0.031	0.017	fertilizer	-0.606
Velthof et al 1997	4	4	2	0.5	NH4-based		0.031	0.006	fertilizer	-1.705
Velthof et al 1997	4	4	2	0.5		DCD	0.017	0.006	inhibitor	-1.099
Wang S et al 2017	4	4	2	0.5			0.184	0.307	N_fertilizer_application	0.512
Wang S et al 2017	4	4	2	0.5			0.184	0.337	N_fertilizer_application	0.605
Wang S et al 2017	4	4	2	0.5			0.184	0.275	N_fertilizer_application	0.4
Wang S et al 2017	4	4	2	0.5			0.467	0.619	N_fertilizer_application	0.282
Wang S et al 2017	4	4	2	0.5			0.467	0.64	N_fertilizer_application	0.315
Wang S et al 2017	4	4	2	0.5			0.467	0.499	N_fertilizer_application	0.066
Wang S et al 2017	4	4	2	0.5		biochar	0.307	0.337	amendment	0.093
Wang S et al 2017	4	4	2	0.5		Nitrapyrin	0.307	0.275	inhibitor	-0.111
Wang S et al 2017	4	4	2	0.5		biochar	0.619	0.64	amendment	0.033
Wang S et al 2017	4	4	2	0.5		Nitrapyrin	0.619	0.499	inhibitor	-0.216
Weier et al 1993	3	3	1.5	0.667			0.01	0.009	N_fertilizer_application	-0.105
Weier et al 1993	3	3	1.5	0.667			0.044	0.124	N_fertilizer_application	1.036
Weier et al 1993	3	3	1.5	0.667			0.076	0.237	N_fertilizer_application	1.137
Weier et al 1993	3	3	1.5	0.667			0.007	2.672	N_fertilizer_application	5.945
Weier et al 1993	3	3	1.5	0.667			0.025	4.77	N_fertilizer_application	5.251
Weier et al 1993	3	3	1.5	0.667			0.071	5.367	N_fertilizer_application	4.325
Weier et al 1993	3	3	1.5	0.667			0.009	1.3	N_fertilizer_application	4.973
Weier et al 1993	3	3	1.5	0.667			0.051	6.51	N_fertilizer_application	4.849
Weier et al 1993	3	3	1.5	0.667			0.079	4.923	N_fertilizer_application	4.132
Weier et al 1993	3	3	1.5	0.667			0.01	0.005	N_fertilizer_application	-0.693
Weier et al 1993	3	3	1.5	0.667			0.044	0.087	N_fertilizer_application	0.682
Weier et al 1993	3	3	1.5	0.667			0.076	0.21	N_fertilizer_application	1.016
Weier et al 1993	3	3	1.5	0.667			0.007	1.309	N_fertilizer_application	5.231
Weier et al 1993	3	3	1.5	0.667			0.025	8.919	N_fertilizer_application	5.877
Weier et al 1993	3	3	1.5	0.667			0.071	11.272	N_fertilizer_application	5.067
Weier et al 1993	3	3	1.5	0.667			0.009	2.606	N_fertilizer_application	5.668
Weier et al 1993	3	3	1.5	0.667			0.051	13.23	N_fertilizer_application	5.558
Weier et al 1993	3	3	1.5	0.667			0.079	9.383	N_fertilizer_application	4.777
Weier et al 1993	3	3	1.5	0.667			0.013	0.006	N_fertilizer_application	-0.773
Weier et al 1993	3	3	1.5	0.667			0.018	0.016	N_fertilizer_application	-0.118
Weier et al 1993	3	3	1.5	0.667			0.203	0.36	N_fertilizer_application	0.573
Weier et al 1993	3	3	1.5	0.667			0.014	0.087	N_fertilizer_application	1.827
Weier et al 1993	3	3	1.5	0.667			0.091	3.684	N_fertilizer_application	3.701
Weier et al 1993	3	3	1.5	0.667			0.195	6.287	N_fertilizer_application	3.473
Weier et al 1993	3	3	1.5	0.667			0.028	0.541	N_fertilizer_application	2.961

Weier et al 1993	3	3	1.5	0.667	0.188	3.717	N_fertilizer_application	2.984
Weier et al 1993	3	3	1.5	0.667	0.187	8.814	N_fertilizer_application	3.853
Weier et al 1993	3	3	1.5	0.667	0.013	0.002	N_fertilizer_application	-1.872
Weier et al 1993	3	3	1.5	0.667	0.018	0.036	N_fertilizer_application	0.693
Weier et al 1993	3	3	1.5	0.667	0.203	0.303	N_fertilizer_application	0.401
Weier et al 1993	3	3	1.5	0.667	0.014	0.07	N_fertilizer_application	1.609
Weier et al 1993	3	3	1.5	0.667	0.091	1.281	N_fertilizer_application	2.645
Weier et al 1993	3	3	1.5	0.667	0.195	14.227	N_fertilizer_application	4.29
Weier et al 1993	3	3	1.5	0.667	0.028	0.23	N_fertilizer_application	2.106
Weier et al 1993	3	3	1.5	0.667	0.188	10.999	N_fertilizer_application	4.069
Weier et al 1993	3	3	1.5	0.667	0.187	26.806	N_fertilizer_application	4.965
Weier et al 1993	3	3	1.5	0.667	0.01	0.003	N_fertilizer_application	-1.204
Weier et al 1993	3	3	1.5	0.667	0.001	0.002	N_fertilizer_application	0.693
Weier et al 1993	3	3	1.5	0.667	0.098	0.006	N_fertilizer_application	-2.793
Weier et al 1993	3	3	1.5	0.667	0.009	0.034	N_fertilizer_application	1.329
Weier et al 1993	3	3	1.5	0.667	0.096	0.11	N_fertilizer_application	0.136
Weier et al 1993	3	3	1.5	0.667	1.396	9.041	N_fertilizer_application	1.868
Weier et al 1993	3	3	1.5	0.667	0.017	0.121	N_fertilizer_application	1.963
Weier et al 1993	3	3	1.5	0.667	0.104	2.21	N_fertilizer_application	3.056
Weier et al 1993	3	3	1.5	0.667	1.697	9.935	N_fertilizer_application	1.767
Weier et al 1993	3	3	1.5	0.667	0.01	0.003	N_fertilizer_application	-1.204
Weier et al 1993	3	3	1.5	0.667	0.001	0.002	N_fertilizer_application	0.693
Weier et al 1993	3	3	1.5	0.667	0.098	0.027	N_fertilizer_application	-1.289
Weier et al 1993	3	3	1.5	0.667	0.009	0.008	N_fertilizer_application	-0.118
Weier et al 1993	3	3	1.5	0.667	0.096	0.134	N_fertilizer_application	0.333
Weier et al 1993	3	3	1.5	0.667	1.396	6.514	N_fertilizer_application	1.54
Weier et al 1993	3	3	1.5	0.667	0.017	0.093	N_fertilizer_application	1.699
Weier et al 1993	3	3	1.5	0.667	0.104	0.773	N_fertilizer_application	2.006
Weier et al 1993	3	3	1.5	0.667	1.697	17.479	N_fertilizer_application	2.332
Weier et al 1993	3	3	1.5	0.667	0.007	0.002	N_fertilizer_application	-1.253
Weier et al 1993	3	3	1.5	0.667	0.001	0.007	N_fertilizer_application	1.946
Weier et al 1993	3	3	1.5	0.667	0.087	0.172	N_fertilizer_application	0.682
Weier et al 1993	3	3	1.5	0.667	0.011	0.033	N_fertilizer_application	1.099
Weier et al 1993	3	3	1.5	0.667	0.156	0.76	N_fertilizer_application	1.583
Weier et al 1993	3	3	1.5	0.667	3.85	12.652	N_fertilizer_application	1.19
Weier et al 1993	3	3	1.5	0.667	0.005	0.023	N_fertilizer_application	1.526
Weier et al 1993	3	3	1.5	0.667	1.654	1.65	N_fertilizer_application	-0.002
Weier et al 1993	3	3	1.5	0.667	4.214	12.669	N_fertilizer_application	1.101
Weier et al 1993	3	3	1.5	0.667	0.007	0.002	N_fertilizer_application	-1.253
Weier et al 1993	3	3	1.5	0.667	0.001	0.001	N_fertilizer_application	0
Weier et al 1993	3	3	1.5	0.667	0.087	0.317	N_fertilizer_application	1.293
Weier et al 1993	3	3	1.5	0.667	0.011	0.002	N_fertilizer_application	-1.705
Weier et al 1993	3	3	1.5	0.667	0.156	0.034	N_fertilizer_application	-1.523
Weier et al 1993	3	3	1.5	0.667	3.85	10.565	N_fertilizer_application	1.009
Weier et al 1993	3	3	1.5	0.667	0.005	0.013	N_fertilizer_application	0.956
Weier et al 1993	3	3	1.5	0.667	1.654	0.268	N_fertilizer_application	-1.82
Weier et al 1993	3	3	1.5	0.667	4.214	28.065	N_fertilizer_application	1.896
Zhong et al 2018	3	3	1.5	0.667	11.547	9.509	residue_retention	-0.194
Zhong et al 2018	3	3	1.5	0.667	10.505	7.615	residue_retention	-0.322
Elliott et al 1991	4	4	2	0.5	0.072	0.236	N_fertilizer_application	1.183

Elliott et al 1991	4	4	2	0.5		0.084	0.175	N_fertilizer_application	0.738
Moster et al 1986	2	2	1	1		0.045	0.163	irrigation	1.281
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	0.815	residue_retention	0.297
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	1.002	residue_retention	0.503
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	1.141	residue_retention	0.634
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	1.188	residue_retention	0.674
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	0.769	residue_retention	0.238
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	0.71	residue_retention	0.16
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	1.037	residue_retention	0.537
Aulakh et al 1991 residue	3	3	1.5	0.667		0.606	1.118	residue_retention	0.613
Mulvaney et al 1997	3	3	1.5	0.667		0.125	1.283	N_fertilizer_application	2.325
Mulvaney et al 1997	3	3	1.5	0.667		0.125	1.096	N_fertilizer_application	2.168
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.393	N_fertilizer_application	1.142
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.73	N_fertilizer_application	1.762
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.423	N_fertilizer_application	1.215
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.513	N_fertilizer_application	1.409
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.348	N_fertilizer_application	1.02
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.428	N_fertilizer_application	1.227
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.306	N_fertilizer_application	0.891
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.462	N_fertilizer_application	1.305
Mulvaney et al 1997	3	3	1.5	0.667		0.125	0.393	N_fertilizer_application	1.142
Mulvaney et al 1997	3	3	1.5	0.667	NH4-based	1.096	1.283	fertilizer	0.157
Mulvaney et al 1997	3	3	1.5	0.667	NH4-based	1.096	0.73	fertilizer	-0.406
Mulvaney et al 1997	3	3	1.5	0.667	NH4-based	1.096	0.513	fertilizer	-0.759
Mulvaney et al 1997	3	3	1.5	0.667	NH4NO3	1.096	0.428	fertilizer	-0.942
Mulvaney et al 1997	3	3	1.5	0.667	NH4-based	1.096	0.462	fertilizer	-0.863
Mulvaney et al 1997	3	3	1.5	0.667	NH4-based	0.393	0.423	fertilizer	0.073
Mulvaney et al 1997	3	3	1.5	0.667	NH4-based	0.393	0.348	fertilizer	-0.122
Mulvaney et al 1997	3	3	1.5	0.667	NH4NO3	0.393	0.306	fertilizer	-0.251
Mulvaney et al 1997	3	3	1.5	0.667	NH4-based	0.393	0.393	fertilizer	-0.001
Horgan et al 2002	3	3	1.5	0.667	NO3-based	0.199	0.301	fertilizer	0.416
Sexstone et al 1988	8	8	4	0.25		0.018	0.035	irrigation	0.636
Sexstone et al 1988	8	8	4	0.25		0.018	0.065	irrigation	1.258
Sexstone et al 1988	8	8	4	0.25		0.061	0.083	irrigation	0.306
Sexstone et al 1988	8	8	4	0.25		0.061	0.13	irrigation	0.758
Sexstone et al 1988	8	8	4	0.25		0.001	0.001	irrigation	0.215
Sexstone et al 1988	8	8	4	0.25		0.001	0.003	irrigation	1.044
Sexstone et al 1988	8	8	4	0.25		0.095	0.193	irrigation	0.706
Sexstone et al 1988	8	8	4	0.25		0.095	0.206	irrigation	0.774
Sexstone et al 1988	8	8	4	0.25		0.148	0.154	irrigation	0.044
Sexstone et al 1988	8	8	4	0.25		0.148	0.222	irrigation	0.408
Sexstone et al 1988	8	8	4	0.25		0.066	0.098	irrigation	0.395
Sexstone et al 1988	8	8	4	0.25		0.066	0.186	irrigation	1.042

Supporting Table S2 Studies used in the meta-analysis to examine the effects of mitigation strategies on soil N_2 production

author/year	repilcation		weight	variance	fertilizer type	amendment/inhibitor	N2 emission from denitrification (kg N ha per day)		management practice	lnRR
	ck	treatment			treatment	treatment	CK	treatment	category	
Ciarlo et al 2008	3	3	1.5	0.67			0.001	0.001	N_fertilizer_application	0.208
Ciarlo et al 2008	3	3	1.5	0.67			0.001	0.001	N_fertilizer_application	0.143
Ciarlo et al 2008	3	3	1.5	0.67			0.001	0.004	split_application	1.658
Ciarlo et al 2008	3	3	1.5	0.67			0.001	0.003	split_application	1.623
Ciarlo et al 2008	3	3	1.5	0.67			0.041	0.144	N_fertilizer_application	1.247
Ciarlo et al 2008	3	3	1.5	0.67			0.041	0.068	N_fertilizer_application	0.495
Ciarlo et al 2008	3	3	1.5	0.67			0.144	0.068	split_application	-0.752
Ciarlo et al 2008	3	3	1.5	0.67			0.046	0.05	split_application	0.069
Ciarlo et al 2008	3	3	1.5	0.67			0.043	0.614	N_fertilizer_application	2.661
Ciarlo et al 2008	3	3	1.5	0.67			0.043	0.473	N_fertilizer_application	2.4
Ciarlo et al 2008	3	3	1.5	0.67			0.614	0.473	split_application	-0.262
Ciarlo et al 2008	3	3	1.5	0.67			0.046	0.041	split_application	-0.116
Ciarlo et al 2008	3	3	1.5	0.67	NO3-based		0.001	0.001	fertilizer_type	0.065
Ciarlo et al 2008	3	3	1.5	0.67	NO3-based		0.003	0.004	fertilizer_type	0.1
Ciarlo et al 2008	3	3	1.5	0.67	NO3-based		0.046	0.144	fertilizer_type	1.134
Ciarlo et al 2008	3	3	1.5	0.67	NO3-based		0.05	0.068	fertilizer_type	0.312
Ciarlo et al 2008	3	3	1.5	0.67	NO3-based		0.046	0.614	fertilizer_type	2.586
Ciarlo et al 2008	3	3	1.5	0.67	NO3-based		0.041	0.473	fertilizer_type	2.44
Duxbury and McConnaughey	3	3	1.5	0.67			0.016	0.02	N_fertilizer_application	0.245
Duxbury and McConnaughey	3	3	1.5	0.67			0.016	0.024	N_fertilizer_application	0.408
Duxbury and McConnaughey	3	3	1.5	0.67	NO3-based		0.024	0.02	fertilizer_type	-0.163
Blackmer and Bremner 1978	3	3	1.5	0.67			0.222	0.778	N_fertilizer_application	1.253
Blackmer and Bremner 1978	3	3	1.5	0.67			0.222	1.333	N_fertilizer_application	1.792
Blackmer and Bremner 1978	3	3	1.5	0.67			0.222	1.333	N_fertilizer_application	1.792
Blackmer and Bremner 1978	3	3	1.5	0.67			0.222	0.9	N_fertilizer_application	1.399
Blackmer and Bremner 1978	3	3	1.5	0.67			0.222	0.511	N_fertilizer_application	0.833
Blackmer and Bremner 1978	3	3	1.5	0.67			0.078	0.622	N_fertilizer_application	2.079
Blackmer and Bremner 1978	3	3	1.5	0.67			0.078	0.544	N_fertilizer_application	1.946
Blackmer and Bremner 1978	3	3	1.5	0.67			0.078	0.333	N_fertilizer_application	1.455
Blackmer and Bremner 1978	3	3	1.5	0.67			0.078	0.078	N_fertilizer_application	0
Blackmer and Bremner 1978	3	3	1.5	0.67			0.078	0.011	N_fertilizer_application	-1.946
Chen T et al 2019	3	3	1.5	0.67			0.023	0.055	N_fertilizer_application	0.869
Chen T et al 2019	3	3	1.5	0.67			0.023	0.062	N_fertilizer_application	0.989
Chen T et al 2019	3	3	1.5	0.67			0.023	0.068	N_fertilizer_application	1.079
Fangueiro et al 2008	5	5	2.5	0.4			0.001	0.002	N_fertilizer_application	0.832
Fangueiro et al 2008	5	5	2.5	0.4			0.001	0.002	N_fertilizer_application	0.816
Fangueiro et al 2008	5	5	2.5	0.4			0.001	0.004	N_fertilizer_application	1.303
Fangueiro et al 2008	5	5	2.5	0.4			0.001	0.004	N_fertilizer_application	1.257
Fangueiro et al 2008	5	5	2.5	0.4			0.001	0.004	N_fertilizer_application	1.308
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.546
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.527
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.655
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.047
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.002
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	0.936
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.075
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.369
Lai and Denton 2018	4	4	2	0.5			0	0	N_fertilizer_application	1.733
Loick et al 2017	4	4	2	0.5			0.29	0.33	N_fertilizer_application	0.129
Loick et al 2017	4	4	2	0.5			0.29	0.15	N_fertilizer_application	-0.659
Loick et al 2017	12	12	6	0.17			0.05	0.1	N_fertilizer_application	0.693
Loick et al 2017	12	12	6	0.17			0.05	0.23	N_fertilizer_application	1.526

Qin SP et al 2017 temperal	4	4	2	0.5		0.213	0.29	N_fertilizer_application	0.308
Zistl-Schlingmann et al 2019	4	4	2	0.5		0.141	1.15	N_fertilizer_application	2.102
Zistl-Schlingmann et al 2019	4	4	2	0.5		0.072	1.529	N_fertilizer_application	3.054
Senbayram, et al 2019	3	3	1.5	0.67		0.113	0.359	N_fertilizer_application	1.157
Senbayram, et al 2019	3	3	1.5	0.67		0.086	0.25	N_fertilizer_application	1.072
Senbayram, et al 2019	3	3	1.5	0.67		0.103	0.164	N_fertilizer_application	0.469
Senbayram, et al 2019	3	3	1.5	0.67		0.103	0.033	N_fertilizer_application	-1.132
Senbayram, et al 2019	3	3	1.5	0.67		0.024	0.093	N_fertilizer_application	1.335
Senbayram, et al 2019	3	3	1.5	0.67		0.024	0.036	N_fertilizer_application	0.386
Senbayram, et al 2018	3	3	1.5	0.67		0.023	0.019	N_fertilizer_application	-0.212
Senbayram, et al 2018	3	3	1.5	0.67		0.019	0.027	residue_retention	0.373
Senbayram, et al 2018	3	3	1.5	0.67		0.019	0.055	residue_retention	1.077
Senbayram, et al 2018	3	3	1.5	0.67		0.019	0.068	residue_retention	1.29
Wallenstein et al 2006	3	3	1.5	0.67		0.018	0.017	N_fertilizer_application	-0.045
Wallenstein et al 2006	3	3	1.5	0.67		0.37	0.362	N_fertilizer_application	-0.023
Wallenstein et al 2006	3	3	1.5	0.67		0.015	0.017	N_fertilizer_application	0.09
Wu D et al 2018	3	3	1.5	0.67		0.937	0.558	N_fertilizer_application	-0.517
Wu D et al 2018	3	3	1.5	0.67		0.558	2.709	residue_retention	1.579
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.001	residue_retention	0.134
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.001	residue_retention	0.409
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.001	residue_retention	-0.014
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.005	residue_retention	2.151
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.009	residue_retention	2.691
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.002	residue_retention	1.291
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.057	residue_retention	4.527
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.071	residue_retention	4.748
Zhu TB et al 2013	3	3	1.5	0.67		0.001	0.069	residue_retention	4.726
Cai G et al 2002	4	4	2	0.5		0.643	0.263	fertilizer_incorporation	-0.896
Cai G et al 2002	4	4	2	0.5		0.263	0.643	N_fertilizer_application	0.896
Cai G et al 2002	4	4	2	0.5		0.355	0.309	irrigation	-0.137
Cai G et al 2002	4	4	2	0.5		0.345	0.355	N_fertilizer_application	0.026
Carter et al 2007	4	4	2	0.5		0.015	0.094	N_fertilizer_application	1.851
Cayuela et al 2013	4	4	2	0.5	biochar	0.434	0.426	amendment	-0.019
Cayuela et al 2013	4	4	2	0.5	biochar	0.001	0.005	amendment	2.2
Cayuela et al 2013	4	4	2	0.5	biochar	1.19	0.593	amendment	-0.696
Cayuela et al 2013	4	4	2	0.5	biochar	0.039	0.186	amendment	1.55
Cayuela et al 2013	4	4	2	0.5	biochar	0.052	0.05	amendment	-0.033
Cayuela et al 2013	4	4	2	0.5	biochar	0.102	0.074	amendment	-0.324
Cayuela et al 2013	4	4	2	0.5	biochar	0.355	0.112	amendment	-1.156
Cayuela et al 2013	4	4	2	0.5	biochar	0.357	0.099	amendment	-1.281
Cayuela et al 2013	4	4	2	0.5	biochar	0.006	0.017	amendment	1.058
Cayuela et al 2013	4	4	2	0.5	biochar	0.089	0.298	amendment	1.204
Cayuela et al 2013	4	4	2	0.5	biochar	0.016	0.026	amendment	0.478
Cayuela et al 2013	4	4	2	0.5	biochar	0.115	0.085	amendment	-0.298
Cayuela et al 2013	4	4	2	0.5	biochar	0.831	0.655	amendment	-0.238
Cayuela et al 2013	4	4	2	0.5	biochar	1.822	0.508	amendment	-1.277
Cayuela et al 2013	4	4	2	0.5	biochar	0.021	0.012	amendment	-0.6
Dendooven et al 1996	4	4	2	0.5		0.117	0.162	N_fertilizer_application	0.323
Dendooven et al 1996	4	4	2	0.5		0.117	0.168	residue_retention	0.357
Dendooven et al 1999	3	3	1.5	0.67		0.153	0.136	N_fertilizer_application	-0.121
Guo XB et al 2013	4	4	2	0.5		0.75	0.409	N_fertilizer_application	-0.606
McGeough et al 2012	4	4	2	0.5	DCD	0.823	0.523	nitrification_inhibitor	-0.452
McGeough et al 2012	4	4	2	0.5	DCD	0.94	0.674	nitrification_inhibitor	-0.333
Miller et al 2008	4	4	2	0.5		0.073	0.114	N_fertilizer_application	0.438
Miller et al 2008	4	4	2	0.5		0.073	0.147	N_fertilizer_application	0.693

Miller et al 2008	4	4	2	0.5			0.073	0.092	N_fertilizer_application	0.223
Miller et al 2008	4	4	2	0.5			0.054	0.088	N_fertilizer_application	0.497
Miller et al 2008	4	4	2	0.5			0.054	0.18	N_fertilizer_application	1.211
Miller et al 2008	4	4	2	0.5			0.054	0.385	N_fertilizer_application	1.973
Miller et al 2008	4	4	2	0.5			0.043	0.092	N_fertilizer_application	0.751
Miller et al 2008	4	4	2	0.5			0.043	0.18	N_fertilizer_application	1.424
Miller et al 2008	4	4	2	0.5			0.043	0.282	N_fertilizer_application	1.876
Rolston et al 1982	3	3	1.5	0.67			0.036	0.136	irrigation	1.322
Rolston et al 1982	3	3	1.5	0.67			0.036	0.052	irrigation	0.358
Rolston et al 1982	3	3	1.5	0.67			0.129	0.345	irrigation	0.983
Rolston et al 1982	3	3	1.5	0.67			0.129	0.463	irrigation	1.278
Rolston et al 1982	3	3	1.5	0.67			0.045	0.159	irrigation	1.253
Rolston et al 1982	3	3	1.5	0.67			0.045	0.052	irrigation	0.135
Tenuta and Sparling 2011	3	3	1.5	0.67			0.025	0.029	N_fertilizer_application	0.144
Tenuta and Sparling 2011	3	3	1.5	0.67			0.024	0.107	N_fertilizer_application	1.499
Weier et al 1996	3	3	1.5	0.67			0.241	0.095	tillage	-0.934
Weier et al 1996	3	3	1.5	0.67			0.123	0.173	tillage	0.341
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.039	0.086	N_fertilizer_application	0.805
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.039	0.129	N_fertilizer_application	1.206
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.039	0.128	N_fertilizer_application	1.196
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.039	0.112	N_fertilizer_application	1.064
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.039	0.127	N_fertilizer_application	1.189
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.029	0.087	N_fertilizer_application	1.117
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.029	0.064	N_fertilizer_application	0.813
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.029	0.083	N_fertilizer_application	1.071
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.029	0.064	N_fertilizer_application	0.812
Lopez-Fernandez et al 2007	3	3	1.5	0.67			0.029	0.101	N_fertilizer_application	1.269
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.086	0.129	fertilizer_type	0.401
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.086	0.128	fertilizer_type	0.391
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.086	0.112	fertilizer_type	0.259
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.086	0.127	fertilizer_type	0.384
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.087	0.064	fertilizer_type	-0.303
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.087	0.083	fertilizer_type	-0.046
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.087	0.064	fertilizer_type	-0.304
Lopez-Fernandez et al 2007	3	3	1.5	0.67	organic fertilizer		0.087	0.101	fertilizer_type	0.152
Maris et al 2015	3	3	1.5	0.67			0.002	0.003	N_fertilizer_application	0.678
Maris et al 2015	3	3	1.5	0.67			0.003	0.005	N_fertilizer_application	0.463
Maris et al 2015	3	3	1.5	0.67			0.005	0.01	N_fertilizer_application	0.694
Maris et al 2015	3	3	1.5	0.67			0.001	0.003	N_fertilizer_application	0.884
Maris et al 2015	3	3	1.5	0.67		DMPP	0.003	0.002	nitrification_inhibitor	-0.39
Maris et al 2015	3	3	1.5	0.67		DMPP	0.01	0.001	nitrification_inhibitor	-2.87
Meijide et al 2007	3	3	1.5	0.67			0.037	0.122	N_fertilizer_application	1.193
Meijide et al 2007	3	3	1.5	0.67			0.037	0.106	N_fertilizer_application	1.052
Meijide et al 2007	3	3	1.5	0.67			0.037	0.083	N_fertilizer_application	0.813
Meijide et al 2007	3	3	1.5	0.67			0.037	0.183	N_fertilizer_application	1.6
Meijide et al 2007	3	3	1.5	0.67			0.037	0.262	N_fertilizer_application	1.958
Meijide et al 2007	3	3	1.5	0.67			0.037	0.143	N_fertilizer_application	1.351
Meijide et al 2007	3	3	1.5	0.67	organic fertilizer		0.143	0.122	fertilizer_type	-0.158
Meijide et al 2007	3	3	1.5	0.67		DCD	0.122	0.083	nitrification_inhibitor	-0.38
Sanchez-Martin et al 2010	3	3	1.5	0.67			0.039	0.07	N_fertilizer_application	0.583
Sanchez-Martin et al 2010	3	3	1.5	0.67			0.072	0.14	N_fertilizer_application	0.665
Ullah et al 2005	3	3	1.5	0.67			0.042	0.072	N_fertilizer_application	0.545
Ullah et al 2005	3	3	1.5	0.67			0.299	0.176	N_fertilizer_application	-0.529
Ullah et al 2005	3	3	1.5	0.67			0.166	0.702	N_fertilizer_application	1.444
Ullah et al 2005	3	3	1.5	0.67			0.292	0.443	N_fertilizer_application	0.417

Vallejo et al 2005	3	3	1.5	0.67			0.069	0.182	N_fertilizer_application	0.978
Vallejo et al 2005	3	3	1.5	0.67			0.069	0.244	N_fertilizer_application	1.268
Vallejo et al 2005	3	3	1.5	0.67			0.069	0.22	N_fertilizer_application	1.165
Vallejo et al 2005	3	3	1.5	0.67			0.182	0.244	fertilizer_incorporation	0.29
Vallejo et al 2005	3	3	1.5	0.67		DCD	0.244	0.22	nitrification_inhibitor	-0.103
Vallejo et al 2006	3	3	1.5	0.67			0.044	0.171	N_fertilizer_application	1.358
Vallejo et al 2006	3	3	1.5	0.67			0.044	0.133	N_fertilizer_application	1.104
Vallejo et al 2006	3	3	1.5	0.67			0.044	0.175	N_fertilizer_application	1.383
Vallejo et al 2006	3	3	1.5	0.67			0.044	0.123	N_fertilizer_application	1.027
Vallejo et al 2006	3	3	1.5	0.67			0.044	0.203	N_fertilizer_application	1.532
Vallejo et al 2006	3	3	1.5	0.67			0.044	0.14	N_fertilizer_application	1.16
Vallejo et al 2006	3	3	1.5	0.67		DCD	0.171	0.175	nitrification_inhibitor	0.024
Vallejo et al 2006	3	3	1.5	0.67	organic fertilizer		0.14	0.171	fertilizer_type	0.198
Vallejo et al 2006	3	3	1.5	0.67	organic fertilizer		0.14	0.133	fertilizer_type	-0.056
Zaman et al 2010	3	3	1.5	0.67		lime	0.001	0.002	amendment	0.844
Zaman et al 2010	3	3	1.5	0.67		zeolite	0.001	0.001	amendment	0.347
Zaman et al 2010	3	3	1.5	0.67		lime	0.014	0.015	amendment	0.043
Zaman et al 2010	3	3	1.5	0.67		zeolite	0.014	0.018	amendment	0.212
Zaman et al 2010	3	3	1.5	0.67		lime	0.027	0.029	amendment	0.09
Zaman et al 2010	3	3	1.5	0.67		zeolite	0.027	0.021	amendment	-0.214
Zaman et al 2010	3	3	1.5	0.67			0.001	0.014	N_fertilizer_application	2.937
Zaman et al 2010	3	3	1.5	0.67			0.001	0.027	N_fertilizer_application	3.559
Zaman et al 2008	3	3	1.5	0.67		zeolite	0.031	0.029	amendment	-0.072
Zaman et al 2008	3	3	1.5	0.67		lime	0.031	0.066	amendment	0.761
Zaman et al 2008	3	3	1.5	0.67		zeolite	0.051	0.057	amendment	0.099
Zaman et al 2008	3	3	1.5	0.67		lime	0.051	0.083	amendment	0.481
Zaman et al 2008	3	3	1.5	0.67			0.029	0.057	N_fertilizer_application	0.687
Zaman et al 2008	3	3	1.5	0.67			0.066	0.083	N_fertilizer_application	0.236
Zaman et al 2008	3	3	1.5	0.67			0.031	0.051	N_fertilizer_application	0.515
Zaman et al 2008	3	3	1.5	0.67		zeolite	0.001	0.002	amendment	0.407
Zaman et al 2008	3	3	1.5	0.67		lime	0.001	0.001	amendment	0.134
Zaman et al 2008	3	3	1.5	0.67		zeolite	0.395	0.38	amendment	-0.039
Zaman et al 2008	3	3	1.5	0.67		lime	0.395	0.637	amendment	0.478
Zaman et al 2008	3	3	1.5	0.67			0.001	0.395	N_fertilizer_application	5.755
Zaman et al 2008	3	3	1.5	0.67			0.001	0.637	N_fertilizer_application	6.1
Zaman et al 2008	3	3	1.5	0.67			0.002	0.38	N_fertilizer_application	5.309
Dambreville et al 2006	8	8	4	0.25			0.1	0.004	N_fertilizer_application	-3.227
Dambreville et al 2006	8	8	4	0.25			0.11	0.003	N_fertilizer_application	-3.742
Friedl et al 2017	4	4	2	0.5		DMPP	1.046	0.284	nitrification_inhibitor	-1.305
Friedl et al 2017	4	4	2	0.5		DMPP	1.355	0.338	nitrification_inhibitor	-1.389
Friedl et al 2017	4	4	2	0.5		DMPP	0.19	0.204	nitrification_inhibitor	0.072
Gillam et al 2008	4	4	2	0.5			0.574	0.03	N_fertilizer_application	-2.951
Gillam et al 2008	4	4	2	0.5			0.374	0.446	N_fertilizer_application	0.176
Krause et al 2017	3	3	1.5	0.67			0.024	0.055	N_fertilizer_application	0.816
Krause et al 2017	3	3	1.5	0.67			0.024	0.06	N_fertilizer_application	0.903
Li x et al 2014	3	3	1.5	0.67			0.173	0.304	fertilizer_incorporation	0.564
Lindau et al 1990	3	3	1.5	0.67	NH4NO3		0.071	0.17	fertilizer_type	0.87
Lindau et al 1990	3	3	1.5	0.67	NO3-based		0.071	0.827	fertilizer_type	2.449
Lindau et al 1990	3	3	1.5	0.67	NH4NO3		0.086	0.162	fertilizer_type	0.636
Lindau et al 1990	3	3	1.5	0.67	NO3-based		0.086	1.977	fertilizer_type	3.138
Liu xj et al 2007	3	3	1.5	0.67			1.082	1.548	tillage	0.358
Liu xj et al 2007	3	3	1.5	0.67			0.417	0.074	tillage	-1.729
Malghani et al 2018	3	3	1.5	0.67		biochar	0.029	0.054	amendment	0.633
Malghani et al 2018	3	3	1.5	0.67		biochar	0.029	0.065	amendment	0.817
Malghani et al 2018	3	3	1.5	0.67		biochar	0.029	0.013	amendment	-0.786

Malghani et al 2018	3	3	1.5	0.67		biochar	0.029	0.016	amendment	-0.603
McMillan et al 2016	3	3	1.5	0.67			0.075	0.224	N_fertilizer_application	1.097
McMillan et al 2016	3	3	1.5	0.67			0.092	0.385	N_fertilizer_application	1.432
McMillan et al 2016	3	3	1.5	0.67			0.112	0.372	N_fertilizer_application	1.203
McMillan et al 2016	3	3	1.5	0.67			0.1	0.792	N_fertilizer_application	2.072
McMillan et al 2016	3	3	1.5	0.67			0.052	1.025	N_fertilizer_application	2.989
McMillan et al 2016	3	3	1.5	0.67			0.119	1.024	N_fertilizer_application	2.151
McMillan et al 2016	3	3	1.5	0.67			0.209	0.192	N_fertilizer_application	-0.087
McMillan et al 2016	3	3	1.5	0.67			0.152	0.172	N_fertilizer_application	0.127
McMillan et al 2016	3	3	1.5	0.67			0.152	0.205	N_fertilizer_application	0.303
McMillan et al 2016	3	3	1.5	0.67			0.147	0.196	N_fertilizer_application	0.287
McMillan et al 2016	3	3	1.5	0.67			0.191	0.268	N_fertilizer_application	0.337
McMillan et al 2016	3	3	1.5	0.67			0.203	0.246	N_fertilizer_application	0.194
McMillan et al 2016	3	3	1.5	0.67		lime	0.075	0.092	amendment	0.206
McMillan et al 2016	3	3	1.5	0.67		lime	0.075	0.112	amendment	0.401
McMillan et al 2016	3	3	1.5	0.67		lime	0.224	0.385	amendment	0.541
McMillan et al 2016	3	3	1.5	0.67		lime	0.224	0.372	amendment	0.506
McMillan et al 2016	3	3	1.5	0.67		lime	0.1	0.052	amendment	-0.659
McMillan et al 2016	3	3	1.5	0.67		lime	0.1	0.119	amendment	0.179
McMillan et al 2016	3	3	1.5	0.67		lime	0.792	1.025	amendment	0.258
McMillan et al 2016	3	3	1.5	0.67		lime	0.792	1.024	amendment	0.257
McMillan et al 2016	3	3	1.5	0.67		lime	0.209	0.152	amendment	-0.32
McMillan et al 2016	3	3	1.5	0.67		lime	0.209	0.152	amendment	-0.322
McMillan et al 2016	3	3	1.5	0.67		lime	0.192	0.172	amendment	-0.106
McMillan et al 2016	3	3	1.5	0.67		lime	0.192	0.205	amendment	0.068
McMillan et al 2016	3	3	1.5	0.67		lime	0.147	0.191	amendment	0.263
McMillan et al 2016	3	3	1.5	0.67		lime	0.147	0.203	amendment	0.322
McMillan et al 2016	3	3	1.5	0.67		lime	0.196	0.268	amendment	0.313
McMillan et al 2016	3	3	1.5	0.67		lime	0.196	0.246	amendment	0.229
Merino et al 2001	4	4	2	0.5			0.039	0.091	N_fertilizer_application	0.834
Merino et al 2001	4	4	2	0.5			0.039	0.104	N_fertilizer_application	0.975
Merino et al 2001	4	4	2	0.5			0.039	0.246	N_fertilizer_application	1.833
Merino et al 2001	4	4	2	0.5			0.039	0.265	N_fertilizer_application	1.905
Merino et al 2001	4	4	2	0.5			0.039	0.128	N_fertilizer_application	1.174
Merino et al 2001	4	4	2	0.5		DCD	0.091	0.104	nitrification_inhibitor	0.141
Merino et al 2001	4	4	2	0.5		DCD	0.246	0.265	nitrification_inhibitor	0.071
Morkved et al 2006	3	3	1.5	0.67			0.023	0.057	N_fertilizer_application	0.887
Morkved et al 2006	3	3	1.5	0.67			0.087	0.046	N_fertilizer_application	-0.639
Morkved et al 2006	3	3	1.5	0.67			0.02	0.1	N_fertilizer_application	1.613
Morkved et al 2006	3	3	1.5	0.67			0.099	0.09	N_fertilizer_application	-0.097
Mukumbuta et al 2018	4	4	2	0.5		lime	0.006	0.021	amendment	1.2
Mukumbuta et al 2018	4	4	2	0.5		lime	0.006	0.089	amendment	2.665
Mukumbuta et al 2018	4	4	2	0.5			0.089	0.898	N_fertilizer_application	2.313
Mukumbuta et al 2018	4	4	2	0.5			0.303	5.722	N_fertilizer_application	2.939
Mukumbuta et al 2018	4	4	2	0.5			0.722	6.06	N_fertilizer_application	2.128
Schuster and Conrad 1992	3	3	1.5	0.67			0	0	N_fertilizer_application	1.674
Schuster and Conrad 1992	3	3	1.5	0.67			0	0	N_fertilizer_application	3.076
Schuster and Conrad 1992	3	3	1.5	0.67			0	0	N_fertilizer_application	0.894
Schuster and Conrad 1992	3	3	1.5	0.67	NH4-based		0	0	fertilizer_type	-1.402
Schuster and Conrad 1992	3	3	1.5	0.67	NO3-based		0	0	fertilizer_type	-2.182
Vallejo et al 2003	3	3	1.5	0.67			0.053	0.117	N_fertilizer_application	0.788
Vallejo et al 2003	3	3	1.5	0.67			0.053	0.224	N_fertilizer_application	1.432
Vallejo et al 2003	3	3	1.5	0.67			0.053	0.155	N_fertilizer_application	1.065
Vallejo et al 2003	3	3	1.5	0.67			0.053	0.187	N_fertilizer_application	1.254
Vallejo et al 2003	3	3	1.5	0.67	organic fertilizer		0.117	0.224	fertilizer_type	0.644

Vallejo et al 2003	3	3	1.5	0.67	organic fertilizer		0.117	0.155	fertilizer_type	0.277
Vallejo et al 2003	3	3	1.5	0.67			0.155	0.187	fertilizer_incorporation	0.189
Vallejo et al 2001	3	3	1.5	0.67			0.005	0.156	N_fertilizer_application	3.424
Vallejo et al 2001	3	3	1.5	0.67			0.005	0.134	N_fertilizer_application	3.268
Vallejo et al 2001	3	3	1.5	0.67			0.005	0.326	N_fertilizer_application	4.159
Vallejo et al 2001	3	3	1.5	0.67			0.797	1.054	N_fertilizer_application	0.28
Vallejo et al 2001	3	3	1.5	0.67			0.797	1.496	N_fertilizer_application	0.63
Vallejo et al 2001	3	3	1.5	0.67			0.797	2.475	N_fertilizer_application	1.134
Vallejo et al 2001	3	3	1.5	0.67			0.108	0.064	N_fertilizer_application	-0.527
Vallejo et al 2001	3	3	1.5	0.67			0.108	0.125	N_fertilizer_application	0.147
Vallejo et al 2001	3	3	1.5	0.67			0.108	0.129	N_fertilizer_application	0.178
Vallejo et al 2001	3	3	1.5	0.67			1.063	1.07	N_fertilizer_application	0.007
Vallejo et al 2001	3	3	1.5	0.67			1.063	0.841	N_fertilizer_application	-0.234
Vallejo et al 2001	3	3	1.5	0.67			1.063	1.427	N_fertilizer_application	0.295
Vallejo et al 2001	3	3	1.5	0.67			0.147	0.085	N_fertilizer_application	-0.548
Vallejo et al 2001	3	3	1.5	0.67			0.147	0.09	N_fertilizer_application	-0.492
Vallejo et al 2001	3	3	1.5	0.67			0.147	0.321	N_fertilizer_application	0.778
Vallejo et al 2001	3	3	1.5	0.67			0.429	0.431	N_fertilizer_application	0.004
Vallejo et al 2001	3	3	1.5	0.67			0.429	0.985	N_fertilizer_application	0.832
Vallejo et al 2001	3	3	1.5	0.67			0.429	1.321	N_fertilizer_application	1.125
Vallejo et al 2001	3	3	1.5	0.67		DCD	0.156	0.134	nitrification_inhibitor	-0.156
Vallejo et al 2001	3	3	1.5	0.67	organic fertilizer		0.156	0.326	fertilizer_type	0.734
Vallejo et al 2001	3	3	1.5	0.67		DCD	1.054	1.496	nitrification_inhibitor	0.351
Vallejo et al 2001	3	3	1.5	0.67	organic fertilizer		1.054	2.475	fertilizer_type	0.854
Vallejo et al 2001	3	3	1.5	0.67		DCD	0.064	0.125	nitrification_inhibitor	0.674
Vallejo et al 2001	3	3	1.5	0.67	organic fertilizer		0.064	0.129	fertilizer_type	0.705
Vallejo et al 2001	3	3	1.5	0.67		DCD	1.07	0.841	nitrification_inhibitor	-0.241
Vallejo et al 2001	3	3	1.5	0.67	organic fertilizer		1.07	1.427	fertilizer_type	0.288
Vallejo et al 2001	3	3	1.5	0.67		DCD	0.085	0.09	nitrification_inhibitor	0.056
Vallejo et al 2001	3	3	1.5	0.67	organic fertilizer		0.085	0.321	fertilizer_type	1.327
Vallejo et al 2001	3	3	1.5	0.67		DCD	0.431	0.985	nitrification_inhibitor	0.828
Vallejo et al 2001	3	3	1.5	0.67	organic fertilizer		0.431	1.321	fertilizer_type	1.121
Velthof et al 1997	4	4	2	0.5	NH4NO3		0.029	0.254	fertilizer_type	2.186
Velthof et al 1997	4	4	2	0.5	NO3-based		0.029	0.206	fertilizer_type	1.974
Velthof et al 1997	4	4	2	0.5	NH4-based		0.029	0.011	fertilizer_type	-0.916
Velthof et al 1997	4	4	2	0.5	NH4-based		0.029	0.003	fertilizer_type	-2.303
Velthof et al 1997	4	4	2	0.5		DCD	0.011	0.003	nitrification_inhibitor	-1.386
Wang S et al 2017	4	4	2	0.5			0.184	0.307	N_fertilizer_application	0.512
Wang S et al 2017	4	4	2	0.5			0.184	0.337	N_fertilizer_application	0.605
Wang S et al 2017	4	4	2	0.5			0.184	0.275	N_fertilizer_application	0.4
Wang S et al 2017	4	4	2	0.5			0.467	0.619	N_fertilizer_application	0.282
Wang S et al 2017	4	4	2	0.5			0.467	0.64	N_fertilizer_application	0.315
Wang S et al 2017	4	4	2	0.5			0.467	0.499	N_fertilizer_application	0.066
Wang S et al 2017	4	4	2	0.5		biochar	0.307	0.337	amendment	0.093
Wang S et al 2017	4	4	2	0.5		Nitrapyrin	0.307	0.275	nitrification_inhibitor	-0.111
Wang S et al 2017	4	4	2	0.5		biochar	0.619	0.64	amendment	0.033
Wang S et al 2017	4	4	2	0.5		Nitrapyrin	0.619	0.499	nitrification_inhibitor	-0.216
Weier et al 1993	3	3	1.5	0.67			0.032	0.092	N_fertilizer_application	1.051
Weier et al 1993	3	3	1.5	0.67			0.055	0.184	N_fertilizer_application	1.2
Weier et al 1993	3	3	1.5	0.67			0.07	4.281	N_fertilizer_application	4.114
Weier et al 1993	3	3	1.5	0.67			0.076	4.904	N_fertilizer_application	4.161
Weier et al 1993	3	3	1.5	0.67			0.055	0.164	N_fertilizer_application	1.086
Weier et al 1993	3	3	1.5	0.67			0.07	6.97	N_fertilizer_application	4.601
Weier et al 1993	3	3	1.5	0.67			0.076	5.908	N_fertilizer_application	4.348
Weier et al 1993	3	3	1.5	0.67			0.19	5.378	N_fertilizer_application	3.345

Weier et al 1993	3	3	1.5	0.67		0.184	2.723	N_fertilizer_application	2.694
Weier et al 1993	3	3	1.5	0.67		0.174	8.688	N_fertilizer_application	3.912
Weier et al 1993	3	3	1.5	0.67		0.014	0.027	N_fertilizer_application	0.676
Weier et al 1993	3	3	1.5	0.67		0.19	7.987	N_fertilizer_application	3.741
Weier et al 1993	3	3	1.5	0.67		0.184	10.176	N_fertilizer_application	4.012
Weier et al 1993	3	3	1.5	0.67		0.174	25.394	N_fertilizer_application	4.985
Weier et al 1993	3	3	1.5	0.67		1.213	7.891	N_fertilizer_application	1.873
Weier et al 1993	3	3	1.5	0.67		1.661	8.524	N_fertilizer_application	1.635
Weier et al 1993	3	3	1.5	0.67		3.948	11.82	N_fertilizer_application	1.097
Weier et al 1993	3	3	1.5	0.67		3.948	17.434	N_fertilizer_application	1.485
Zhong et al 2018	3	3	1.5	0.67		2.336	1.709	residue_retention	-0.312
Zhong et al 2018	3	3	1.5	0.67		1.832	1.559	residue_retention	-0.161
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	0.792	residue_retention	0.355
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	0.943	residue_retention	0.53
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	0.947	residue_retention	0.533
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	1.112	residue_retention	0.694
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	0.727	residue_retention	0.269
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	0.673	residue_retention	0.192
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	0.996	residue_retention	0.584
Aulakh et al 1991 residue	3	3	1.5	0.67		0.556	1.047	residue_retention	0.634
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.343	N_fertilizer_application	1.645
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.271	N_fertilizer_application	1.411
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.149	N_fertilizer_application	0.811
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.205	N_fertilizer_application	1.129
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.13	N_fertilizer_application	0.679
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.124	N_fertilizer_application	0.628
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.08	N_fertilizer_application	0.185
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.04	N_fertilizer_application	-0.51
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.068	N_fertilizer_application	0.023
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.094	N_fertilizer_application	0.357
Mulvaney et al 1997	3	3	1.5	0.67		0.066	0.089	N_fertilizer_application	0.293
Mulvaney et al 1997	3	3	1.5	0.67	NH4-based	0.271	0.343	fertilizer_type	0.235
Mulvaney et al 1997	3	3	1.5	0.67	NH4-based	0.271	0.205	fertilizer_type	-0.281
Mulvaney et al 1997	3	3	1.5	0.67	NH4-based	0.271	0.124	fertilizer_type	-0.782
Mulvaney et al 1997	3	3	1.5	0.67	NH4NO3	0.271	0.04	fertilizer_type	-1.921
Mulvaney et al 1997	3	3	1.5	0.67	NH4-based	0.271	0.094	fertilizer_type	-1.054
Mulvaney et al 1997	3	3	1.5	0.67	NH4-based	0.149	0.13	fertilizer_type	-0.132
Mulvaney et al 1997	3	3	1.5	0.67	NH4-based	0.149	0.08	fertilizer_type	-0.627
Mulvaney et al 1997	3	3	1.5	0.67	NH4NO3	0.149	0.068	fertilizer_type	-0.788
Mulvaney et al 1997	3	3	1.5	0.67	NH4-based	0.149	0.089	fertilizer_type	-0.518
Horgan et al 2002	3	3	1.5	0.67	NO3-based	0.198	0.141	fertilizer_type	-0.339

CHAPTER 6

N_2O production pathways in forest soils: coupling Australian case study and global literature data

This chapter is based on an accepted manuscript entitled:

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N_2O production pathways in forest soils: coupling Australian case study and global literature data

Abstract

Forest soil is the largest source of nitrous oxide (N_2O) emissions from terrestrial ecosystems and the contribution of N_2O production by different pathways varies widely. It is necessary to evaluate the contribution of N_2O production pathways and its driving factors in forest soils and thus to improve the simulation performance and investigate the boundary conditions of N_2O production pathways. However, the patterns of N_2O production pathways at the global scale and in Australian forest soils remains uncertain. In this study, by conducting a ^{15}N tracing experiment, we found that based on 13 forest soils sampled within Australia from subtropical, temperate and arid regions, forest soils in temperate areas had the highest N_2O emission rate (19.5 μg N kg^{-1} soil d^{-1}), followed by subtropical and arid soils (3.84 and 3.80 μg N kg^{-1} soil d^{-1}, respectively). Heterotrophic nitrification dominated N_2O production in Australian forest soils; its contribution followed the order of arid (78%) > subtropical (69%) > temperate (59%). N_2O from heterotrophic nitrification was negatively related to mean annual temperature (MAT) and the contribution of heterotrophic nitrification to N_2O production was negatively related to soil total nitrogen (N) and total carbon (C) content. These results partially agreed with the global literature data synthesis, which showed that in addition to heterotrophic nitrification (42%), denitrification (43.5%) was a main pathway of N_2O production in global forest soils. Globally, we found soil pH, moisture content, total nitrogen content, total carbon content and MAT attributed to the contribution of heterotrophic nitrification and denitrification to N_2O production. Our findings improve the understanding of N_2O production pathways in forest soils and have implications for the simulation and projection of global N_2O production from forest soils.

1. Introduction

Nitrous oxide (N_2O) is a potent greenhouse gas with a global warming potential 298 times that of carbon dioxide (CO_2) over a 100-year timeframe (WMO, 2013), contributing to global warming and stratospheric ozone depletion (Ravishankara *et al.*, 2009). The global N_2O emissions from terrestrial ecosystems were estimated to be 12.5−13.3 Tg N yr^{-1} (Xu *et al.*, 2008; Tian *et al.*, 2013). Tian *et al.* (2013) estimated that forest soil accounted for the largest emissions of N_2O (4.3 Tg N yr^{-1}) from 1981 to 2020. Autotrophic nitrification, heterotrophic nitrification and denitrification are the main N_2O production pathways in soils (Eylar and Schmidt, 1959; Barnard *et al.*, 2005). Previous studies have shown that N_2O emissions from croplands and grassland soils are mainly governed by autotrophic nitrification and denitrification (Tortoso and Hutchinson, 1990; Li *et al.*, 2016), which are regulated by soil properties. For example, soil C:N ratio could influence gross nitrate (NO_3^-) production rates by altering the AOA/AOB ratio and subsequently change the proportion of N_2O production from autotrophic nitrification (Duan *et al.*, 2019b). More available soil organic carbon (SOC) provided more energy source and increased soil O_2 consumption, which could promote denitrification and associated N_2O emission, thus lowered N_2O/N_2 ratio (Zhang *et al.*, 2018). Soil pH was also a dominant factor that regulated N_2O production pathways. Autotrophic nitrifiers were inhibited by low pH, so autotrophic nitrification and associated N_2O production would be low in acidic soils (Cheng *et al.*, 2015). Moreover, decreasing soil pH could inhibit N_2O reductase and increase the $N_2O/(N_2+N_2O)$ ratio from denitrification, resulting in an increased contribution of denitrification derived N_2O production (Cheng *et al.*, 2015). Generally, the optimum temperature for soil nitrification is around 25 to 35°C, and the optimum temperature of denitrification is higher because denitrifies may adapt to higher soil temperature (Duan *et al.*, 2019a). Maag and Vinther (1996) showed that the proportion

of N_2O from nitrification decreased with increasing temperature while the denitrification activity positively responded to increased temperature. Soil moisture is considered as an important factor regulating N_2O production pathways (Maag and Vinther, 1996), higher soil moisture content (55-85% of water filled pore space) could create an anaerobic condition, leading to higher denitrification derived N_2O production (Liu *et al.*, 2017). However, these studies were mainly focus on site-specific characteristics in agroecosystems, and few studies have been conducted in forest ecosystems.

In contrast to croplands or grassland soils, previous studies demonstrated that heterotrophic nitrification was the primary production way of N_2O emissions in acidic Chinese forest soils (Zhang *et al.*, 2011; Zhang *et al.*, 2018). High organic carbon content with high C:N ratio and low soil pH in those soils provide a favourable microsite for heterotrophic nitrifiers (Zhang *et al.*, 2015). The contribution of N_2O production from different pathways varies widely in forest ecosystems. Some studies demonstrated that denitrification was the main source of N_2O in spruce and beech forest in Norway (Ambus, 1998) and Amazon forest soils (Perez *et al.*, 2006), where soils were acidic with limited available NH_4^+. Besides, only scattered studies were conducted in Australia (Breuer *et al.*, 2000; Kiese and Butterbach-Bahl, 2002), i.e., the magnitude and dynamic changes of N_2O emissions on Australian subtropical and tropical rainforests. The lack of comprehensive studies leads to high uncertainty in national N_2O inventory estimation.We conducted a large-scale field sampling in subtropical, temperate and arid regions within Central and Eastern Australia, followed by a laboratory ^{15}N tracing experiment and combined with a global literature search for N_2O production pathways in forest soils, to 1) evaluate the contribution of N_2O production pathways of Australian forest soils in subtropical, temperate and arid regions; 2) to quantify the N_2O production pathways from forest soils

globally; and 3) to investigate the relationships between N_2O production pathways in forest soils and driving factors.

2. Materials and methods

2.1 Soil samples

Thirteen forest (dominated by Eucalyptus species) soils were sampled across Eastern coastal areas and inland areas of central Australia in May 2019. These soils were located in arid (n= 6), humid subtropical (n= 3) and temperate (n= 4) climates (Figure 1). At each site, topsoil (0-10cm) was collected, homogenized thoroughly, and transported on ice to the laboratory. The soils were oven-dried at 40°C after removing roots and leaf residues. These soils were then ground and sieved through a 2.0mm mesh. For each site, the mean annual precipitation (MAP [mm]) and mean annual temperature (MAT [°C]) of each sampling location were extracted from the WorldClim database version 2 (http://www.worldclim.org/) using the '*raster*' and '*sp*' packages in R (Pebesma and Bivand, 2005; Hijmans *et al.*, 2019). Soil pH was measured by a 1: 2.5 (*w:w*) of soil and water ratio with a pH meter (Thermo Scientific Inc., Waltham MA, US). NH_4^+-N and NO_3^--N were analysed with 2 M KCl solution with a segmented flow analyser (Skalar Analytical B. V. Tinstraat, Breda, Netherland). Total nitrogen [TN (%)] and total carbon [TC (%)] were determined by an isotope ratio mass spectrometer (Sercon Hydra, Crewe, UK).

2.2 ^{15}N tracing experiment

Soil microcosms were established in 250ml flasks containing 20g of soil (oven-dry equivalent). Soils were pre-incubated at 25°C for three weeks to stabilize the soil microbial communities. Soil moisture contents were adjusted to 50% WFPS during pre-incubation. After pre-incubation, two labelled treatments with three replicates, i.e., $NH_4{}^{15}NO_3$ (9.7537 atom% excess) and $^{15}NH_4NO_3$ (9.8837 atom% excess) were applied at

60 μg N g^{-1} dry soil (comprising 30 μg NH_4^+-N g^{-1} soil and 30 μg NO_3^--N g^{-1} soil). Soil moisture content in the flasks was adjusted to 55% WFPS with N solution added. Soils were incubated in the dark for 6 days at 25 °C.

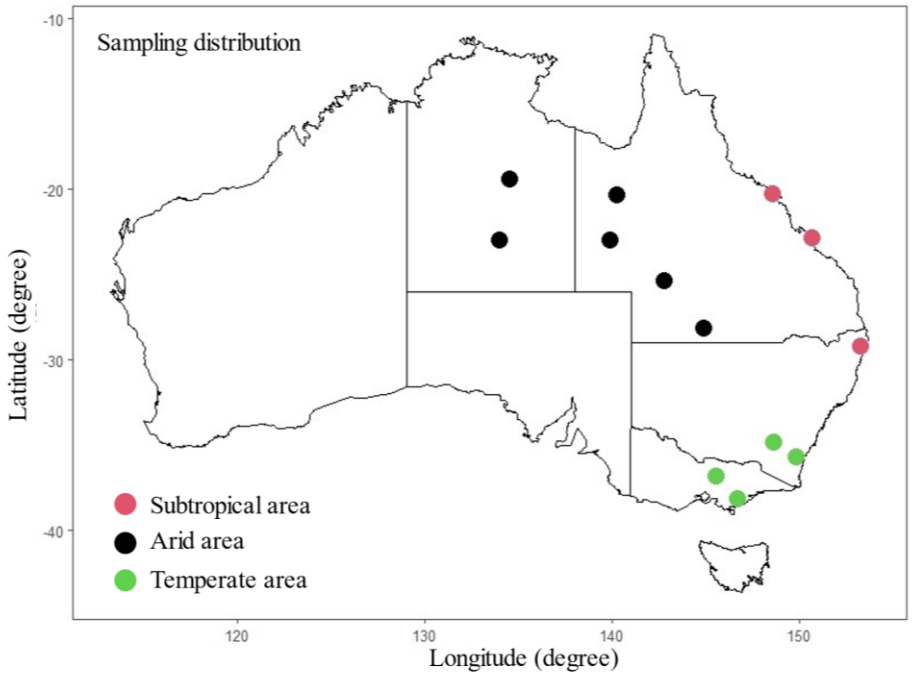


Figure 1. The location of soils collected for the incubation study

2.3 Laboratory analyses

Headspace gas samples were collected at days 0, 2 and 6 after N application. On each gas sampling day, the flasks were opened and flushed with ambient air and resealed for 6 h before sampling. Gas samples were collected from each flask and injected into two pre-evacuated vials (12 ml) to determine the N_2O concentration by gas chromatograph (7890A; Agilent Technologies, Santa Clara, USA). A separate set of gas samples was injected into a 60 ml evacuated vial for $^{15}N_2O$ analysis by isotope ratio mass spectrometer (IRMS) (Sercon Hydra, Crewe, UK). Soils were destructively sampled for isotopic composition analysis after gas sampling. Soils were extracted by 2 M KCl solution to determine NH_4^+-N and NO_3^--N content with a segmented flow analyzer (Skalar

Analytical B. V. Tinstraat, Netherlands) and isotopic composition using a micro-diffusion method as described by Saghir *et al.* (1993) followed by IRMS. After extraction, soils were washed using distilled water, and oven-dried at 60 ^{o}C and the isotopic composition of the insoluble organic N was determined by IRMS. After gas sampling and NH_4^+-N and NO_3^--N extraction, soil in each flask was washed with distilled water and then oven-dried at 60 ^{o}C to determine the isotopic composition of the insoluble organic N by IRMS.

2.4 Data analysis and calculations

Autotrophic nitrification, heterotrophic nitrification and denitrification were targeted in this study as the pathways of N_2O production. Three pools were considered as possible substrates for N_2O, which differed in their ^{15}N atom fraction, viz. a_n (an autotrophic nitrification pool, assumed to be equivalent to NH_4^+-N), a_h (a heterotrophic nitrification pool, assumed to be equivalent to insoluble organic N), a_d (a denitrification pool, assumed to be equivalent to NO_3^--N) (Stevens *et al.*, 1997; Rütting *et al.*, 2010). The ^{15}N atom fraction of the resulting N_2O can be determined according to Rütting *et al.* (2010)

$$a_{N2O} = n_d a_d + n_a a_a + n_h a_h \quad (1)$$

$$n_d + n_a + n_h = 1 \quad (2)$$

Where, a_{N2O} is the ^{15}N abundance of total N_2O produced during the incubation; n_d, n_a and n_h are N_2O emission fractions originating from the NO_3^-, NH_4^+ and insoluble organic N pool via denitrification, autotrophic nitrification and heterotrophic nitrification, respectively; a_d, a_a and a_h represent the ^{15}N abundance of NO_3^-, $NH_4^+$and insoluble organic N, respectively. The value of ^{15}N atom% excess from NO_3^-, NH_4^+ and insoluble organic N in the paired $^{15}NH_4NO_3$ and $NH_4{}^{15}NO_3$ treatments were put into equations (1) and (2), which enabled derivation of n_d, n_a, and n_h, respectively.

N_2O production rates originating from the NO_3^- pool (denitrification), the NH_4^+ pool (autotrophic nitrification) and the insoluble organic N pool (heterotrophic nitrification) were calculated using equations (3) to (5) as follows:

$$N_2O_d = n_d \times N_2O_t \quad (3)$$

$$N_2O_a = n_a \times N_2O_t \quad (4)$$

$$N_2O_h = n_h \times N_2O_t \quad (5)$$

Where N_2O_t is the total N_2O emission rate from incubated soil.

One-way ANOVA was used to compare the N_2O production rates and the contribution of N_2O from different pathways among different soils. Pearson correlation analyses were performed to determine the relationships between N_2O production pathways (rates and contribution) and soil/environmental properties. All analyses were conducted by SPSS (Version 25, SPSS Inc., Chicago, IL, USA).

2.5 Global literature data collection

To investigate the global pattern of N_2O production pathways, we extracted data of relative contribution of each pathway to N_2O production in forest soils from published peer-reviewed papers. Data extracted from literature should be met the following criteria: 1) the study should be conducted in forest soils. 2) the relative contribution of nitrification and denitrification to N_2O production should be reported. We then compiled a database with 100 observations from 17 studies (including this study). The geographic coordinates (latitude and longitude), MAT, MAP, soil pH, soil total nitrogen (TN [%]), soil total carbon (TC [%]), soil C:N ratio, NH_4^+ and NO_3^- concentrations (mg kg^{-1} dry soil) were extracted.

3. Results

3.1. N_2O production in Australian forest soils

According to the climatic zones, 13 soils can be categorized as arid, temperate, and subtropical soils (Figure 1). The MAT ranged from 13.3 to 25.7 °C and the MAP ranged widely from 272 to 1407mm (Table 2). Soils were characterized by neutral to slightly acidic, (pH value: 5.2 - 6.8). Soils located in temperate areas had the highest TN and TC content (0.27% and 3.91%, respectively) while Arid soils had the lowest TN and TC (0.11% and 1.38%, respectively). NH_4^+ was highest in temperate soils (14 mg kg^{-1} dry soil) and followed by subtropical (12.6 mg kg^{-1} dry soil) and arid (6.5 mg kg^{-1} dry soil) soils. NO_3^- content were ranged from 36.5 to 42.3 mg kg^{-1} dry soil across different soils.

Table 1. Soil properties of sampling sites in Australia

Climatic regions	n	MAT[§] (°C)	MAP[#] (mm)	pH	TN (%)	TC (%)	C: N ratio	NH_4^+ (mg kg^{-1} dry soil)	NO_3^- (mg kg^{-1} dry soil)
Arid	6	23.4± 0.9	356±34	6.8± 0.3	0.11± 0.05	1.38± 0.69	12.0± 0.8	6.5±1.0	37.5±3.5
Temperate	4	13.8± 0.2	773+61	5.2± 0.3	0.27± 0.13	3.91± 1.86	14.8± 0.8	14.0±3.2	36.5±5.2
Subtropical	3	21.6± 1.2	1275± 75	5.6± 0.5	0.23± 0.07	3.50± 0.78	16.1± 1.3	12.6±4.6	42.3±20.5
p		<0.01	<0.01	0.02	0.40	0.20	0.03	0.11	0.90

[§]MAT: mean annual temperature [#]MAP: mean annual precipitation. Values in the parentheses are standard errors. ANOVA was used to identify significant differences of soils among climatic zones and reported as significant where $p<0.05$.

During 6-day incubation, soils in temperate areas showed the highest rate of N_2O production with 19.5 µg N Kg^{-1} soil d^{-1}, which is significantly greater than the N_2O production in Arid soils and subtropical soils (3.8 and 3.84 µg N Kg^{-1} soil d^{-1}, respectively) (Figure 2a). N_2O production from heterotrophic nitrification was 2.63, 2.47 and 9.5 µg N

Kg^{-1} soil d^{-1} for arid, subtropical, and temperate soils, respectively. Moreover, heterotrophic nitrification accounted for the greatest proportion to total N_2O production in all soils. The contribution of N_2O from heterotrophic nitrification followed the order of arid (78%) > subtropical (69%) > temperate (59%). On the contrary, autotrophic nitrification contributed a smaller proportion to N_2O emission in all soils. The contribution of autotrophic nitrification to N_2O production followed the order of temperate soils (29%) > subtropical soils (20%) > arid soils (11%). N_2O production from denitrification remained the lowest in all soils (11-13%) (Figure 2b).

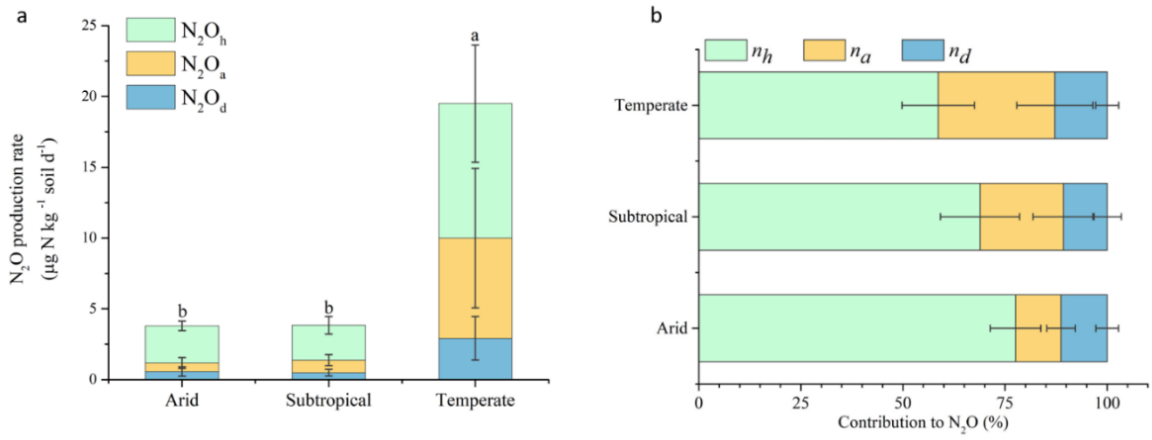


Figure 2. N_2O production rates a) and pathways b) in Australian forest soils from different climatic zones. N_2O_h, N_2O_a and N_2O_d are N_2O emission rates via heterotrophic nitrification, autotrophic nitrification and denitrification, respectively. n_h, n_a and n_d are the contribution of heterotrophic nitrification, autotrophic nitrification and denitrification to N_2O production, respectively. Different letters in Figure 2a on the top indicate that the total values are significantly different at $p < 0.05$.

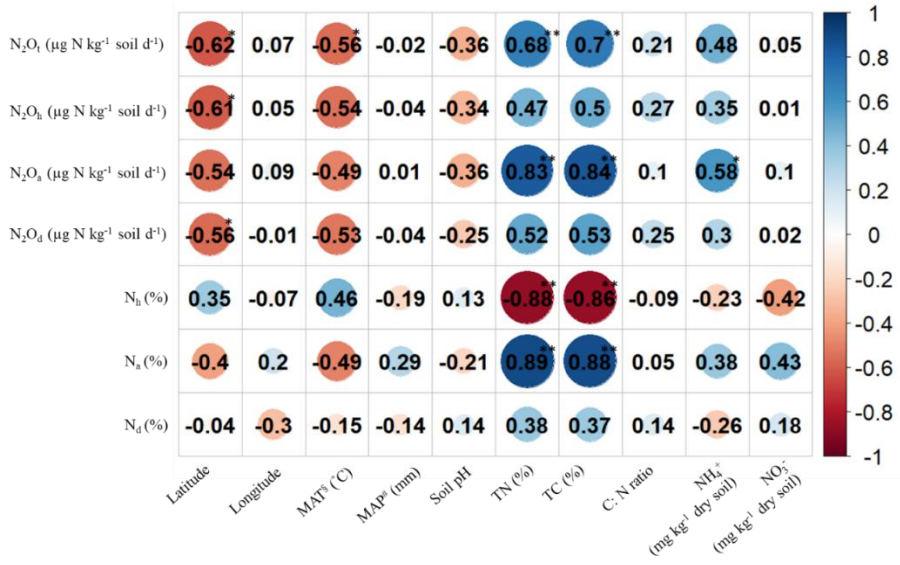


Figure 3. Correlation analysis of N_2O production rate and contribution with climatic variables and soil properties. * $p < 0.05$ ** $p < 0.01$.

Total N_2O production was positively correlated with soil TN and TC ($p<0.01$). N_2O from autotrophic nitrification was positively correlated with soil TN, TC and NH_4^+ content ($p< 0.01$, 0.01 and 0.05, respectively) (Figure 3). The contribution of heterotrophic nitrification to N_2O production was negatively correlated with soil properties, e.g., soil TN and TC content ($p<0.01$, Figure 3). Total N_2O production, N_2O from nitrification and denitrification were negatively correlated with the latitude significantly ($p<0.05$, Figure 3). Total N_2O production and N_2O from nitrification decreased with lower MAT (Figure 3b, $p<0.05$).

3.2. N_2O production in forest soils from literature-based database

We extracted 100 observations of relative contribution of each pathway to N_2O production in forest soils from 17 previous published peer-reviewed papers. These studies were conducted mainly in European, Chinese forest soils. These forest soils mostly located in relatively humid areas with an average MAP of 1130 mm and MAT of around 12 ^oC. Most soils are acidic soils with an average soil pH of 4.7 (3.5-7). Soil TN and TC

ranged from 0.02 to 1.1% and 0.33 to 14.1%, respectively. Soil C:N ratio was up to 36. Soil NO_3^- and NH_4^+ contents had large variances with averages of 12.5 mg kg^{-1} dry soil and 18.3 mg kg^{-1} dry soil, respectively. Soil WFPS ranged from 16 to 96% (Table 2).

Across the global database, we found that heterotrophic nitrification and denitrification are the main pathways to N_2O production (42% and 43.5%, respectively) (Figure 4). The contribution of N_2O from heterotrophic nitrification followed the order of subtropical soils (45%)> temperate soils (44%) > continental soils (33%). The contribution of denitrification to N_2O emission was highest in temperate areas (47%) and followed by continental and subtropical areas (46% and 41%, respectively). N_2O emission from autotrophic nitrification was the lowest in all soils, accounted for 14% (subtropical), 10% (temperate) and 20.5% (continental) of total N_2O production.

Table 2. Soil properties from literature-based data

	n	Minimum	Maximum	Mean	Std. Deviation
MAP[#] (mm)	99	500	3500	1130	605
MAT[*] (^oC)	99	0.5	27	12	8
pH	91	3.5	7	4.7	0.9
SOC (%)	55	0.1	23.3	4.06	4.8
TN (%)	89	0.02	1.4	0.26	0.22
TC (%)	38	1.2	14.1	4.71	3.3
C:N ratio	70	8.4	35.9	16.4	5.1
NO_3^- (mg kg^{-1} dry soil)	86	0.11	93.0	12.89	16.72
NH_4^+ (mg kg^{-1} dry soil)	68	0	283.3	14.5	37.0
WFPS (%)	95	16	96	51	15.3

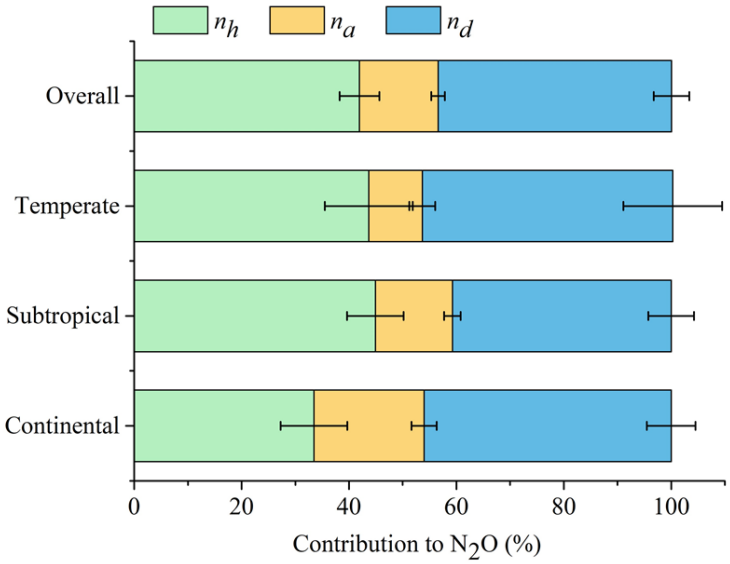


Figure 4. The N_2O production pathways from forest soils from global studies (n=39), n_h, n_a and n_d are the contribution of heterotrophic nitrification, autotrophic nitrification and denitrification to N_2O production, respectively.

Table 3 Spearman's correlation analysis of the contribution of N_2O production with climatic variables and soil properties globally. * $p < 0.05$ ** $p < 0.01$.

	Latitude	Longitude	MAP (mm)	MAT (^oC)	Soil pH	TN (%)	TC (%)	C: N	NO_3^- (mg kg^{-1} dry soil)	NH_4^+ (mg kg^{-1} dry soil)	WFPS
n_h	**-0.38****	**0.38****	-0.12	**0.30***	**0.42****	-0.25	**-0.73****	-0.09	-0.04	**0.45****	0.10
N	52	52	52	52	52	50	13	38	42	42	52
n_a	-0.06	0.23	-0.07	-0.07	-0.02	**0.48****	**0.78****	0.19	0.13	-0.24	-0.18
N	52	52	52	52	52	50	13	38	42	42	52
n_d	**0.30****	**-0.34****	0.05	**-0.19***	**-0.26****	**0.33****	**0.50****	**0.23***	-0.07	**-0.28***	**0.30****
N	112	112	112	112	104	102	51	83	99	81	108

The contribution of heterotrophic nitrification to N_2O production was negatively correlated with soil TC content ($p<0.01$, Table 3) and latitude while it was positively correlated with longitude, MAT, soil pH and soil NH_4^+ content. The contribution of autotrophic nitrification to N_2O production was correlated with soil TC and TN significantly ($p<0.01$, Table 3). The contribution of denitrificaiton to N_2O production was not only correlated with long term environmetal factor, i.e., MAT, but also correlated with soil properties such as soil pH, soil TN, TC, WFPS and so on (Table 3, $p<0.01$).

4. Discussion

We found that N_2O production from temperate soils (19.5 μg N kg^{-1} soil d^{-1}) was significantly greater than that from arid and subtropical soils (3.80 and 3.84 μg N kg^{-1} soil d^{-1}, respectively) (Figure 2a). In contrast, previous studies suggest that tropical and subtropical forest soils are major sources of N_2O (Breuer *et al.*, 2000; Zhang *et al.*, 2011), attributed to their higher organic carbon and nitrogen contents than the soils in temperate regions (Breuer *et al.*, 2000; Zhuang *et al.*, 2012). We observed a significant and positive relationship between N_2O production and soil TC and TN contents (Figure 3). This suggests that soil C and N availability might drive the variation in N_2O production and its pathways in Australian forest soils across climatic regions (Figure 2).

Heterotrophic nitrification was the dominant pathway of N_2O production regardless of climatic zones or soil properties (Figure 2). N_2O production from heterotrophic nitrification was 2.63, 2.47 and 9.5 μg N kg^{-1} soil d^{-1} for arid, subtropical and temperate soils, respectively. Its contribution followed the order of arid (78%) > subtropical (69%) > temperate (59%). This finding agrees with previous studies in global databases, which showed that heterotrophic nitrification contributed to 42% of total N_2O production in forest soils (Figure 4). The dominant role of heterotrophic nitrification in N_2O production in acidic soils was attributed to the stronger adaptability of heterotrophic nitrifiers than

autotrophic nitrifiers under acidic conditions regardless (Braker and Conrad, 2011). Heterotrophic microbes were more competitive for ammonium than autotrophic nitrifiers which were inhibited by a low soil pH, leading to a higher heterotrophic nitrification rate and subsequent N_2O emission (Verhagen and Laanbroek, 1991).

On the contrary, autotrophic nitrification contributed a small proportion to N_2O emissions in forest soils in both Australia and other regions worldwide (Figure 2b and 4). One reason is low soil pH in forest soils may inhibit the autotrophic bacterial activity (Table 1 and 2). When pH <6.5, the lack of ammonia as the substrate for ammonia monooxygenase limits the growth of autotrophic nitrifiers (Li *et al.*, 2018). Higher contribution of autotrophic nitrification to N_2O production in temperate soils (29%) than arid and subtropical soils (11% and 20%, respectively) was observed in Australian forest soils, likely owing to higher soil TN and NH_4^+ content in temperate soils. NH_4^+ is the substrate and the main regulator of autotrophic nitrifiers in forest ecosystems (Ste-Marie and Paré, 1999) and soil TN can also positively affect the activity of nitrifying organisms (Sahrawat, 2008).

Besides, denitrification was another main pathway of N_2O globally (Figure 4). The contribution of denitrification to N_2O production would increase when denitrification is stimulated under high soil moisture content (Bateman and Baggs, 2005) and the greater nitrogen and carbon availability via litter input (Barton *et al.*, 1999) (Table 3). However, denitrification was a minor pathway of N_2O production (11-13%) in Australian forest soils (Figure 2b); its contribution to N_2O production was much lower than that observed in forest soils in tropical and subtropical areas (53%-88%) with high annual rainfall (>1200mm) (Vanitchung *et al.*, 2011; Han *et al.*, 2018). Soils in temperate and arid areas in Australian soils are probably well-aerated with limited water supply thus denitrification is generally very low (<1 kg N ha^{-1} $year^{-1}$).

Soil TC and TN are important to N_2O production pathways as well. Our analysis demonstrated positive relationships between soil TC and TN with the contribution of autotrophic nitrification and denitrification in both Australian and global forest soils (Figure 3, Table 3). However, the relationships between the contribution of heterotrophic nitrification to N_2O production with TN and TC were negative (Figure 3, Table 3). This pattern may imply that with the increase of soil TN and TC, N_2O production from different pathways all increased simultaneously. With lower soils TN and TC contents, denitrification and autotrophic nitrification could be inhibited by limited substrates, while heterotrophic nitrification could be carried out by a wide range of microorganisms, e.g., fungi (Zhang *et al.*, 2011). With increased TN and TC contents, the increase magnitudes of denitrification and autotrophic nitrification were much higher than that of heterotrophic nitrification, which caused the contribution of heterotrophic nitrification to N_2O production decreased.

We also found that N_2O production pathways varied with latitudes (i.e., MAT and MAP). MAP and MAT affect the long-term soil properties and subsequently N_2O production pathways. A negative relationship between MAT and N_2O emission from nitrification was observed in Australian forest soils (Figure 3). This indicated that nitrification was sensitive to high temperature and the adaptation of soil nitrifiers to climate (Malhi and McGill, 1982). Moreover, according to Maag and Vinther (1996), N_2O emission from nitrification has been reported to increase with decreased temperature since nitrite tends to accumulate at low temperatures (Russell *et al.*, 2002). Our results extended the climatic dependence of temperature relationships in N_2O from nitrification to warm arid and cool temperate regions. This finding was in contrast to a previous study in forest soils, which demonstrated that N_2O from nitrification increased with temperature from 15 ^oC to 35 ^oC (Focht, 1974).

5. Conclusion

We examined the magnitude and geographical pattern of N_2O production pathways in forest soil in both Australia and worldwide. By the incubation experiment using ^{15}N tracing technique, we found that in Australia, temperate soils had the highest N_2O production rate (19.5 μg N kg^{-1} soil d^{-1}), followed by subtropical and arid soils (3.84 and 3.80 μg N kg^{-1} soil d^{-1}, respectively). Heterotrophic nitrification was a main pathway of the N_2O production in forest soils either in Australian forest soils or other soils worldwide, accounting for 33-78% of total N_2O emission. Denitrification was another important N_2O production pathway in forest soils other than Australian soils, contributing 41-47% of total N_2O emission. N_2O production and its pathways in forest soils were highly related to soil and environmental variances, such as MAT, soil TN and TC. Our results showed that temperate forest soils may have high potential of N_2O emission especially that produced via heterotrophic nitrification. This work provided fundamental results to help with future simulation and projection of global N_2O production in forest soils.

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CHAPTER 7

Spatiotemporal trend of nitrate leaching in global agroecosystems: a machine learning model evaluation

This chapter is based on a ready submitted manuscript entitled:

Pan, B., Xia, L., Lam, S. K., Liang, X., & Chen, D. Modelling the spatiotemporal trend of nitrate leaching in agroecosystems. Ready to submit.

Modelling the spatiotemporal trend of nitrate leaching in global agroecosystems: a machine learning approach

Abstract

Nitrate (NO_3-N) leaching reduces N use efficiency, causes eutrophication and contaminates groundwater. However, measurement and simulation of NO_3-N leaching in agroecosystems remain challenging. Here we integrated data mining and machine learning models to predict NO_3-N leaching in global agroecosystems. By compiling literature-based database of NO_3-N leaching from field experiments worldwide, we developed a machine learning model (NL_{NO3} model) to reliably predict NO_3-N leaching (R^2=0.75) in 1961, 2000 and 2010 using a few easily accessible input variables. We found that the total NO_3-N leaching in agroecosystems increased from 23.2 Tg N yr^{-1} in 1961 to 32.8 Tg N yr^{-1} in 2000 and 39.7 Tg N yr^{-1} in 2010. Hotspots of NO_3-N leaching in global agroecosystems expanded from Europe in 1961 to China, South Asia and Brazil in 2000 and 2010. The highly spatiotemporal diversity of NO_3-N leaching was mainly driven by soil properties (e.g., soil TN, soil pH, soil texture), aridity index and farming practices (e.g., N fertilization and irrigation). This study provided a novel approach to estimate the spatiotemporal pattern of NO_3-N leaching worldwide and the NL_{NO3} model could be incorporated into process-based models to simulate NO_3-N leaching with easily accessible input variables.

1. Introduction

Nitrogen (N) is an essential nutrient for agricultural production. Synthetic N fertilizer use has increased rapidly since 1960s to meet the growing demand for food, feed and fibre (Erisman *et al.*, 2008). However, reactive N (Nr: all species of N except dinitrogen N (N_2)) losses to the atmosphere and waterbodies from excessive N fertilization in agricultural systems has led to ecosystem degradation, air and water pollution, human health damage

and biodiversity loss (Erisman *et al.*, 2013). Nitrate (NO_3-N) leaching from the root zone to groundwater is one of the most important Nr loss pathways which not only reduces N use efficiency (NUE) in agroecosystems (Di and Cameron, 2002), but also leads to groundwater contamination and eutrophication (Robertson and Vitousek, 2009).

Quantifying NO_3-N leaching is critical to understanding N cycling and achieving sustainable agriculture. However, the simulation and measurements of NO_3-N leaching are still challenging (van der Laan *et al.*, 2014). From top-down estimation, IPCC adopted a default value of 0.3 to represent the N fertilization induced NO_3-N leaching in agricultural systems (IPCC, 2006) while about 16-17% of applied N fertilizers was estimated to be lost as N leaching in cropland by a global N flow model and a combined isotope-based model with global mass-balance model (Liu *et al.*, 2010; Wang *et al.*, 2017). However, top-down estimation cannot capture the spatiotemporal heterogeneity of NO_3-N leaching as NO_3-N leaching is highly related to soil properties, environmental conditions and farming practices (Wang *et al.*, 2019).

Many process-based models have also embedded submodules to simulate N leaching, such as APSIM-SWIM (Keating *et al.*, 2003), WNMM (Li *et al.*, 2007), RZWQM (Ma *et al.*, 1998), GLEAMS (Leonard *et al.*, 1987), LEACHM (Hutson, 2003), HYDRUS (Simunek *et al.*, 2008), DNDC (Li *et al.*, 2006), etc. However, these models require detailed initialization, parameterization, as well as complex soil parameters and hydrodynamic coefficients to simulate N leaching, e.g., for Richard's equation and convection-dispersion equation, which are difficult to obtain. Moreover, the uncertainties related to model settings and lack of representation of the key processes in N dynamics may also limit the applicability of these process-based models (van der Laan *et al.*, 2014). In addition, some empirical and simpler models were developed to estimate NO_3-N leaching in specific agricultural systems. For example, Di and Cameron (2000) developed

a NLE model to estimate N leaching in dairy pasture systems, whereas Simmelsgaard and Djurhuus (1998) used an empirical model to estimate NO_3-N leaching in Danish crop systems. These empirical models adopted a series of equations to reflect the relationships between NO_3-N leaching and influencing factors derived from site-specific studies. However, the capacities of these empirical models are limited to a certain scale/system and cannot be applied in a wide range of soil and environmental conditions. Moreover, these models cannot capture the complex interactions between input variables, which could restraint the model performance.

To address the limitations of current top-down estimation, process-based models and empirical models, we adopted a machine-learning based stochastic gradient boosting (SGB) model to identify the spatiotemporal patterns of NO_3-N leaching and improve the prediction of N dynamics across diverse global agroecosystems. SGB is a tree-based machine model; each tree is built in a sequential error-correcting process to converge to an accurate model. SGB model was proposed and modified based on the gradient boosting decision tree (Friedman, 2001, 2002) with the modification of sampling algorithms. Rather than boosting all the sample data at each iteration, a SGB model adopts random subsampling, i.e., random selection of subsample from training data at each iteration, to reduce the computational complexity and improve the model learning speed. It has been proven to be a powerful tool to handle both numerical and categorical variables (Friedman, 2001, 2002). A SGB model is especially effective when quantifying nonlinear N transformations that are regulated by variables with complicated interactions, e.g., N_2O emission from nitrification (Pan *et al.*, 2021) and gross nitrification and immobilization rate (Zhang *et al.*, 2021).

In this study, by compiling a global database of NO_3-N leaching studies conducted under field conditions, we aimed to 1) develop a reliable machine learning model of NO_3-N

leaching in global agroecosystems; 2) investigate the global spatiotemporal trend of NO_3-N leaching in 1961, 2000 and 2010; 3) identify the hotspots of NO_3-N leaching worldwide. This study provided a novel approach to quantify NO_3-N leaching worldwide and the NL_{NO3} model could be incorporated into process-based models for better simulation of NO_3-N leaching at various scales.

2. Methods and materials

2.1 Database compilation

We conducted literature search from Web of Science (ISI) and Google Scholar using keywords including nitrate/ NO_3^- leaching, N application, mitigation strategy, management practices, fertilizer type, soil amendment, and inhibitor. Studies included in our database fulfilled the following selection criteria: a) studies were conducted under field conditions; b) experimental design was clearly presented; c) studies were restricted to agroecosystems. A total of 1818 observations (1319 from cropland and 499 from grassland) from 286 studies were included. In the database, we extracted geographic coordinates (latitude [LATI] and longitude [LONG]) of the study sites, environmental conditions (mean annual temperature [MAT] and precipitation [MAP], potential evapotranspiration rate [PET] and aridity index (the ratio of precipitation to evapotranspiration) [AI]), soil texture classified as coarse, medium, fine and very fine based on Soil and Terrain Digital Database (SOTER) convention (S_TEX) (Van Engelen and Dijkshoorn, 2013), soil pH (S_PH), soil total N (S_TN), soil organic carbon (SOC) content, soil C/N ratio, initial NH_4^+ and NO_3^- concentrations, N application rate (N_IN), irrigation rate (IR) and land use system (LUS). The NO_3-N leaching data were extracted over the whole year. We recorded the average value of year-round for monocultural cropping systems and rotation/mixed cropping systems. The database covered different agroecosystem types, e.g., cropland, grassland, rotation and mixed cropping systems, and

horticultural land. These sites spanned from 122.8°W to 176.3°E and from 46.4°S to 63.2°N (Figure 1).

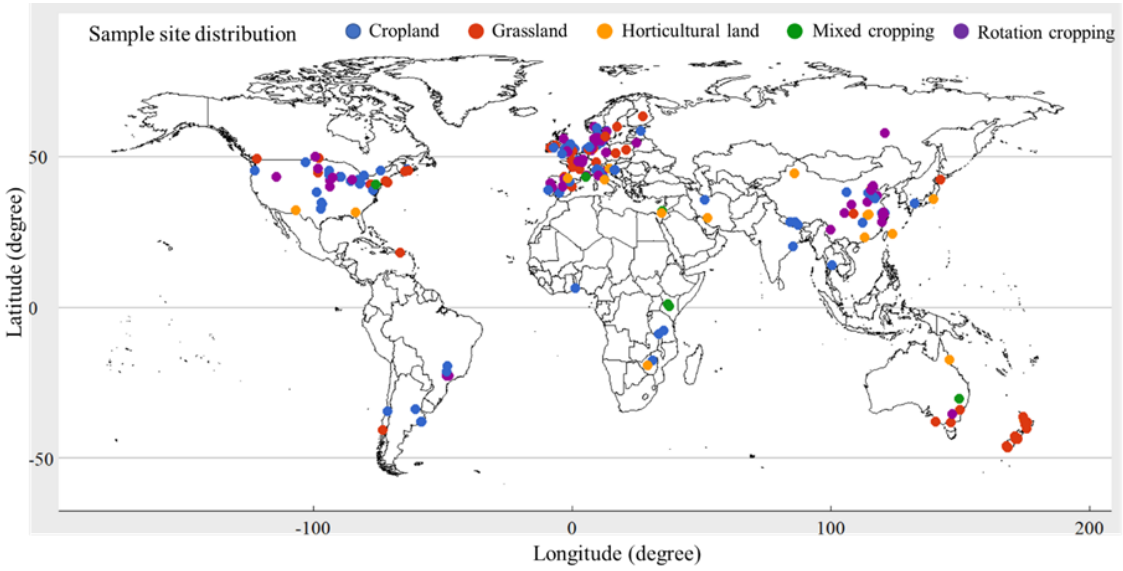


Figure 1. Study site distribution in the database

2.2 Machine learning model building and selection

We applied a SGB model to build a NO_3-N leaching model for global agroecosystems (NL_{NO3} model) in 1961, 2000 and 2010. There are four parameters to initialize NL_{NO3} model, namely learn rate (LR), subsample fraction (SF), number of trees (NT), and minimum terminal node (MINCHILD). Learn rate controls the contribution of each tree to the final model. Subsample fraction is involved in the learning process to prevent the model from overfitting. The number of trees is based on the learn rate. Minimum terminal nodes are regulated by the size of dataset. We tested different combinations of LR, SF, NT and MINCHILD parameters (Friedman, 2001; Pan *et al.*, 2021) and found the optimal configurations of NL_{NO3} model were 0.1, 0.9,1000 and 3, respectively. We followed a series of stepwise procedures (Thompson, 1978) to eliminate redundant input variables and optimize the number of variables for this NL_{NO3} model (Table S1). The stepwise variable selection method is efficient in computation and has been widely used in machine

learning variable selection (Rakotomamonjy, 2003). NL_{NO3} model selected N_IN, AI, IR, LONG. S_TEX, S_PH, S_TN and LUS (M5 in Table S1) as the important variables in predicting NO_3-N leaching. We then introduced a constraint of AI in NL_{NO3} model according to Wang *et al.* (2014). Under AI < 0.32 and no irrigation application, NO_3-N leaching can be neglected under such extremely dry conditions. Therefore, the complete model is described as follows:

$$NL_{NO3} = \sigma_{AI} \cdot M_{SGB} \quad (1)$$

$$M_{SGB} = \nabla F\ (N_IN, AI, IR, LONG, S_TEX, S_PH, S_TN, LUS) \quad (2)$$

$$\sigma_{AI} = \begin{cases} 0 & if\ AI < 0.32\ \&\ IR = 0 \\ 1 & otherwise \end{cases} \quad (3)$$

Among these equations, σ_{AI} is the constraint coefficient of AI, M_{SGB} is the original training model. $N_IN, AI, IR, LONG, S_TEX, S_PH, S_TN\ and\ LUS$ are the input variables of M_{SGB}. The accuracy of the model was evaluated by 10-fold cross-validation, where the whole dataset was first used for learning, and subsequently divided into ten folds. For each of the 10 folds, nine folds were used as a training set and the remaining fold as a test set. The test results from each fold were averaged to estimate the whole model performance. Regression coefficients of determination (R^2), mean square error (MSE) and mean absolute percentage error (MAPE) were used to measure the percentage of variation explained by the model and the model accuracy for NL_{NO3} models.

$$R^2 = \frac{\sum_{i=1}^{n} P_i - O_{avg}{}^2}{\sum_{i=1}^{n} O_i - O_{avg}{}^2} \quad (4)$$

$$\mathrm{MSE} = \frac{1}{n}\sum_{i=1}^{n}(P_i - O_i)^2 \quad (5)$$

$$\text{MAPE} = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{O_i - P_i}{O_i}\right| \quad (6)$$

Where n is the number of data pairs, O_i is the observed value, P_i is its predicted value. O_{avg} and P_{avg} are the averaged observed and its predicted values, respectively.

2.3 Data source

The soil property database (S_TEX, S_PH, S_TN) with a spatial resolution of 1 km^2 was obtained from the World Inventory of Soil Emission (WISE) database developed by the International Soil Reference and Information Centre (ISRIC) (www.isric.org). Climate data of MAP and PET in year 1961, 2000 and 2010 were sourced from CRU TS 3.21 data (0.5 degree by 0.5 degree) (https://crudata.uea.ac.uk/cru/data/hrg/), AI was calculated based on AI= MAP/PET. Irrigation data (year 1961 and year 2000) were sourced from Puma and Cook (2010). The irrigation rate (0.5 degree by 0.5 degree) of year 2010 was calculated based on year 2000 with an increase of 8% (AQUASTAT) (https://www.wri.org/aqueduct/). LUS with a spatial resolution of 10 km^2 in year 1961, 2000 and 2010 was obtained from Ellis and Klein Goldewijk (2019) based on HYDE model (v3.2) (Klein Goldewijk *et al.*, 2017). N_IN in cropland included synthetic fertilizer and manure N application rates with a 0.5-degree resolution, which are obtained from Lu and Tian (2016) and Zhang *et al.* (2017), respectively. N fertilization in grassland contained synthetic fertilizer, manure N application and manure deposition which were sourced from Xu *et al.* (2018) with a 0.5-degree resolution. All NL_{NO3} model computations were performed using TreeNet® (Salford Systems) and global distribution maps were plotted by The ESRI ArcGIS software (v. 10.4.1 ESRI).

3. Results

3.1 NO_3-N leaching based on field experiments

NO_3-N leaching varied widely across global agroecosystems, ranging from 2.8 to 219 kg N ha^{-1} yr^{-1} (5^{th}–95^{th} percentile) with an average of 59.2 kg N ha^{-1} yr^{-1} when the average N fertilization (synthetic fertilizer + manure application) was 282 kg N ha^{-1} yr^{-1}, accounting for ~21% of total N fertilization. The collected observations in this database covered a wide range of MAT and MAP with an average of 11.4 °C and 805 mm, respectively. The range of aridity index was from 0.13 to 1.33 (5^{th}–95^{th} percentile). This database was used to build the machine learning model (NL_{NO3} model) and subsequent analyses.

Table 1 Summary statistics of NO_3-N leaching database compiled in this study

	n	Mean ± SEM*	Median	5^{th}–95^{th} percentile
NO_3-N leaching (kg N ha^{-1} yr^{-1})	1818	59.2±1.9	32.4	2.8-219
MAP (mm)	1816	805± 8.5	735	314-1383
MAT (°C)	1816	11.4± 0.1	10.8	5.9-18.8
Evapotranspiration rate (mm)	1816	1173± 8.9	1170	808-2022
Aridity index	1816	0.77± 0.01	0.79	0.13-1.33
Soil pH	1201	6.55± 0.03	6.4	5-8.4
SOC (%)	1095	1.79± 0.05	1.27	0.38-4.75
Soil TN (%)	790	0.40± 0.05	0.15	0.04-0.67
Soil C/N ratio	406	13± 0.5	101.4	8-18
Soil CEC (cmol c/kg^{-1})	362	17.4± 0.8	14	2.3-48
N application rate (kg N ha^{-1} yr^{-1})	1818	282± 6.5	180	26.6-921
Irrigation rate (mm yr^{-1})	742	536.7± 19.7	422	45-1399

*SEM is the standard error of the mean value

3.2 Performance of NL_{NO3} model in predicting NO_3-N leaching

Based on stepwise variable selection, NL_{NO3} model (M5 in Table S1) has the highest R^2 (0.75) and lowest MSE (0.4) with only eight input variables. When the number of input variables was reduced (M1-M4), the model performance decreased significantly, whereas including more input variables (M6-M12) did not improve the R^2 or reduce the MSE. In this regard, M5 was considered the optimal model for further upscaling prediction of global NO_3-N leaching (Figure 2a). N_IN, AI and IR were the most important variables in the model, followed by LONG and soil properties, e.g., S_TEX, S_PH and S_TN (Figure 2b).

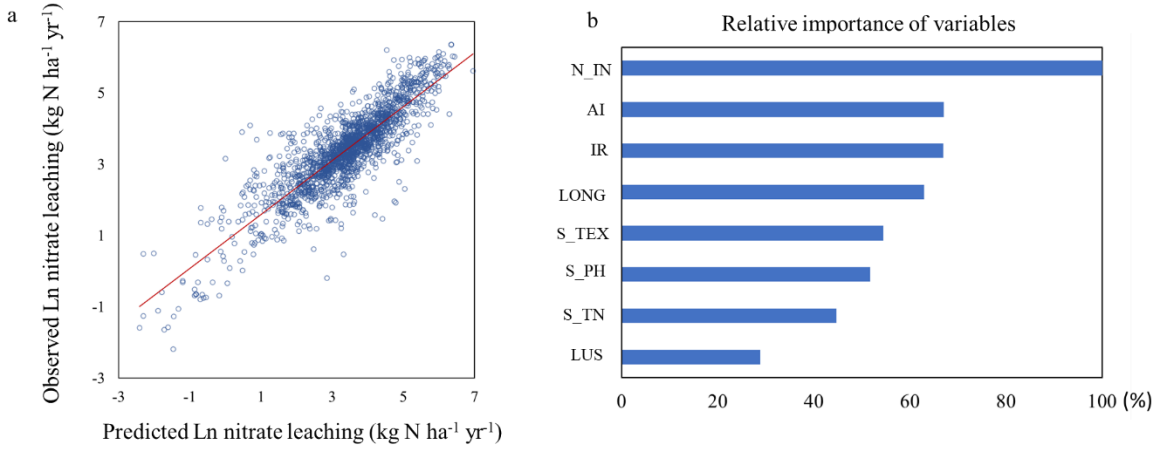


Figure 2. a) Scatter plot of the observed against predicted NO_3-N leaching by NL_{NO3} model (n=1818, R^2=0.75, MSE=0.4, MAPE = 0.27); b) relative importance of variables in NL_{NO3} model.

3.3 Global prediction of NO_3-N leaching based on NL_{NO3} model

According to the prediction of NL_{NO3} model, average NO_3-N leaching in 1961 was 5.8 kg N ha $^{-1}$ yr^{-1} and increased to 9.9 kg N ha $^{-1}$ yr^{-1} and 12.8 kg N ha $^{-1}$ yr^{-1} in 2000 and 2010, respectively (Table 2). NO_3-N leaching in cropland was higher than in grassland worldwide and showed an increasing trend from 1961 to 2010 (from 6.4 kg N ha $^{-1}$ yr^{-1} to 17.1 kg N ha $^{-1}$ yr^{-1}) (Table S2). NO_3-N leaching in grassland remained stable with an

average around 4 kg N ha $^{-1}$ yr^{-1} to 4.7 kg N ha $^{-1}$ yr^{-1} in three timepoints (Table S2). Total NO_3-N leaching in agroecosystems was 23.2 Tg N yr^{-1} in 1961 and increased to 39.7 Tg N yr^{-1} in 2010. In 1961, NO_3-N leaching accounted for ~30% of total N fertilization but dropped to ~18% in the 2000 and 2010.

Table 2. The N fertilization and simulated NO_3-N leaching worldwide in 1961, 2000 and 2010

Year	1961	2000	2010
Average NO_3-N leaching (kg N ha $^{-1}$ yr^{-1})	5.8	9.9	12.8
Total N fertilization* (Tg N yr^{-1})	77.8	184.9	221.7
Total NO_3-N leaching§ (Tg N yr^{-1})	23.2	32.8	39.7
NO_3-N leaching to total N application (%)	29.8	17.7	17.9

* Total N application in cropland includes synthetic N fertilizer and manure application. Total N fertilization in grassland includes synthetic N fertilizer, manure application and manure deposition. §Total NO_3-N leaching is the sum of NO_3-N leaching in cropland and grassland (Table S2).

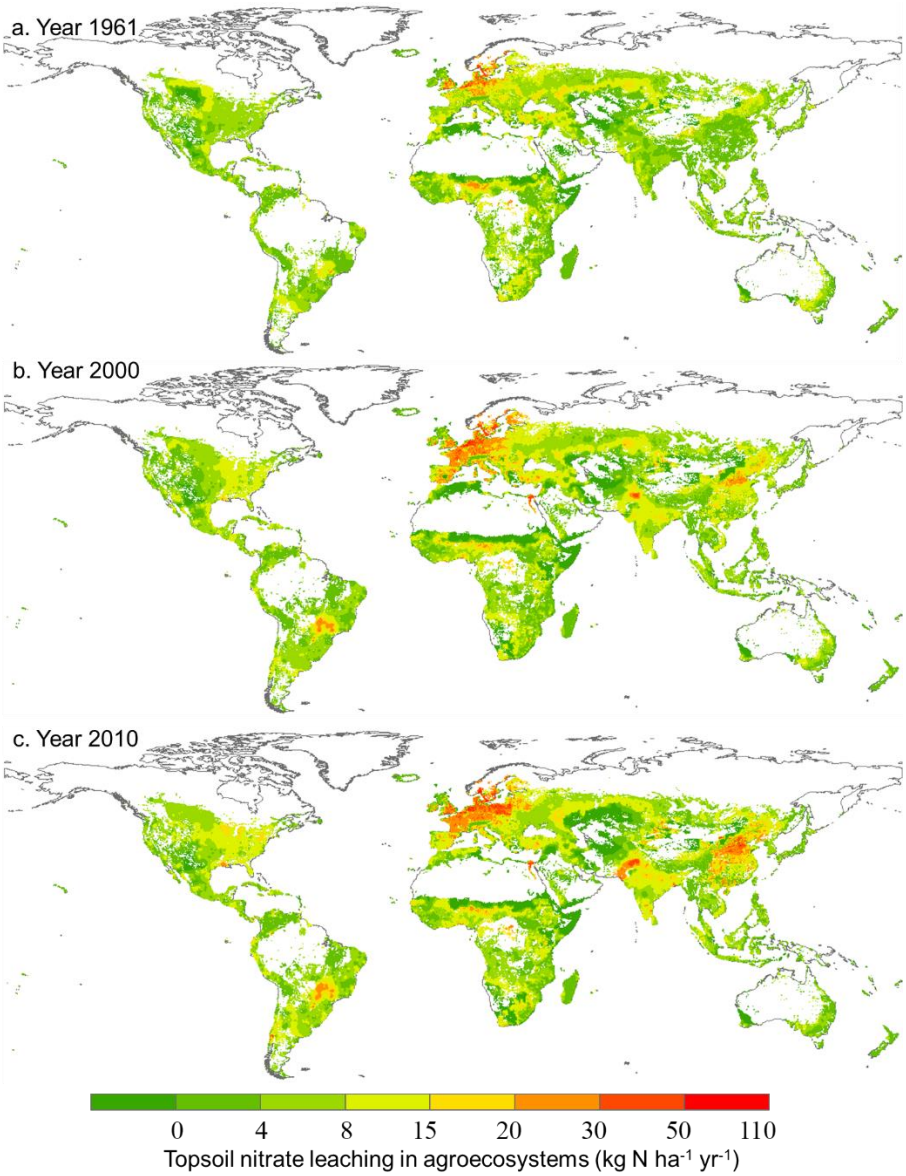


Figure 3. Global distribution of NO_3-N leaching in agroecosystems in year 1961, 2000 and 2010.

The prediction of global NO_3-N leaching in agroecosystems showed a high spatiotemporal variation (Figure 3). Europe had the highest NO_3-N leaching throughout three timepoints and expanded to a larger area in the 2000s. Compared to 1961 (4.1 kg N ha $^{-1}$ yr^{-1}), NO_3-N leaching in China increased by 127% (9.9 kg N ha $^{-1}$ yr^{-1}) and 224% (13.3 kg N ha $^{-1}$ yr^{-1}) in 2000 and 2010, respectively. Similar pattern can be observed in South Asia regions, NO_3-N leaching increased from 4 kg N ha $^{-1}$ yr^{-1} in 1961 to 6.6 kg N

ha $^{-1}$ yr^{-1} in 2000 and 9.7 kg N ha $^{-1}$ yr^{-1} in 2010. The hotspots (>20 kg N ha $^{-1}$ yr^{-1}) expansion can be observed in Brazil in 2000 and 2010 (Figure 3b &c).

NO_3-N leaching was strongly positively related to N_IN, IR and S_PH (Figure 4). NO_3-N leaching tended to slightly decrease with higher AI. NO_3-N leaching also showed a longitudinal pattern globally. NO_3-N leaching was highest near meridian areas and tended to decrease from meridian areas to west and east hemisphere.

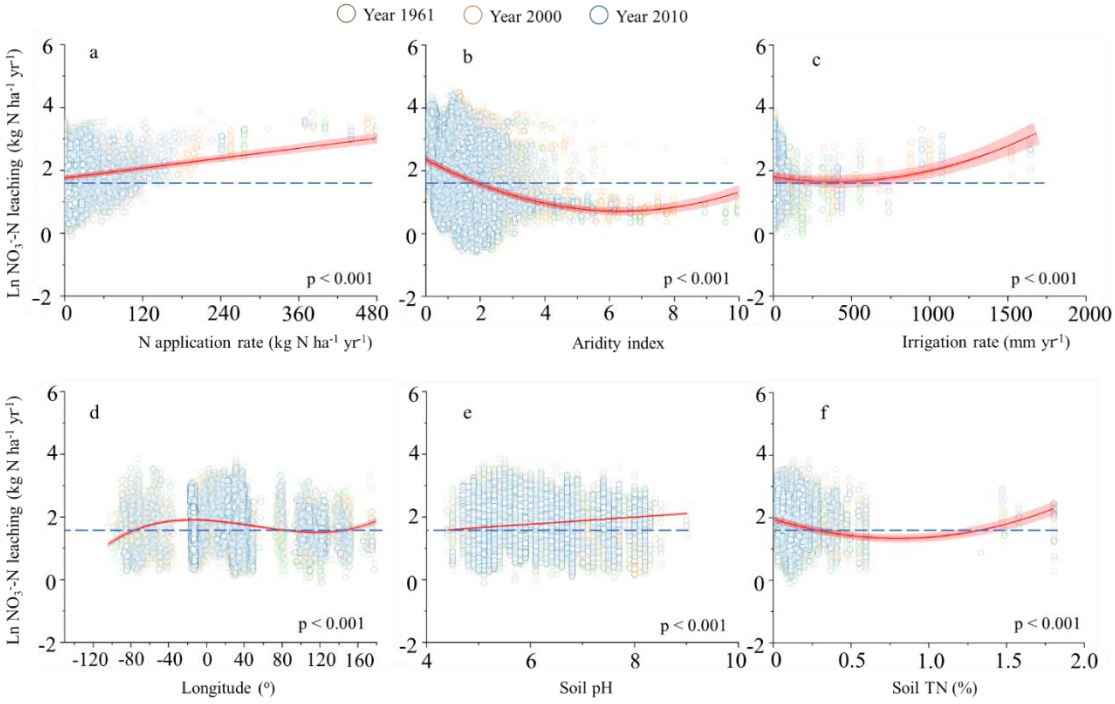


Figure 4. Relationships between NO_3-N leaching and (a) N_IN, (b) AI, (c) IR, (d) LONG, (e) S_PH and (f) S_TN. Shaded bands in figures indicate 95% confidence intervals, and the dashed lines represent the average NO_3-N leaching.

4. Discussion

4.1 NO_3-N leaching is a main N loss pathway in agroecosystems.

Based on NL_{NO3} model, the average NO_3-N leaching rate increased from 1961 to 2010. In 2010, the total NO_3-N leaching worldwide reached 39.7 Tg N yr^{-1}, accounting for ~18% of total N application to agricultural land. This was within the range of previous model estimation, e.g., Lin *et al.* (2001) estimated NO_3-N leaching from agricultural land was

around 19% in 1995. The value of NO_3-N leaching to total N application in 2000 and 2010 (18%) is much lower than the IPCC default emission factor of 30% (IPCC, 2006). This implies that the amount of NO_3-N leaching may be overestimated using the IPCC default factor. Furthermore, this constant value used to estimate the amount of NO_3-N leaching is problematic. We found that the proportion of NO_3-N leaching from agricultural land to total N fertilization varied from 1961 to 2010 (30% and 18%, respectively) with fertilization rate, irrigation rate, and environmental conditions. The amount of NO_3-N leaching contributed more to total Nr loss in cropland than that in grassland across all three timepoints (Table S2). Horticultural land usually receives high N and water input to guarantee productivity, causing high NO_3-N leaching. However, in most grassland systems, extensive management maintains a low N status in the soil, thereby limiting the amount of NO_3-N leaching due (Di and Cameron, 2002). Besides, N in grassland can be fixed by legumes efficiently, and captured by mowing, cutting for hay or silage, resulting in low leaching losses (Cameron *et al.*, 2013). Compared to 1961, the total NO_3-N leaching amount increased by 40% in 2000 and 70% in 2010 (Table 2). This indicates that since the green revolution, the rapid increase of N fertilizer use and irrigation has substantially increased NO_3-N leaching especially in cropland (Liu *et al.*, 2010; Wang *et al.*, 2017).

4.2 Spatiotemporal pattern of NO_3-N leaching and its drivers

The global temporal trend of NO_3-N leaching was mainly affected by the different levels of N application rates and irrigation rates. Previous studies have well-documented that NO_3-N leaching increased with the growth of N application rate (Eagle *et al.*, 2017; Qasim *et al.*, 2020) (Figure 4a). Besides, irrigation also played a critical role in contributing to NO_3-N leaching through the regulation of NO_3^- and its mobility by altering soil hydrological properties (Eagle *et al.*, 2017) (Figure 4c). When water supply exceeds

the water holding capacity, NO_3^- washed out from the rhizosphere and leached into the groundwater (Di and Cameron, 2002). In 1961, hotspots of NO_3-N leaching were mainly distributed at Europe countries since only developed countries had relatively higher N application rate at that time (Frink *et al.*, 1999) (20-100 kg N ha^{-1} yr^{-1}) (Figure S3). With the development of green revolution, the amount of N fertilization and irrigation increased rapidly, leading to more hotspots of NO_3-N leaching in 2000 and 2010 (Figure 3b, c), such as China and south Asia after 2000 (Figure S4).

Other than N fertilizer application rate and irrigation rate as mentioned above, the interactions between AI, soil TN, soil pH and soil texture have been shown (Figure 4 and Figure S5) to affect NO_3-N leaching in agricultural systems, such as cropland in Southern Brazil and grasslands in sub-Saharan Africa. This finding is consistent with previous studies (Liu *et al.*, 2010; Carbonell *et al.*, 2021). Soil pH affected NO_3-N leaching indirectly by influencing nitrification process in the soil (Cameron *et al.*, 2013). N is more prone to leaching after it is converted to NO_3^- by nitrification. With the increase of soil pH, soil nitrification and the amount of NO_3^- produced increased accordingly (Sahrawat, 2008), which would potentially promote NO_3-N leaching (Gao *et al.*, 2016). More soil TN may increase the soil N pool, accelerate N mineralization and subsequently enhance NO_3-N leaching (Di and Cameron, 2002). Coarse soil texture with high water permeability or percolation rate could increase NO_3-N leaching to the groundwater (Figure S5).

However, NO_3-N leaching in very humid tropical areas such as Indonesia and Malaysia with AI >1.5 were relatively low throughout three timepoints (<6 kg N ha^{-1} yr^{-1}), accounting for a relatively smaller proportion of N fertilizer inputs (6-13%), (Figure 3). The low NO_3-N leaching in humid tropical agricultural areas is also reported by Yoshikawa and Shiozawa (2008), who calculated the N budget in a tropical agricultural

watershed in Indonesia and found that NO_3-N leaching only accounted for a small proportion of the total N loss (13%) where 42% of N input was lost via NH_3 volatilization and denitrification. Normally, the higher precipitation in humid conditions could also increase the risk of NO_3-N leaching (Martinez-Feria *et al.*, 2019), especially in coarse-textured soils. NO_3-N leaching losses were less from the fine- texture soil in humid conditions, mainly due to its poor water permeability and slower percolation rate (Figure S5) (Di and Cameron, 2002). Meanwhile, denitrification can be promoted under waterlogging conditions that induced by higher precipitation in humid areas, leading to more gaseous N loss and less NO_3-N leaching (Sexstone *et al.*, 1985). Besides, acidic soils could limit nitrification rate (Morkved *et al.*, 2007), resulting in less available NO_3^- that can be leached.

The various distribution of NO_3-N leaching and their interactions among different driving factors indicated that site-specifical mitigation strategies should be adopted to reduce NO_3-N leaching, including optimizing irrigation management and adopting advance irrigation facilities (drip irrigation) in coarse-textured soils (Barton and Colmer, 2006), applying '4R' stewardship (using the right fertilizer with the right amount at the right time and the right placement) in heavy N input areas (Quemada *et al.*, 2013), developing precision agriculture in developed countries, using nitrification inhibitors in cropland (Eagle *et al.*, 2017).

4.3 Implications of predictions based on NL_{NO3} model

By combining literature-based database and machine learning models, we provided a reliable bottom-up estimation of NO_3-N leaching at a global scale. Predictions of NO_3-N leaching using existing process-based models are based on the combined water transport and N transport simulations (Keating *et al.*, 2003; Li *et al.*, 2007). Detailed soil and hydraulic parameters are required during calibration and validation in simulating NO_3-N

leaching. However, predisposed parameterization may cause greater errors (van der Laan *et al.*, 2014). Giltrap *et al.* (2014) has compared the performance of NZ-DNDC and APSIM models in simulating NO_3-N leaching on horticultural soils and found that neither model could achieve a reliable estimation of NO_3-N leaching. One possible reason is that these process-based models failed to include important parameters in N transport algorithms (Giltrap *et al.*, 2015). Unlike previous empirical models and process-based models, our NL_{NO3} model could capture the spatiotemporal responses of NO_3-N leaching by including soil properties, environmental conditions and farming practices as input variables, which are easily accessible.

With the accuracy and easily accessible of NL_{NO3} model, it is expected that this NL_{NO3} model could be further applied to scenario analysis. For example, the hotspots and global pattern of NO_3-N leaching under different climate change scenarios and N management practices could be estimated to inform better N management. Furthermore, this NL_{NO3} model can be directly embedded into a process-based model as a submodule to improve the simulation performance of process-based models for a particular system. On the other hand, the negative externalities associated with NO_3-N leaching worldwide have gained more attention, for example, based on the European Nitrogen Assessment, the social cost of NO_3-N leaching to groundwater under agricultural land in the EU27 in 2008 was estimated between €0-2 billion euro per year for human health impacts and €25-95 billion euro per year for ecosystems impacts (Brink *et al.*, 2011; Van Grinsven *et al.*, 2013). However, the damage costs of NO_3-N leaching to the society and the environment remain largely unquantified in most regions owing to the comprehensive data requirements for specific local circumstances. In this regard, this NL_{NO3} model could be used to estimate the damage costs of NO_3-N leaching at a global scale to highlight the importance of reducing NO_3-N pollution.

4.4 Uncertainty

The global data sets we used for simulation NO_3-N leaching were obtained from different sources with different resolutions (e.g., 0.5-degree resolution of aridity index, 10km^2 resolution of land uses systems). When we resampled and integrated the databases with different resolutions, deviations may occur during this process and result in uncertainties in our simulation. Secondly, deviations may exist in some input variables obtained from global databases at the grid cell. For example, the database of N fertilizer application rate used in this study is simulated by other models, and may affect the performance of our machine learning model.

5. Conclusion

We developed a machine learning model to predict NO_3-N leaching in global agroecosystems using our global literature-based database. This model can accurately simulate NO_3-N leaching with a few easily accessible input variables. According to the model estimation, NO_3-N leaching represented a major Nr loss pathway in global agroecosystems in 1961, 2000 and 2010. The global average NO_3-N leaching increased from 5.8 kg N ha $^{-1}$ yr^{-1} in 1961 to 9.9 kg N ha $^{-1}$ yr^{-1} in 2000 and 12.8 kg N ha $^{-1}$ yr^{-1} in 2010, respectively. Total NO_3-N leaching in agroecosystems accounted for 18% of total N fertilizer input in 2000 and 2010. Hotspots of NO_3-N leaching in agroecosystems expanded from Europe in 1961 to China, South Asia and Brazil in 2000 and 2010. The high spatiotemporal heterogeneity of NO_3-N leaching was mainly driven by soil properties (soil TN, soil pH, soil texture), aridity index and farming practices (N fertilization and irrigation). This study provides a novel approach to estimate NO_3-N leaching worldwide and the machine learning based NL_{NO3} model can be incorporated as a submodule into process-based models for an accurate projection of NO_3-N leaching globally.

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Supporting Information for

Modelling the spatiotemporal trend of nitrate leaching in global agroecosystems

Contents of this file

Table S1. Model prediction of NL_{NO3} by stepwise selection of variables

Table S2. The global N application and simulated NO_3^--N leaching loss in cropland and grassland in 1961, 2000 and 2010

Figure S1. Distribution of a) soil pH and b) TN in agroecosystems

Figure S2. Global distribution of arid index (AI)

Figure S3. Global distribution of N application rate (N_IN)

Figure S4. Global distribution of irrigation application rate (IR)

Figure S5. Average NO_3^--N leaching rate in different soil texture. Different letters in the figure indicate that the values are significantly different at $p < 0.05$.

Reference list of included studies.

Table S1. Model prediction of NL_{NO3} by stepwise selection of variables

Model	Stepwise variable selection	R^2	MSE	MAPE
M1	N_IN, IR, LONG, AI	0.66	0.58	0.33
M2	N_IN, IR, LONG, AI, S_TEX	0.67	0.58	0.32
M3	N_IN, IR, LONG, AI, S_TEX, S_PH	0.67	0.56	0.33
M4	N_IN, IR, LONG, AI, S_TEX, S_PH, S_TN	0.68	0.55	0.33
M5	N_IN, IR, LONG, AI, S_TEX, S_PH, S_TN, LUS	0.75	0.44	0.27
M6	N_IN, IR, LONG, MAP, AI, S_TEX, S_PH, S_TN, LUS	0.74	0.45	0.27
M7	N_IN, IR, LATI, LONG, MAP, AI, S_TEX, S_PH, S_TN, LUS	0.74	0.45	0.26
M8	N_IN, IR, LATI, LONG, MAT, MAP, AI, S_TEX, S_PH, S_TN, LUS	0.74	0.44	0.26
M9	N_IN, IR, LATI, LONG, MAT, MAP, AI, S_TEX, S_PH, SOC, S_TN, LUS	0.74	0.45	0.27
M10	N_IN, IR, LATI, LONG, MAT, MAP, AI, S_TEX, S_PH, SOC, S_TN, S_CN, LUS	0.74	0.45	0.26
M11	N_IN, IR, LATI, LONG, MAT, MAP, AI, S_TEX, S_PH, SOC, S_TN, S_CN, LUS, S_TC	0.75	0.44	0.26
M12	N_IN, IR, LATI, LONG, MAT, MAP, AI, S_TEX, S_PH, SOC, S_TN, S_CN, LUS, S_TC, S_CEC	0.74	0.45	0.26

Table S2. The global N application and simulated NO_3^--N leaching loss in cropland and grassland in 1961, 2000 and 2010

Year	1961		2000		2010	
Land use	Cropland	Grassland	Cropland	Grassland	Cropland	Grassland
Total N application (Tg N yr^{-1})	25.6	52.2	100.6	84.3	125.3	96.4
Average NO_3-N leaching (kg N ha^{-1} yr^{-1})	6.4	4.7	12.8	4.0	17.1	4.2
Total NO_3^--N leaching (Tg N yr^{-1})	8.6	14.5	19.1	13.6	25.9	13.8
NO_3^--N leaching to total N application (%)	33.7	27.8	19	16.2	20.7	14.3

* Total N application in cropland includes synthetic N fertilizer and manure application, Total N input in grassland includes synthetic N fertilizer, manure application and manure deposition.

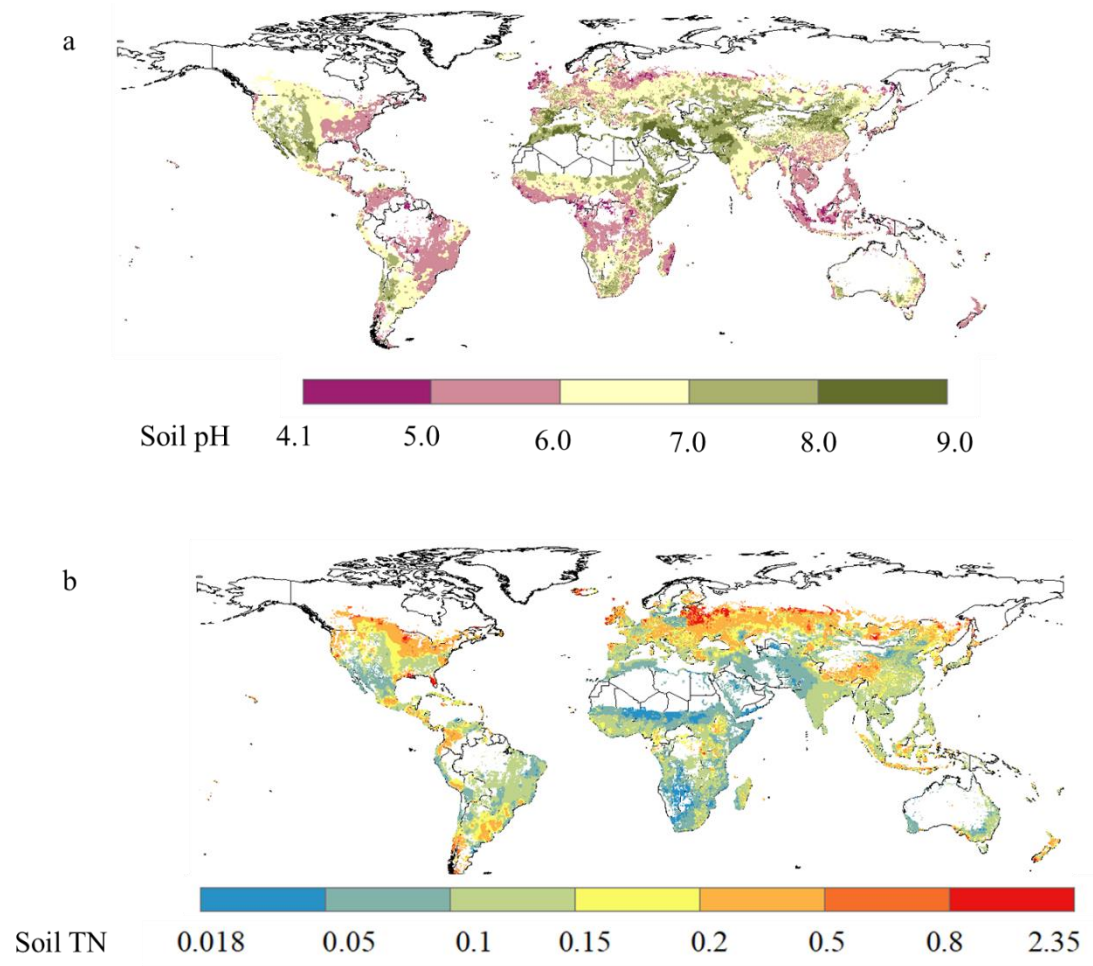


Figure S1. Distribution of a) soil pH and b) TN in agroecosystems

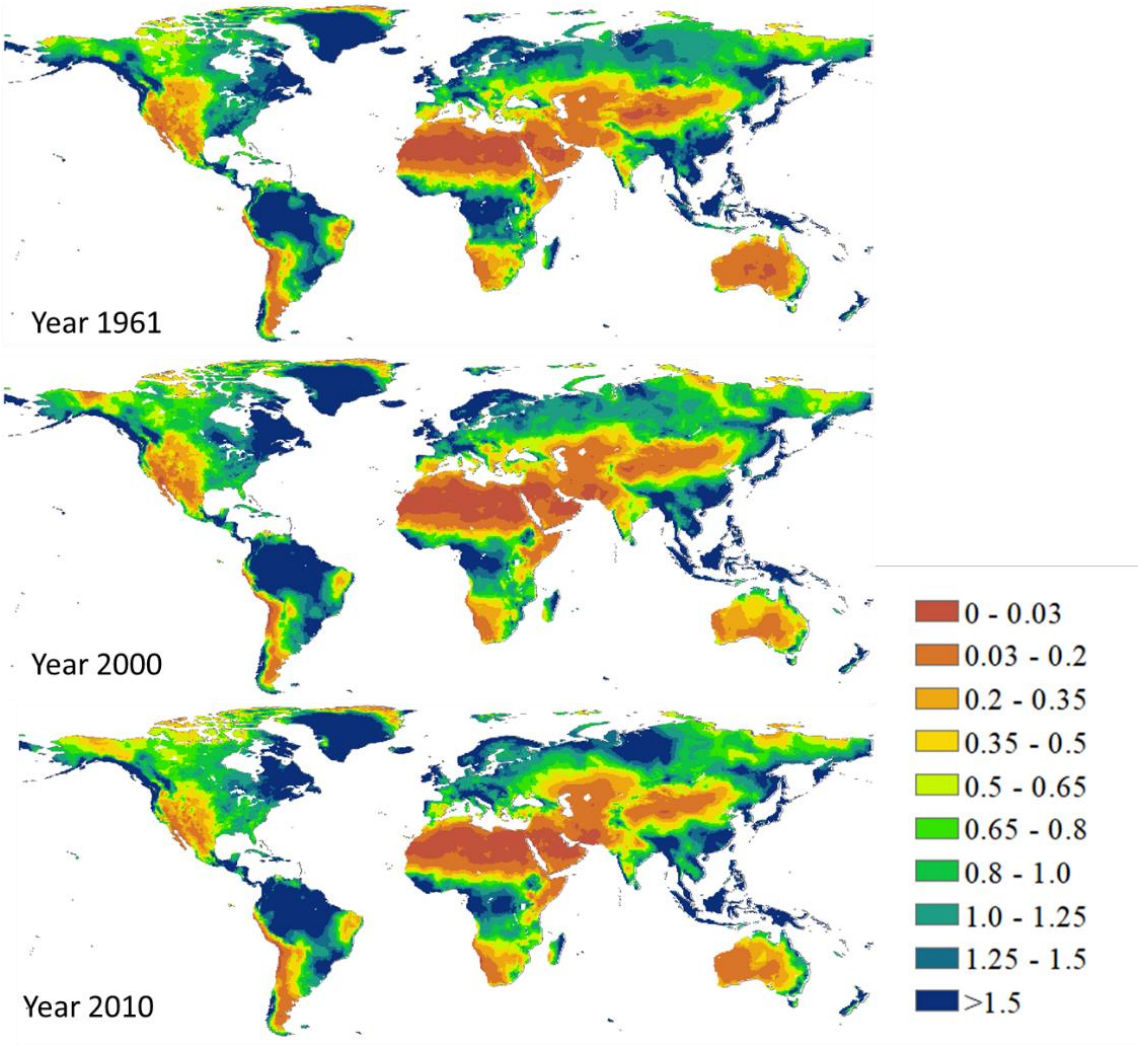


Figure S2. Global distribution of arid index (AI)

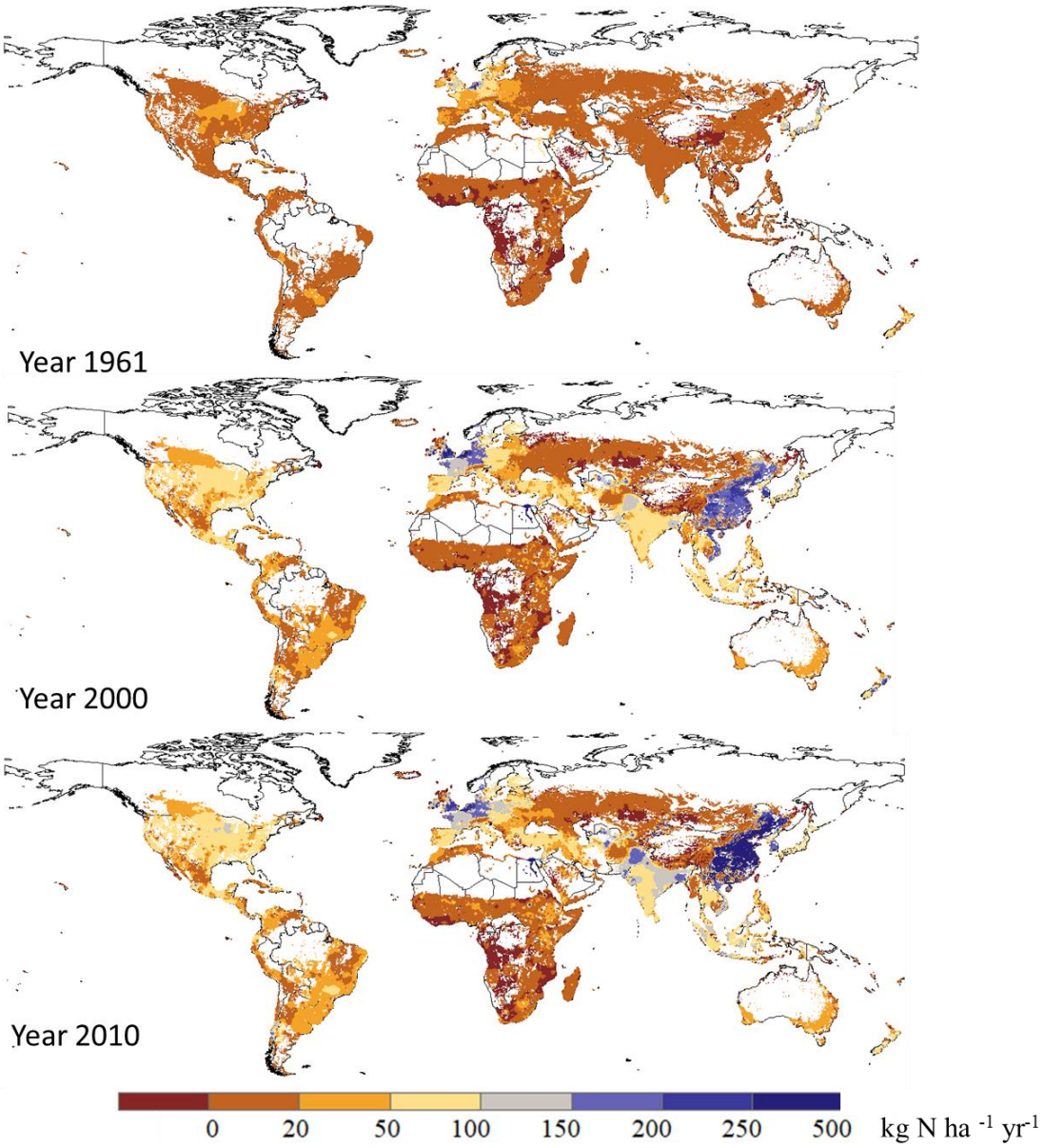


Figure S3. Global distribution of N application rate (N_IN)

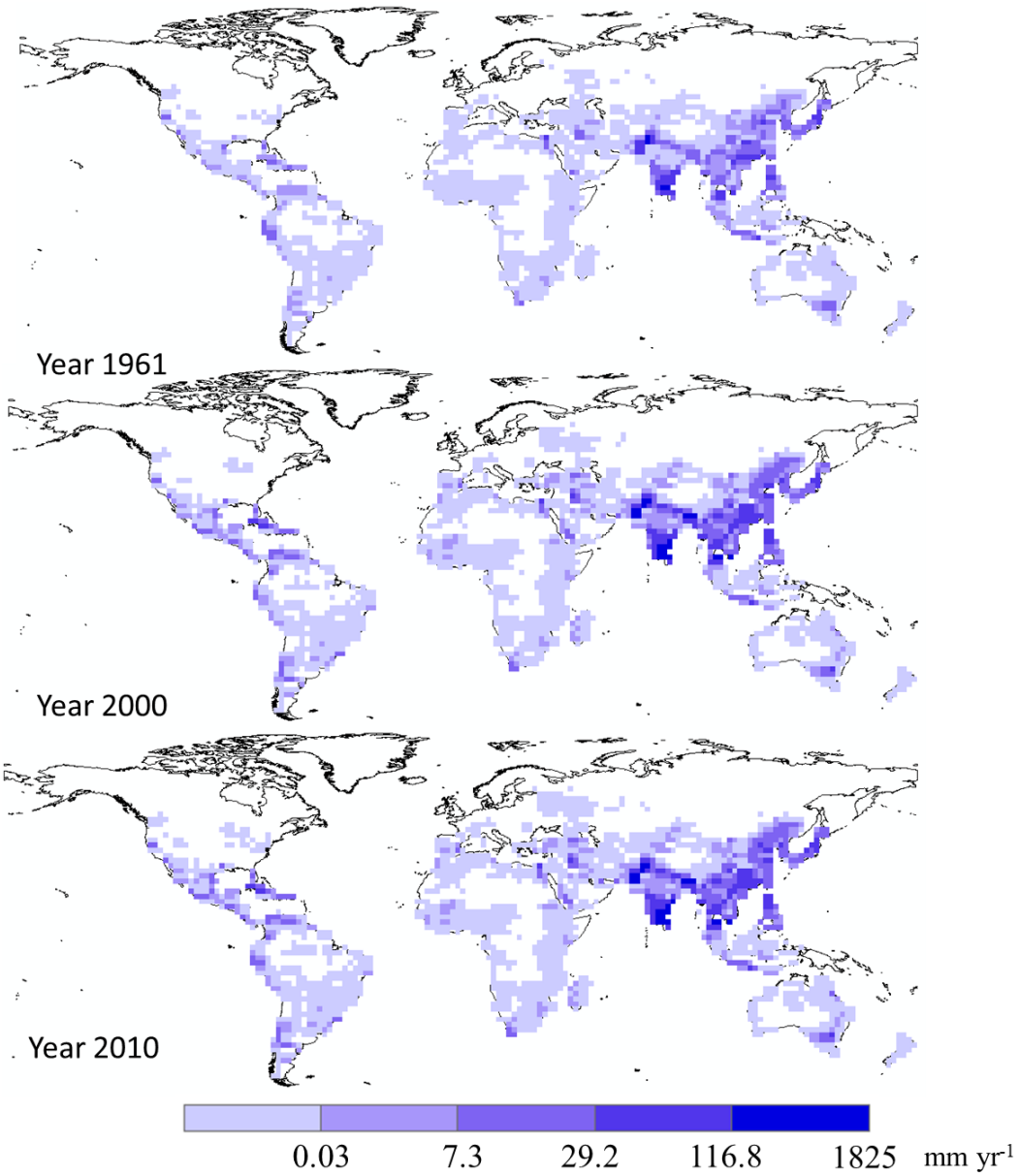


Figure S4. Global distribution of irrigation application rate (IR)

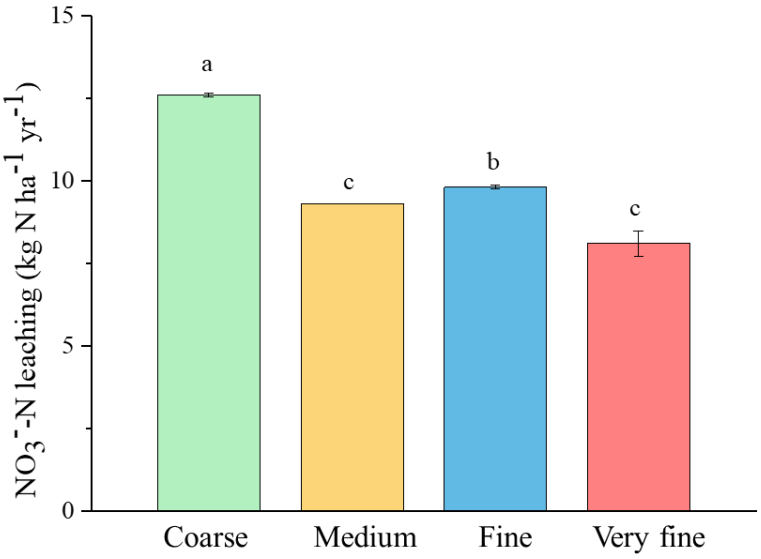


Figure S5. Average NO_3^--N leaching rate in different soil texture. Different letters in the figure indicate that the values are significantly different at $p < 0.05$.

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CHAPTER 8

GENERAL DISCUSSION AND CONCLUSIONS

8.1 Major findings

The literature review in Chapter 2 demonstrated that the understanding of N cycling in agroecosystems is essential to improve N management and to sustain environmental quality. Process-based models are developed to simulate the complex interactions of water, carbon and N cycles of terrestrial ecosystems. However, limitations of current process-based models were mainly associated with data availability, complex relationships between input and output variables and difficulty of model parameter estimation. To address the limitations of process-based models, improve the prediction and simulation of N losses, increase NUE and environmental quality, comprehensive databases of N loss from different pathways should be compiled by data mining, advanced machine learning models should be provided to reflect the linkage between N loss pathways and soil, environmental and climatic conditions. Chapter 3 described the the general methods of each study.

To predict nitrification rate (R_{nit}) and the fraction of nitrification as N_2O emissions ($f_{N_2O_Nit}$), data mining and machine learning based SGB models were innovatively applied. Firstly, a global database on R_{nit} and $f_{N_2O_Nit}$ was compiled. The database revealed that the average potential R_{nit} in the topsoil was 1.4 kg N ha^{-1} d^{-1}, and $f_{N_2O_Nit}$ ranged from 0.004 to 9.19% (average 0.46%). The SGB models were more accurate in predicting $R_{nit}$and associated N_2O emissions by using the same input variables compared to three widely used process-based models. The SGB models can be extrapolated to a global scale with only a few input variables without compromising accuracy. The potential R_{nit} was predicted by MAT, soil C/N ratio and soil pH (R^2 = 0.76). $f_{N_2O_Nit}$

varied by over 200 times, which challenged the common practice of using a constant value in process-based models. Instead $f_{\mathrm{N_2O_Nit}}$ should be adjusted according to edaphic and environmental conditions. Large spatial variation in nitrification and its fraction as N_2O emissions was mainly driven by long-term environmental and edaphic factors. R_{nit} and $f_{\mathrm{N_2O_Nit}}$ were highest in subtropical and equatorial regions but lowest in tropical and northern latitude regions. This study demonstrates the novel application of machine learning models in predicting R_{nit} and $f_{\mathrm{N_2O_Nit}}$ and provides insights into advancing process-based models for projecting N dynamics and greenhouse gas emissions (Chapter 4).

To conduct a global assessment of soil denitrification rate, $N_2O/(N_2O+N_2)$, and their driving factors and mitigation strategies, 225 studies (3367 observations) were extracted from global literature. The results showed that N loss through denitrification varied greatly across land uses and climatic regions with an average of 0.25 kg N $ha^{-1}d^{-1}$. The average emission factor of denitrification (EF_D) was 4.8%. The wide range of $N_2O/(N_2O+N_2)$ (mean: 0.33) demonstrated that the adoption of a fixed ratio in some process-based models for estimating N_2 emissions from denitrification is not suitable. N_2 loss accounted for 67% of total denitrification. N loss as N_2, although harmless to the environment, deserves more attention from the perspective of improving N use efficiency. Soil denitrification rate was positively related to soil WFPS, NO_3^- content and soil temperature and decreased with higher soil oxygen content. N_2 emissions increased with latitude, WFPS, soil mineral N but decreased with soil oxygen content. $N_2O/(N_2O+N_2)$ ratio increased with soil oxygen content and NO_3^- content but decreased with organic C, C/N ratio and WFPS. A meta-analysis of the effects of potential mitigation strategies on reducing denitrification rate and N_2 emission showed that optimizing N application rates, using ammonium-based fertilizers compared to nitrate-based fertilizers, biochar

amendment and application of nitrification inhibitors could effectively reduce soil denitrification rate and associated N_2 emissions by 34-219% and 15-226%, respectively. These findings highlight that N loss via soil denitrification and N_2 emissions cannot be neglected, and that mitigation strategies should be adopted to reduce N loss and improve N use efficiency. Our study provides a solid foundation to large-scale estimations of denitrification and the refinement of relevant parameters used in the submodels of denitrification in process-based models (Chapter 5).

To evaluate the contribution of N_2O production pathways and its driving factors in forest soils, the magnitude and geographical pattern of N_2O production pathways in forest soil in both Australia and worldwide were investigated. The results obtained from an incubation experiment with ^{15}N tracing technique showed that based on 13 forest soils sampled within Australia, forest soils in temperate areas had the highest N_2O emission rate (19.5 μg N kg^{-1} soil d^{-1}), followed by subtropical and arid soils (3.84 and 3.80 μg N kg^{-1} soil d^{-1}, respectively). Heterotrophic nitrification dominated N_2O production in Australian forest soils; its contribution followed the order of arid (78%) > subtropical (69%) > temperate (59%). N_2O from heterotrophic nitrification was negatively related to MAT and the contribution of heterotrophic nitrification to N_2O production was negatively related to soil TN and TC. These results partially agreed with the global literature data synthesis, which showed that in addition to heterotrophic nitrification (42%), denitrification (43.5%) was a main pathway of N_2O production in global forest soils. Globally, soil pH, moisture content, soil TN content, TC content and MAT attributed to the contribution of heterotrophic nitrification and denitrification to N_2O production. Such findings improved the understanding of N_2O production pathways in forest soils and had implications for the simulation and projection of global N_2O production from forest soils (Chapter 6).

To predict NO_3-N leaching from agroecosystems, a machine learning model (NL_{NO3} model) was developed based on a global literature-based database of NO_3-N leaching from agroecosystems (1818 observations from 286 studies). The NL_{NO3} model can reliably predict NO_3-N leaching using a few easily accessible input variables (R^2=0.75). The global spatiotemporal pattern and hotspots were identified based on NL_{NO3} model. The total NO_3-N leaching in agroecosystems increased from 23.2 Tg N yr^{-1} in 1961 to 32.8 Tg N yr^{-1} in 2000 and 39.7 Tg N yr^{-1} in 2010. Total NO_3-N leaching in agroecosystems accounted for 18% of total N fertilizer input in 2000 and 2010. Hotspots of NO_3-N leaching in agroecosystems expanded from Europe in 1961 to China, South Asia and Brazil in 2000 and 2010. The high spatiotemporal heterogeneity of NO_3-N leaching was mainly driven by soil properties (soil TN, soil pH, soil texture), aridity index and farming practices (N fertilizer application rate and irrigation rate). This study provided a novel approach to estimate spatiotemporal NO_3-N leaching and could be incorporated into process-based models to simulate NO_3-N leaching with easily accessible input variables (Chapter 7).

8.2 Implications of the study

8.2.1 Improving the simulation capacity of process-based model

In Chapters 4-7, N loss from nitrification, denitrification and leaching were quantified under different soil, environmental and management conditions at a global scale based on the comprehensive literature-based databases. The nitrification rate, denitrification rate and nitrate leaching rate summarized in this study can be used as the benchmark for calibration and to quantify the uncertainty of process-based models. Besides, the parameters used in process-based models to estimate the N_2O from nitrification, N_2 and N_2O from denitrification and the contribution of N_2O from nitrification and denitrification have been proven to be problematic (Chapter 4-6). The data-driven SGB models could

derive above parameters site-specifically according to the edaphic and environmental conditions that can be incorporated into process-based models to simulate the N loss and improve the model performances.

8.2.2 The application of data mining and machine learning model

In Chapter 4 and Chapter 7, two SGB models based on literature-based databases were developed to predict nitrification rate, and nitrate leaching, respectively. By stepwise variable selection, SGB models could eliminate redundant variables and use a few easily accessible input variables to achieve satisfactory performance of simulations (R^2= 0.76 and 0.75, respectively). Therefore, databases based on literature provide sufficient information to build satisfactory machine learning models. Fewer input variables enabled SGB models to perform global predictions and provided fundamental data for further analysis, such as scenario analysis and damage cost calculation of N loss. For example, in SGB model of nitrification, MAT is an important input variable, by altering MATs under different climate change scenarios, the effect of climate change on soil nitrification rate can be simulated and the corresponding management adaptions could be applied. In Chapter 7, SGB model gave a bottom-up estimation of NO_3-N leaching at a global scale. This model results provided a unique opportunity in estimating the damage costs of NO_3-N leaching at a global scale and abatement costs to address the significant challenges associated with reducing NO_3-N pollution.

8.2.3 Improving N management and NUE

In this study, the relationships between N loss pathways and soil, environmental and management conditions were identified (Chapter 5,6,7), the mitigation strategies of nitrate leaching were assessed by meta-analysis in Chapter 5, the hotspots of nitrate leaching at the global scale were simulated by SGB models (Chapter 7). These indicated

that to reduce N loss in terrestrial ecosystems, improve NUE and sustain environmental quality, management practices should be adopted. This includes: 1) optimizing N application rates, using ammonium-based fertilizers compared to nitrate-based fertilizers, biochar amendment and application of nitrification inhibitors, and advanced irrigation technique could effectively reduce soil denitrification rate and associated N_2 emissions. 2) Mitigation strategies should be adopted to reduce NO_3-N leaching site-specifically through, for example, optimizing irrigation management and adopting advanced irrigation facilities (drip irrigation) in coarse-textured soils (Barton and Colmer, 2006), applying '4R' stewardship (using the right fertilizer with the right amount at the right time and the right placement) in heavy N input areas (Quemada *et al.*, 2013), developing precision agriculture in developed countries, using nitrification inhibitors in cropland (Eagle *et al.*, 2017).

8.3 Limitations of the study

The first limitation is related to the data availability. Even this study is based on extensive literature-based databases of N loss from different pathways and provides satisfactory performance of machine learning models, some of these databases e.g., nitrification still contain a low number of observations. This indicated limited geographical coverage, which could lead to uncertainties of simulation. Moreover, even SGB models can well handle relatively small databases, the performance of SGB model could be further improved by including more data for learning and training. Furthermore, according to the analysis, N loss from different pathways varied largely across different soil, environmental and management conditions. More studies should be conducted to provide more available data for future simulation and investigation. Secondly, for global simulation such as nitrification and nitrate leaching, deviations may exist in some input variables obtained from global databases at the grid cell. For example, the database of N

fertilizer application rate used in nitrate leaching prediction is simulated by other models, and this may affect the performance of the machine learning model Thirdly, in SGB models of nitrification and nitrate leaching, constraint coefficients were applied in both models to regulate the simulation results. However, the simulation errors may still exist due to the potential bias of databases. Last but the not least, the difficulties in measuring N process may lead to uncertainties of data and subsequent simulation results. For example, previous studies adopted acetylene inhibition technique (AIT) to measure denitrification, which had been proven to underestimate denitrification rate both in situ and laboratory conditions (Wang and Yan, 2016). Advanced techniques should be adopted to such variations.

8.4 Suggestions for further study

This study demonstrated the possibilities of data mining in analysing and revealing patterns of N loss pathways. Global prediction of N loss was performed in Chapters 4 and 7 by integrating data mining and machine learning models. In the future study, other N loss pathways, e.g., NH_3 volatilization can be conducted to complete a comprehensive N loss prediction on a global scale. Furthermore, N processes interact with each other and are highly affected by the interactions between edaphic, environmental and management conditions in terrestrial ecosystems. In the future study, the application of data mining and machine learning methods can be extended to the simulation and quantification of the N cycling processes.

In this study, the limitations of process-based models have been demonstrated and the advantages of data mining and machine learning methods have been evaluated. In the future study, the next level of hybrid model can be proposed and applied. First, data mining combined with machine learning models can be leveraged to predict missing data during process-based model initialisation. For instance, machine learning models can be

built on the data collected from the soil of certain types at some locations and then used to predict missing data for the soil of a similar type and/or at a similar location. It will provide a more accurate estimation of missing data and thus ensure that process-based models run properly. Secondly, the data-driven machine learning model can be embedded as the submodule in process-based to improve the model performance. This brand-new module is parallel with the existing modules of other N processes. Also, for a module that already exists, a data-driven machine learning model can be built as its alternative and switch from the process-based model to the machine learning model, or vice versa, when performance becomes unsatisfactory.

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