ABSTRACT

LI, QI. Characterization of Atmospheric Reactive Nitrogen Emissions from Global Agricultural Sources. (Under the direction of Dr. Viney P. Aneja).

Reactive nitrogen (Nr) includes inorganic chemically reduced nitrogen compounds such as ammonia (NH3) and ionic ammonium (NH4+); inorganic chemically oxidized nitrogen compounds such as nitrogen oxides (NOX), nitric acid (HNO3), nitrate ion ($NO3^-$), and nitrous oxide (NO2); and organic nitrogen compounds such as urea, amino acids, and proteins. They are biologically active, chemically reactive, and radiatively active in the Earth's atmosphere and biosphere, which are different from non-reactive nitrogen gas.

Here, three statistical model (N2O_STAT, NH3_STAT, and NO_STAT) are developed for characterizing atmospheric Nr emissions from agricultural sources. We obtained Nr emissions and physicochemical variables (i.e., air temperature, soil temperature, soil moisture, soil pH, and Ni input to the soil) from published journal articles. The statistical model was developed by expressing a multiple linear regression equation between Nr emission and the physicochemical variables. The model was evaluated for 2012 Nr emissions. Results of the model are compared with other global and regional models (e.g. EDGAR, etc.). The results indicate that, in comparison to other data sets, the model generates a lower global N2O estimate by 9-20% (N2O_STAT: 3.75 Tg N yr-1; EDGAR: 4.49 Tg N yr-1; FAO: 4.07 Tg N yr-1), a lower global NH3 estimates by 57%, (NH3_STAT: 14.2 Tg N yr-1; EDGAR: 33.0 Tg N yr-1), and a lower global NOx estimates by 87%, (NO_STAT: 0.2 Tg N yr-1; EDGAR: 1.6 Tg N yr-1). We also performed a region-based analysis (U.S., India, and China) using the N2O_STAT and NH3_STAT models. Based on results, the difference in the global estimates is attributed to the lower estimates in major agricultural countries like China and India. Statistical models can capture the spatial distribution of global Nr emissions by utilizing a more simplified approach

than those used by other readily available data sets. Moreover, three models provide an opportunity of predict future Nr emissions in a changing world.

Characterization of Atmospheric Reactive Nitrogen Emissions from Global Agricultural Sources

by
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APPROVED BY:

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DEDICATION

For my Mom and Dad.

For my friends in U.S. and in China.

For Super Junior.

BIOGRAPHY

Qi Li was born in Anyang, Henan Province, China. She finished her elementary, middle, and high schools in her hometown and was admitted to Ocean University of China in 2011. As freshmen, she joined Environmental Protection Association in college and showed strong interests in environment issues, especially in air quality problems.

In August of 2015, Qi began her study in Department of Marine, Earth, and Atmospheric Sciences at North Carolina State University. After finishing required coursework, she joined Air Quality Research Group in January of 2017. She was fortune enough to have her thesis advisor, Dr. Viney Aneja, provide her with broad topics of reactive nitrogen studies. At the same time, she also enrolled in Statistics MS program at NC State to get a comprehensive training in data analysis. With strong background in both atmospheric science and statistics, Qi was able to develop a statistical model to estimate the global emission of reactive nitrogen from agricultural soil during her master's project. Upon completion of her degree, Qi is employed by a healthcare company as a data scientist.

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CHAPTER 1 Introductions

Reactive nitrogen includes inorganic chemically reduced nitrogen compounds such as ammonia (NH3) and ionic ammonium (NH4+); inorganic chemically oxidized nitrogen compounds such as nitrogen oxides (NOX), nitric acid (HNO3), nitrate ion (NO3−), and nitrous oxide (NO2); and organic nitrogen compounds such as urea, amino acids, and proteins. They are biologically active, chemically reactive, and radiatively active in the Earth's atmosphere and biosphere, which are different from non-reactive nitrogen gas (N2).

1.1. N2O

The largest human influence on climate change since the end of the 20th century is the emission of greenhouse gases to the atmosphere. In addition to carbon dioxide (CO2), nitrous oxide (N2O) is one of the most important greenhouse gases, which has a global warming potential (GWP) of ~300 for a 100-year timescale (Myhre et al., 2013). N2O also contributes to the destruction of ozone in the stratosphere (Ravinshankara et al., 2009). The lifetime of N2O in the atmosphere is ~114 years, indicating its long-term influence on a planetary scale (Forster et al 2007).

Since 1978, globally-averaged N2O levels in the atmosphere have been collected at Moana Loa, Hawaii. Prior to 1978, N2O levels have been extracted from ice core data from Antarctica. N2O concentrations in the atmosphere were relatively stable until the start of the Industrial Revolution. Since that time, N2O concentration has increased by 18%, from ~270 ppb to the current value of ~331 ppb (Figure 1.1). This increase is primarily due to intensive human activities, particularly related to agriculture.

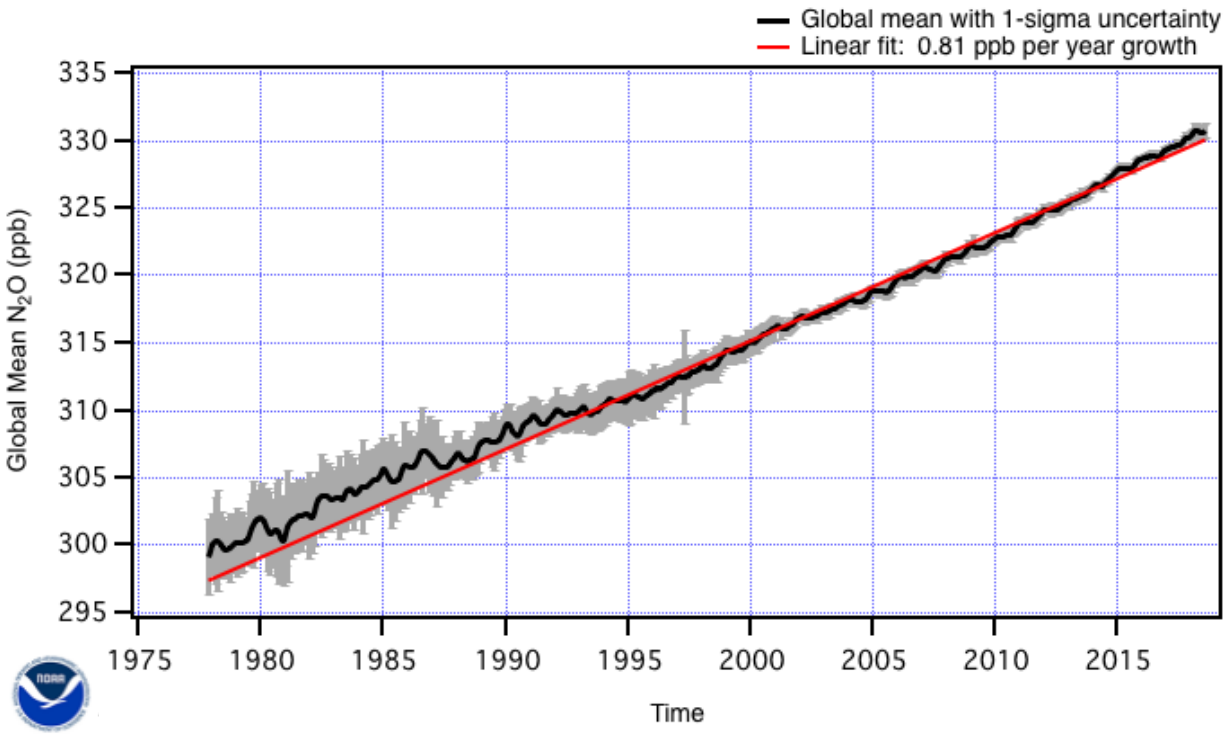

Figure 1.1 Atmospheric N_2O concentration trend from the combined Global Monitoring Division data set for the period 1977-2018 (NOAA, 2018).

Ciais et al. (2013) have estimated the yearly emission of N2O to be 17.9 Tg N yr-1 for the period 2006-2011. N2O emissions from natural sources are 11.0 Tg N yr-1, which includes sources from land and ocean. Agricultural activities are the primary anthropogenic source of N2O, contributing to 5.6 Tg N yr-1 (Schlesinger and Bernhardt, 2013). With increasing food, fiber and energy production, along with climate change and agricultural intensification, N2O emissions from soils have increased since 1990, from 3.5 Tg N yr-1 to 4.1 Tg N yr-1 (IPCC, 2006).

During the period 1961-2010, global N2O emissions from manure usage have grown from 1.17 to 2.03 Tg N yr-1 at an average rate of 1.10% yr-1 (Smith et al., 2014). Meanwhile during the same period, N2O emissions from synthetic fertilizers have increased 0.14 to 1.40 Tg N yr-1 at an average rate of 3.90% yr-1 globally (Tubiello et al., 2013). This indicates that synthetic fertilizers will have a greater contribution to N2O emission than manure within less

than 10 years (Smith et al., 2014). Besides agriculture, other sources such as fossil fuel combustion, industrial processes, and biomass burning also contribute to the total N2O emissions, but these are all relatively small sources. Intensive fertilizer usage has increased the emissions of reactive nitrogen compounds, including N2O from soils through nitrification and denitrification processes (Figure 1.2). Denitrification is a microbially facilitated process that leads to significant N losses from agricultural systems. During denitrification, nitrate and nitrite are reduced, and a portion converted into N2O (Butterbach-Bahl et al., 2013). Nitrification is the aerobic microbial oxidation of ammonium or ammonia to nitrite followed by oxidation of nitrite to nitrate. It is an important process in the nitrogen cycle in which N2O is generated as a by-product that leaks from microbial cells into soil and eventually into atmosphere (Schlesinger and Bernhardt, 2013).

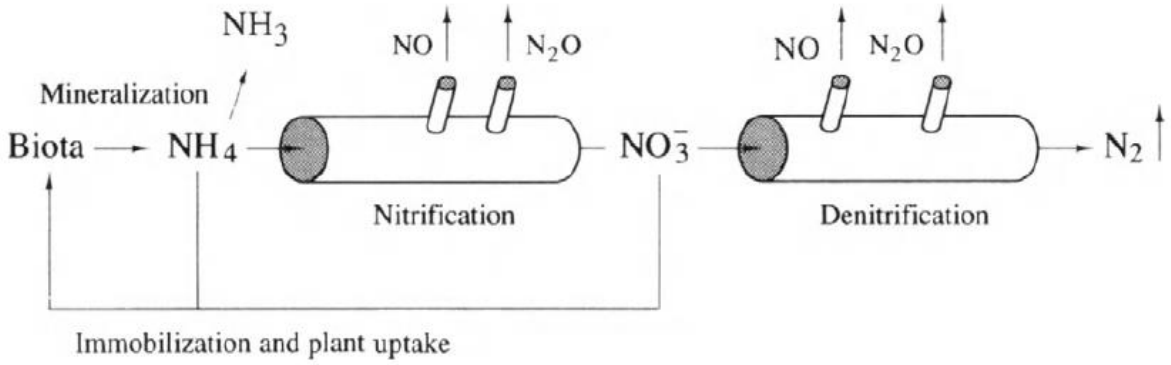

Figure 1.2 N_2O production through nitrification and denitrification in soil (Firestone and Davidson, 1989, as modified by Schlesinger and Bernhardt, 2013).

The ratio of N2O/NO3- (nitrification) and N2O/N2 (denitrification) products are important parameters determining the change of global budget, and they are affected by various physical, chemical and biological factors and their interacting effects (Granli and Bockman, 1994). To develop a predictive model for N2O emission under different agroecosystems, climate variables, soil characteristics, and cropping practices must be considered. Granli and Bockman (1994) provided a review on factors that control N2O emissions from soil, including moisture

and aeration, temperature, soluble and decomposable carbon, soil and fertilizer nitrogen, soil pH and salinity. Table 1.1 summarizes the variables that affect product ratios during nitrification and denitrification in agricultural soils. In nitrification, N2O emission will increase with decreasing O2 concentration, increasing water above field capacity, low NH4+ concentrations, increasing or decreasing pH, and increasing temperature. The production of N2O by denitrification is enhanced in conditions characterized by high nitrate, low C availability, low pH, low temperature, low N2O reductase activity, and 60 to 90% water filled pore space (WFPS) (Stehfest and Bouwman, 2006; Rashti et al., 2015; Wang et al., 2017).

Table 1.1 Variables that affect product ratios during nitrification and denitrification in agricultural soils (Granli and Bockman, 1994; Firestone and Davidson, 1989).

Process	Variable	Effect
		Will increase N_2O/NO_3^- ratio
Nitrification	$[O_2]$	Decreasing O_2 concentration
	$[H_2O]$	Increasing H_2O above field capacity
	$[NH_4^+]$	Low NH_4^+ concentrations
	pH	Increasing or decreasing pH
	Temperature	Increasing temperature
		Will increase N_2O/N_2 ratio
Denitrification	$[NO_3^-]$ or $[NO_2^-]$	Increasing oxidant
	$[O_2]$	Increasing O_2
	Carbon	Decreasing C availability
	pH	Decreasing pH
	Temperature	Decreasing temperature
	Enzyme status	Low N_2O reductase activity
	$[H_2O]$	Decreasing between 60 and 90% WFPS

Uncertainty of estimates of sources and sinks still exist in global N2O budget, especially from agricultural soils (Oertel et al., 2016). Inadequate understanding of the real mechanism in N2O emission from agricultural soils may be responsible for limited attention to agriculture as a

major contributor of the increasing atmospheric N2O. With increasing food and energy production, along with climate change and agricultural intensification, N2O emissions from soils and groundwater will continue to increase, suggesting the necessity of an updated global budget for N2O that accounts for these recent changes.

1.2. NH3

Among all the reactive nitrogen species, NH3 is an important component of the global nitrogen cycle (Fowler et al., 2013, 2015), as well as a large anthropogenic source of reactive nitrogen.I Its emission is primarily from agricultural sources (Schlesinger and Hartley 1992, Aneja et al., 2009). A large loss of reactive nitrogen from loss as ammonia (NH3), mainly from the use of animal manure and synthetic fertilizer, is a major problem in agricultural systems (Aneja et al., 2008; Bouwman et al., 2011). Nitrogen loss into the environment represents a big economic cost in lost nutrients. If the majority of the NH3 emissions can be prevented/conserved and recovered, it could save a significant amount of money (assuming approximately 33% nitrogen use efficiency by crops, the savings annually maybe about $15.9 billion (Raun and Johnson, 1999)). This problem has produced impacts not only in agricultural systems, but also in adjacent, and downwind ecosystems. NH3 reacts with other atmospheric compounds and serves as a precursor of fine particulate matter (PM2.5), which is listed as one of the six crucial atmospheric pollutant by U.S. Environmental Protection Agency (U.S. EPA). Exposure to high PM2.5 concentration can lead to adverse health effects and premature death (Kwok et al., 2013). It can also reduce visibility and lead to regional haze (Zhuang et al., 2014). In addition, NH3 is deposited in terrestrial, aquatic and marine ecosystems, where it causes ecosystem degradation (Bobbink et al., 2010). Through indirect microbial transformations, NH3 emission also contributes to the elevated levels of N2O emission by increasing the rate of nitrogen cycling in the natural environment (Davidson, 2009). N2O is one of the most important greenhouse gases, which contributes to the global warming.

Since last century, the use of synthetic nitrogen fertilizers and cultivation of nitrogen-fixing crops has expanded significantly (Battye et al, 2017). FAO predicts that the fertilizer

usage in south Asia will more than double between 2006 and 2060. Due to these increasing agricultural activities, nitrogen compounds emitted to the atmosphere have also increased dramatically, including significant increases in NH_3 concentrations, which are seen globally especially in agricultural regions. From 2002 to 2016, there have been substantial increases in atmospheric NH_3 emissions over several of the world's major agricultural regions, e.g., US (2.61% yr-1), the European Union (EU) (1.83% yr-1), and China (2.27% yr-1) (Warner et al., 2017).

NH_3 emitted from agricultural activities has the largest contribution to the atmospheric budget of reactive N (Reis et al., 2009). There are other sources of NH_3, including biomass burning, industrial processes, vehicular emissions, and volatilization from soils and oceans. Recent studies indicate that there has been an increase in total NH_3 emission over the past few decades. The estimated global NH_3 emission for 1990 is about 54 Tg N yr^{-1} (Bouwman et al. 1997).. However, an assessment by Schlesinger and Hartley (1992) indicates the global annual sources of NH_3 in the atmosphere to be about 75 Tg of N as NH_3. It also shows that the anthropogenic sources contribute about 80 % of global NH_3. Therefore, a comprehensive review of emissions of NH_3 from various anthropogenic sources is needed, so that effective control strategies can be formulated to reduce such emissions (Behera et al, 2013).

NH_3 may be used in fertilizers either as its salts, in solution or anhydrously. Ammonium fertilizers play a crucial role in high-yield crop production and contribute to large NH_3 emission (Warner et al, 2017). Around 2% of the world energy is used to produce the annual 120Mt Nr for fertilizer and other uses, mainly as ammonia through the Haber-Bosch process (Abrol et al., 2017). The Haber-Bosch process combines atmospheric nitrogen with hydrogen under high pressures and temperatures to produce NH_3. This process, on one hand, feeds more population

with more food; but enhanced N-fixation due to human activities also impacts the nitrogen cycle. Nitrification in agricultural soils has the effect of reducing the release of NH3 to the atmosphere. Some of the NO3 produced is later denitrified. However, the contribution and increases of reactive nitrogen by the Haber-Bosch process is not fully offset by denitrification. From the global perspective (Battye et al., 2017), the global denitrification flux is estimated to be 600 Tg N y-1, which would be sufficient to balance the rate of nitrogen fixation, but there are imbalances on local and regional scales that lead to adverse environmental problems.

Accurate estimates of NH3 emission are necessary for global inventories. Global estimates are also important for developing better models to assess the impact of NH3 emission on the atmosphere and the deposition of ammonia in terrestrial ecosystems. However, the NH3 emissions estimates used in current modeling efforts are subject to considerable uncertainties (Battye et al, 2003).

There are many factors that affect NH3 emission from soil. For example, ammonia emission increases with increasing nitrogen content in fertilizer and pH in soils. It also increases exponentially with temperature (Riddick et al., 2016). NH3 emission also requires a minimum level of soil moisture for microbial activities (Warner et al., 2017).

1.3. NOx

Nitrogen oxides (NOx = NO +NO2) are important trace constituents in the troposphere that regulate the consumption and production of photochemical oxidants, ozone (O3) and hydroxy radicals (Warneck 1988). Although these gases do not directly affect the radiative balance, the formation of ozone and aerosols in the troposphere can results in negative consequence on air quality, radiative balance, dynamics, and chemistry of the atmosphere. Most of air pollution problems are caused by NOx through chemical reactions, e.g. smog, acid rain. NO2, as the most prevalent form of NOx in the atmosphere, has been listed in criteria air pollutants by U.S. Environmental Protection Agency (U.S. EPA). In the National Ambient Air Quality Standards (NAAQS), the primary and secondary standard for NO2 is 0.053 parts per million (ppm).

Tropospheric ozone pollutions is the one of the most significant air pollution problems in the United States, and most developing countries. It is harmful to both human health and vegetable and crop growth. NOx is one of the most important precursors of tropospheric ozone formation. NO2 reacts in the presence of air and sunlight to produce NO. When NO concentration is below 3-8 ppt, NO reacts with O3 to produce NO2 and O2, but when NO concentration is higher, NO catalyzes the oxidation of CH4, CO and volatile organic compounds (VOC) to produce O3 (Warneck 1988), and NO gets recycled to NO2 by free radicals. In rural environments, the reaction of NO with biogenic VOC can be a predominant source of ozone. NOx in troposphere can be captured by moisture in the form of nitric acid (HNO3) to form acid rain, which directly accelerating acidification and eutrophication processes in the regional ecosystems. Acid rain is extremely harmful to not only the ecosystem, but also some segments of our economy. Based on these facts, NOx emission needs to be reduced and well regulated.

NOx can come from both natural sources, such as lightning, soil microbial activities and biomass burning, as well as anthropogenic sources such as fossil fuel combustion. Fossil fuel combustion contributes more than half of the global NO2 budget. For NO, there are great portion of sources attributable to NO emissions from soils. The estimate of NO emission from soils can be up to 40% (Hutchinson, 1995). Published field measurements and inventories show substantial NO emissions from tropical savannas (Poth et al 1995), successional pastures (Keller and Reiners, 1994), and intensively managed agriculture (Valente and Thornton, 1993).

Many factors could control the NO emission from soil, e.g. soil temperature, soil pH, soil moisture, as well as available N into the soil. Many researchers have designed experiments to test how these factors affect the rate of NO emission rate from soil. Pilegaard (2013) points out that the most important factor that affects NO emission is the availability of nitrogen in the soil. It can come from nitrogen fixation, nitrogen deposition, or input as fertilizer/manure. In agricultural soil, NO emission is most affected by the available nitrogen from fertilizer/manure. Soil water amount controls the rate of O2 supply, which directly affects the nitrification and denitrification. Goldberg & Gebauer (2009) observed that NO emission decreased after precipitation, but increased during the drought. In addition, due to the positive effect of soil temperature on microbial processes, NO emission generally increases with soil temperature. Schindlbacher et al. (2003) indicated that there is a positive exponential relationship between NO emission and soil temperature. Although there is no direct relationship between the rate of NO emission and soil pH, nitrification process intensified with high soil pH.

With recent advanced technology, there are an increasing number of field studies and modeling w models that predict NOx emissions. Therefore, understanding the comprehensive mechanism of NO emission from soil and the knowledge of controlling factors for NO emission

have been improved. However, uncertainties for present global and regional estimates for NO emission still exist, and the estimates show a great range between the lowest and highest estimates. Also, most measurements are short-term, which do not represent the spatial and temporal variation of NO emission. This indicates a necessity to use the existing field measurement results and statistical models, with few input parameters, to estimate NO emission from agricultural soil.

1.4. Objectives

The goal of this project is to develop a statistical model to predict Nr emissions from agricultural soils containing synthetic and organic fertilizers using physicochemical properties of ecosystems from different regions. We also analyze the spatial distribution of Nr emissions from agricultural soils, and compare the results with other modeled emission inventories (i.e. EDGAR v.4.3.2, EPA, and FAOSTAT). This project offers global and regional scale estimates for the sources of N2O, NH3, and NOx from fertilizer and manure application on land used for agricultural activities for the base year 2012. Statistical models provide an innovative way to estimate Nr emissions as well as an opportunity of predict future Nr emissions in a changing world.

CHAPTER 2 Methodology

2.1. Flowchart

Figure 2.1 is a flowchart that demonstrates the methodology we have adopted to estimate NH3 global emissions from agricultural soil. Based on an initial literature review, we found four variables of importance in controlling NH3 emissions from soil: (1) soil moisture, (2) soil and air temperature, (3) synthetic and organic fertilizer usage, and (4) soil pH. We performed a series of statistical analyses utilizing the R studio statistical software (https://github.com/rstudio/rstudio) to examine the distribution of data and to fit an appropriate regression model with Nr as the response (dependent) variable and other variables as independent predictors. Lastly, we used Integrated Land and Water Information System (ILWIS) v.3.31 Academic (https://www.itc.nl/ilwis/download/ilwis33/), a GIS tool to prepare the global data sets and apply the statistical model for predicting NH3 emissions.

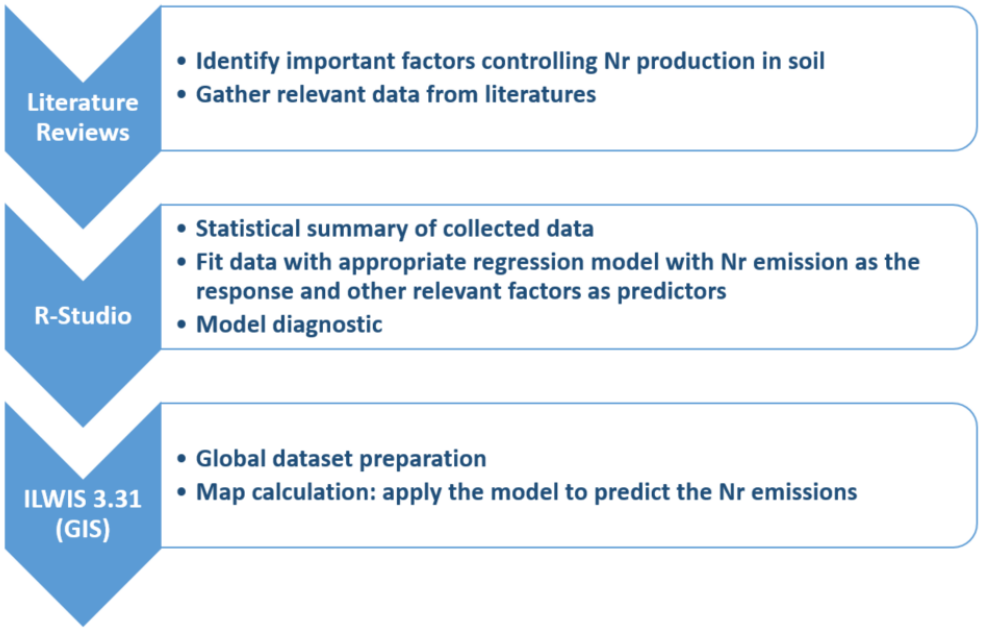

Figure 2.1 Flowchart of the methodology.

2.2. Data Collection

Data collection includes two parts, (1) data sets for developing the statistical model (Table 2.1a), and (2) global data sets for extrapolating the results to larger areas (Table 2.1b). For the statistical model, we conducted a literature review regarding agricultural soil N2O and NH3 emission (published after 2000), and NO emissions (published after 1990).

Table 2.1a. Data for statistical model development.

Parameter	Unit
N_r emissions	kg N ha^{-1} $year^{-1}$
Air temperature	°C
Soil temperature	°C
Soil pH	-
Soil moisture	%
Fertilizer N content usage	kg N ha^{-1} $year^{-1}$
Manure N content usage	kg N ha^{-1} $year^{-1}$

Table 2.1b. Global data set used for prediction.

Parameter	Data sets
Soil temperature; Soil pH	Harmonized World Soil Database v 1.2 http://www.fao.org/soils-portal/soil-survey/soil-maps-and-databases/harmonized-world-soil-database-v12/en/
Soil moisture	The Global Soil Dataset for Earth System Modeling http://globalchange.bnu.edu.cn/research/soilw
Air temperature	ERA-Interim Global Atmospheric Reanalysis https://www.ecmwf.int/en/forecasts/datasets/archive-datasets/reanalysis-datasets/era-interim
Cropland cover, fertilizer usage, manure usage	Adjusted EarthStat's Cropland and Pasture Area http://www.earthstat.org/

2.3. Model Setup

2.3.1. N2O

Based on the statistical analysis, we found that the data distribution, shown as the histogram of N2O emissions (Figure 2.2a), was skewed to the right. To normalize the data, we transformed the data to its logarithmic value (Figure 2.2b) and determined that the data were then close to a normal distribution.

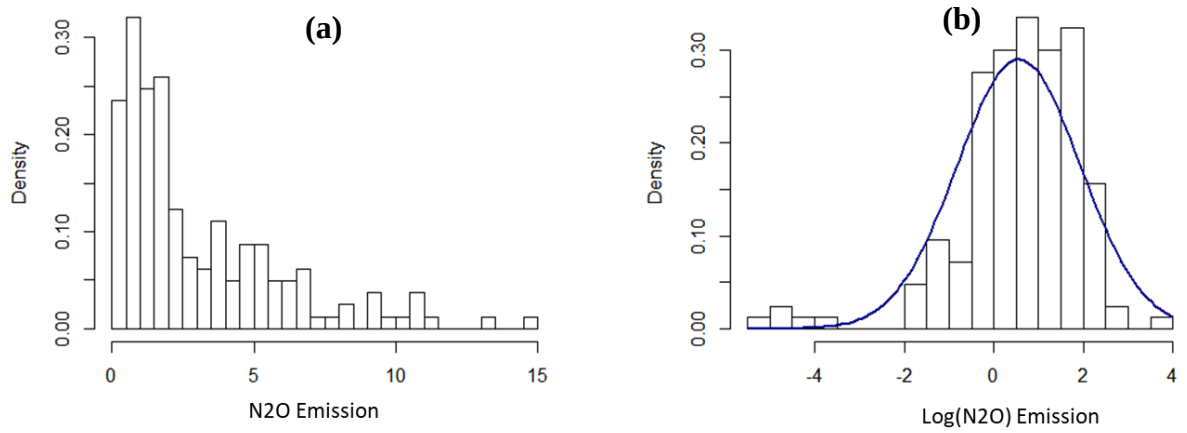

Figure 2.2. (a) Histogram of N_2O emission; and (b) log of N_2O emission.

Then, we used a multiple linear regression model to fit the response variable (the log of N2O emissions), using the physicochemical variables as predictors. The statistically derived model (hereinafter: N2O_STAT) to predict N2O emissions from agricultural soils is mathematically expressed as the following:

$$\boldsymbol{N_2O\ emission} = (\exp\left[A + B \times T_{soil} + C \times SM + D \times pH_{soil} + E \times N\ input + F \times Fertilizer\ type\right]) \times \frac{28}{44} \quad (1)$$

Where, Tsoil refers to soil temperature (°C), SM soil moisture (%), and the coefficients A, B, C, D, E, and F are statistically derived parameters (Table 2.2). N input is differentiated by synthetic or organic fertilizer, and is expressed as kg N ha-1 yr-1. The units for predicted N2O

emission are kg N ha-1 yr-1. Initially, air temperature was included as a variable in the model, but it was later omitted from the equation as it has insignificant correlation with N2O emission.

Table 2.2 summarizes the coefficients and p-values of each variable. Based on the p-value, the soil moisture, soil pH, fertilizer usage and fertilizer type are statistically significant. The Residual standard error is 0.928, and R-squared is 0.2. The F-test shows that this multivariate linear regression model is statistically significant at 90% confidence level.

Table 2.2 Summary of the N2O_STAT model.

Variable	Parameter	Coefficient	P-value
A	Intercept	1.3437	0.0295
B	Soil temperature	0.0291	0.0515
C	Soil moisture	0.0196	0.0003
D	Soil pH	-0.3454	0.0007
E	Nitrogen input	0.0003	0.5802
F	Fertilizer type	0.4567	0.0073

2.3.2. NH3

Similar for NH3, we found that the data distribution, shown as the histogram of NH3 emissions (Figure 2.3a), was skewed to the right. To normalize the data, we transformed the data to its logarithmic value (Figure 2.3b) and determined that the data were then close to a normal distribution.

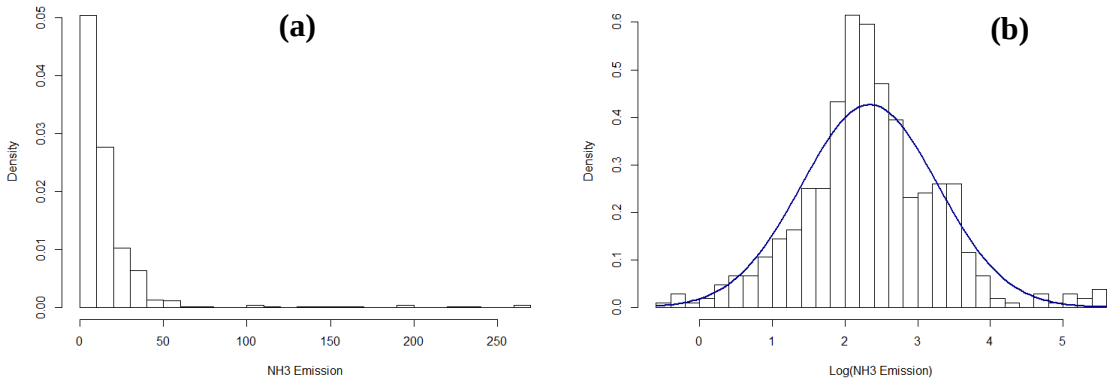

Figure 2.3. (a) Histogram of NH_3 emission; and (b) log of NH_3 emission.

Then, using the same methodology, we used a multiple linear regression model to fit the response variable (the log of NH3 emissions), using the physicochemical variables as predictors. The statistically derived model (hereinafter: NH3_STAT) to predict NH3 emissions from agricultural soils is mathematically expressed as the following:

$$\boldsymbol{NH3\ emission} = (\exp\left[\boldsymbol{A} + \boldsymbol{B} \times \boldsymbol{T_{soil}} + \boldsymbol{C} \times \boldsymbol{SM} + \boldsymbol{D} \times \boldsymbol{pH_{soil}} + \boldsymbol{E} \times \boldsymbol{log(N\ input)} + \boldsymbol{F} \times \boldsymbol{Fertilizer\ type}\right]) \times \frac{\boldsymbol{14}}{\boldsymbol{17}} \qquad (2)$$

Where, Tsoil refers to soil temperature (°C), SM soil moisture (%), and the coefficients A, B, C, D, E, and F are statistically derived parameters (Table 2.3). Table 2.3 summarizes the coefficients and p-values of each variable. Based on the p-value, the soil moisture, soil pH, fertilizer usage and fertilizer type are statistically significant. The Residual standard error is 0.743, and R-squared is 0.4. F-test shows that this multivariate linear regression model is statistically significant (99% confidence level).

Table 2.3 Summary of the NH3_STAT model.

Variable	Parameter	Coefficient	P-value
A	Intercept	-4.641412	< 2e-16
B	Soil temperature	0.015031	0.020691
C	Soil moisture	0.006613	0.008920
D	Soil pH	0.086288	0.04883
E	Log(Nitrogen input)	1.166524	< 2e-16
F	Fertilizer type	0.508622	0.000569

2.3.3. NOx

Similar to previous Nr gases, we found that the data distribution, shown as the histogram of NOx emissions (Figure 2.4a), was skewed to the right. To normalize the data, we transformed the data to its logarithmic value (Figure 2.4b). However, the distribution of log(NO) not appear as a normal distribution. This might due to the relatively small data set. Most data are located between -2 and 2. Therefore, we assume the data are normally distributed.

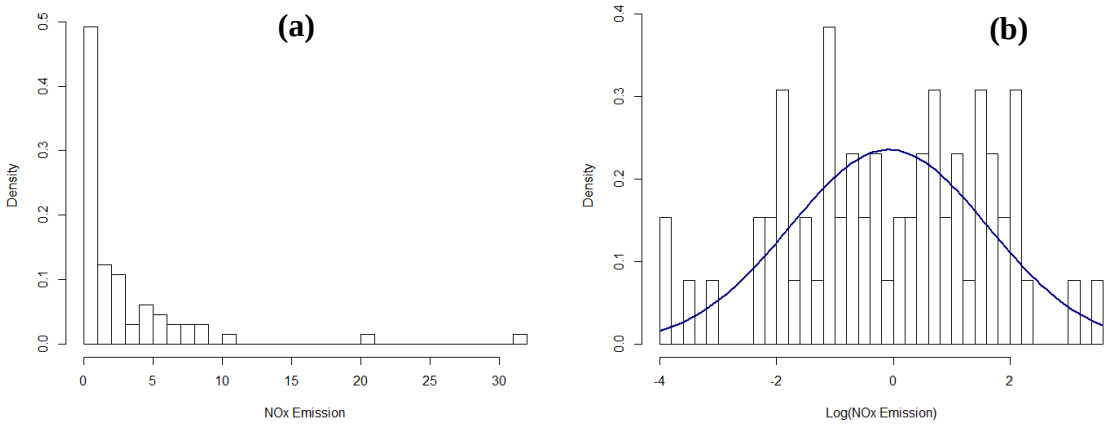

Figure 2.4. (a) Histogram of NOx emission; and (b) log of NOx emission.

Then, we used a multiple linear regression model to fit the response variable (the log of NOx emissions), using the physicochemical variables as predictors. The statistically derived model (hereinafter: NO_STAT) to predict NOx emissions from agricultural soils is mathematically expressed as the following:

$$\boldsymbol{NO\ emission} = (\exp\left[\boldsymbol{A} + \boldsymbol{B} \times \boldsymbol{T_{soil}} + \boldsymbol{C} \times \boldsymbol{SM} + \boldsymbol{D} \times \boldsymbol{pH_{soil}} + \boldsymbol{E} \times \boldsymbol{log(N\ input)} + \boldsymbol{F} \times \boldsymbol{Fertilizer\ type}\right]) \times \frac{\mathbf{14}}{\mathbf{30}} \qquad (3)$$

Where, Tsoil refers to soil temperature (°C), SM soil moisture (%), and the coefficients A, B, C, D, E, and F are statistically derived parameters (Table 2.4). Table 2.4 summarizes the coefficients and p-values of each variable. Based on the p-value, the soil moisture, soil pH, fertilizer usage and fertilizer type are statistically significant. The Residual standard error is

1.345, and R-squared is 0.4. F-test shows that this multivariate linear regression model is statistically significant (99% confidence level).

Table 2.4 Summary of the NO_STAT model.

Variable	Parameter	Coefficient	P-value
A	Intercept	-6.161950	0.0140
B	Soil temperature	0.022040	0.3375
C	Soil moisture	0.017233	0.0436
D	Soil pH	- 0.132707	0.5883
E	Log(Nitrogen input)	1.229934	1.61e-05
F	Fertilizer type	-0.077396	0.9241

2.4. Model Diagnostics

After running the regression analysis, we verified that the three models work well for fitting the data and comply with the assumptions of linear regression models. A two-step model diagnostic was performed by analyzing the variance and distribution of the residuals (Figure 2.5, Figure 2.6, and Figure 2.7). The equal distribution with no distinct patterns of residuals around the horizontal provides a good indication the likelihood of a linear relationship. Additionally, the residuals are well fitted on the straight line, indicating that the data are indeed normally distributed. The outliers indicated by residuals that are far from the line are excluded in further analysis.

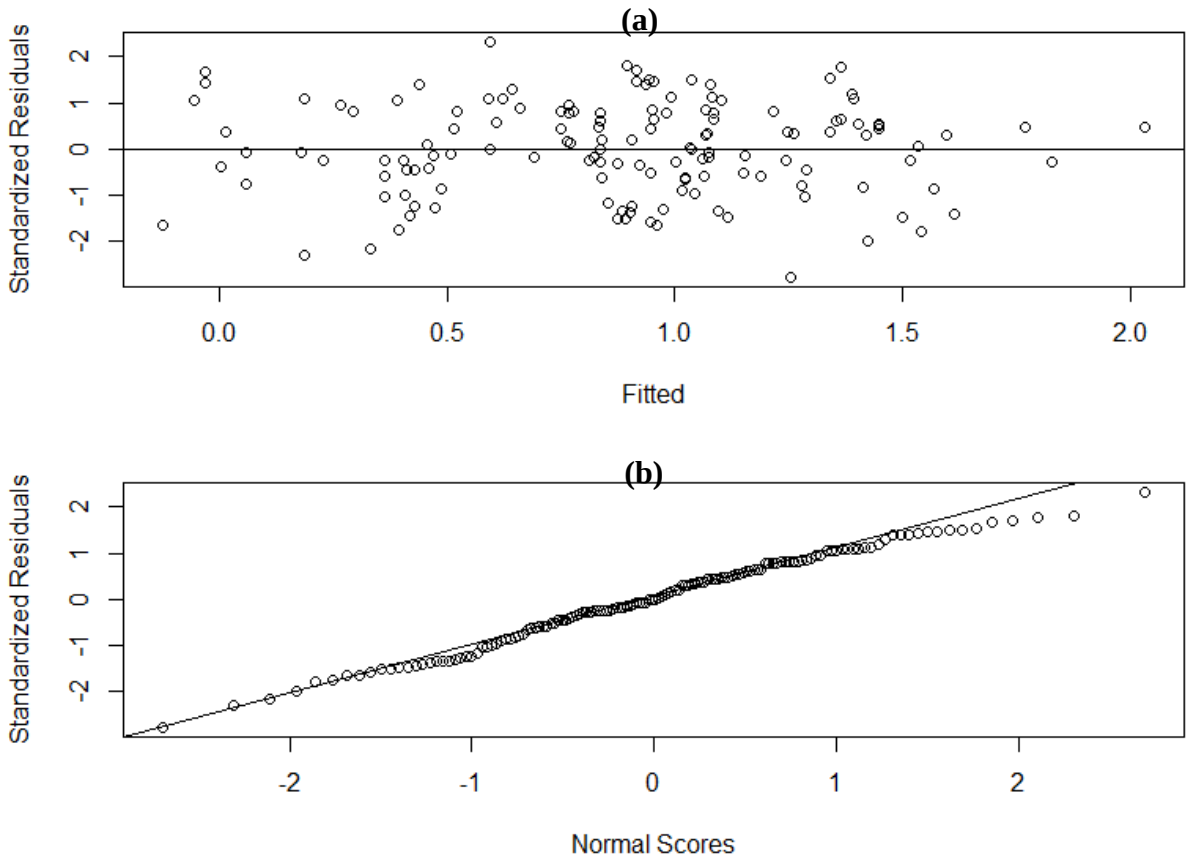

Figure 2.5. N2O Model diagnostic: (a) Standardized residual; (b) Q-Q Plot.

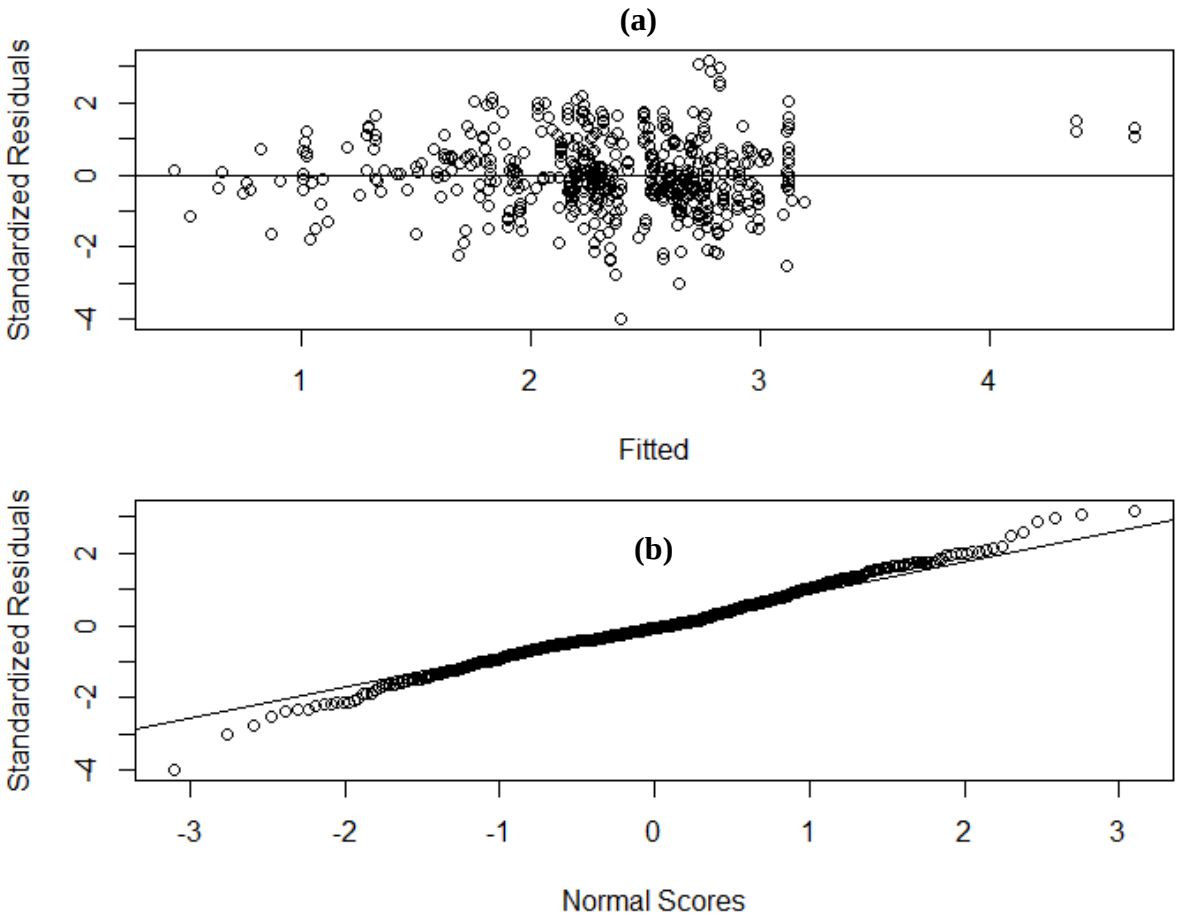

Figure 2.6. NH3 Model diagnostic: (a) Standardized residual; (b) Q-Q Plot.

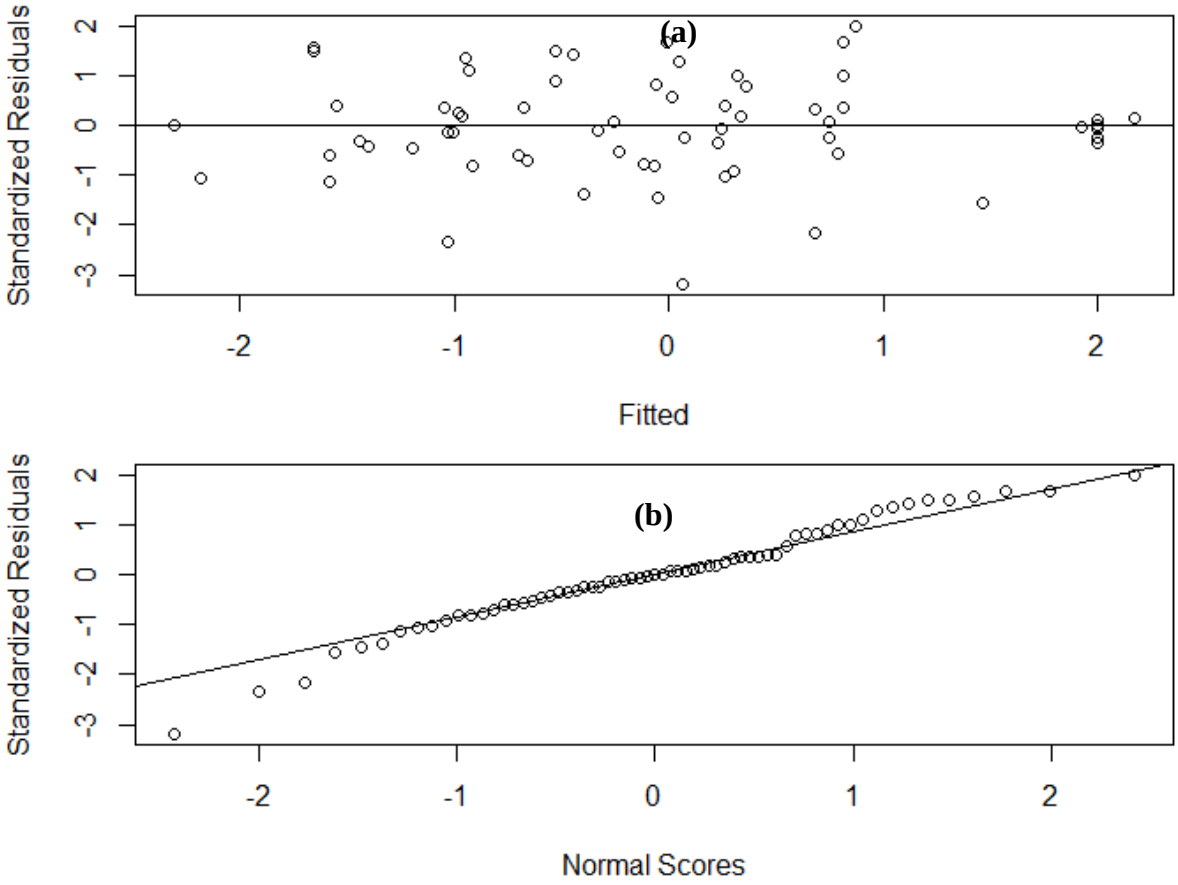

Figure 2.7. NOx Model diagnostic: (a) Standardized residual; (b) Q-Q Plot.

We tested different model forms, i.e., linear form (classical emission factor approach) and exponential form (this study). The exponential form (r2 = 0.2, and residual standard error = 0.93) while the linear form (r2 = 0.14, and residual standard error = 1.02), suggesting that the exponential model performs slightly better in modeling the reactive nitrogen emission.

2.5. Other Emission Inventories

EDGAR: The Emission Database for Global Atmospheric Research compiles anthropogenic global N2O, NH3, NO emissions and trends from 1970 to 2012 based on international statistics and emission factors (Janssens-Maenhout et al. 2017). The activity data for agricultural sectors originate mainly from International Fertilizer Industry Association (IFA)

and the Food and Agriculture Organization Statistics Division (FAOSTAT). The N_2O emission factor for direct soil emissions from the use of synthetic fertilizer and from manure and from crop residues is based on the 2006 IPCC Guidelines (IPCC, 2006). For the comparison purposes, we use the 2012 global N_2O data set for agricultural soils (subsector 4C+4D). However, for NH3 and NOx, we use the 2012 global data set for agriculture sectors (subsector 4B+4C+4D+4F), which includes Enteric fermentation, Manure management, Rice cultivation, and Agricultural soils.

FAOSTAT: Food and Agriculture organization of the United Nations develops methods and standards for food and agriculture statistics, provides technical assistance services and disseminates data for global monitoring. It provides the N2O global emissions from synthetic fertilizers, manure applied to soils, manure left on pasture, and crop residues in 2012.

EPA greenhouse gas inventory report and USGS county-level data sets: We calculated N2O emission from agriculture soils over the continental U.S. based on subsectors described in the EPA report (EPA, 2018). The subsector emissions are taken from the county-level information compiled by USGS from the 2012 census of agriculture (Gronberg and Spahr, 2012; Gronberg and Arnold, 2017). The county-level information consists of fertilizer usage, manure applications, and crop residue. Then we calculated the emissions which are interpolated to generate a spatial distribution of the emission for the United States.

EPA National Emission Inventory 2014 version 2: We calculated NH3 emission from agricultural soils over the continental U.S. based on the county-level emission data (EPA, 2018). We selected the emission from selected source classification codes (SCCs) listed on Table 2.5. The information consists of NH3 emissions from different types of fertilizer, manure from different types of animals, and fractions from unclassified sources.

Table 2.5. Selected NEI2014 v.2 source classification codes (SCCs) included in this study.

SCC	Item	Source
2801700001	Anhydrous Ammonia	Fertilizer Application
2801700002	Aqueous Ammonia	Fertilizer Application
2801700003	Nitrogen Solutions	Fertilizer Application
2801700004	Urea	Fertilizer Application
2801700005	Ammonium Nitrate	Fertilizer Application
2801700006	Ammonium Sulfate	Fertilizer Application
2801700007	Ammonium Thiosulfate	Fertilizer Application
2801700008	Other Straight Nitrogen	Fertilizer Application
2801700009	Ammonium Phosphates	Fertilizer Application
2801700010	N-P-K (multi-grade nutrient fertilizers)	Fertilizer Application
2801700011	Calcium Ammonium Nitrate	Fertilizer Application
2801700012	Potassium Nitrate	Fertilizer Application
2801700013	Diammonium Phosphate	Fertilizer Application
2801700014	Monoammonium Phosphate	Fertilizer Application
2801700015	Liquid Ammonium Polyphosphate	Fertilizer Application
2801700099	Miscellaneous Fertilizers	Fertilizer Application
2805001300	Land application of manure	Beef cattle - finishing operations on feedlots (drylots)
2805002000	Not elsewhere classified	Beef cattle production composite (93%)
2805007300	Land application of manure	Poultry production - layers with dry manure management systems
2805008300	Land application of manure	Poultry production - layers with wet manure management systems
2805009300	Land application of manure	Poultry production - broilers
2805010300	Land application of manure	Poultry production - turkeys
2805018000	Not elsewhere classified	Dairy cattle production composite (26%)
2805019300	Land application of manure	Dairy cattle - flush dairy
2805021300	Land application of manure	Dairy cattle - scrape dairy
2805022300	Land application of manure	Dairy cattle - deep pit dairy
2805023300	Land application of manure	Dairy cattle - drylot/pasture dairy
2805025000	Not elsewhere classified	Swine production composite (4%)
2805030000	Not elsewhere classified	Poultry production composite (7%)
2805035000	Not elsewhere classified	Horse production composite (26%)
2805039300	Land application of manure	Swine production - operations with lagoons (unspecified animal age)
2805045000	Not elsewhere classified	Goats production composite (26%)
2805047300	Land application of manure	Swine production - deep-pit house operations (unspecified animal age)

CHAPTER 3 Characteristic of N2O Emission

3.1. Global

After calculating the coefficients of all variables, we applied the N2O_STAT model for each grid cell and generated a global map for N2O emissions. Figure 3.1(a) gives the spatial distribution of global N2O emission from agricultural soils calculated using N2O_STAT in kg N2O yr-1 grid cell-1. The resolution of this map is 5 arc-minute by 5 arc-minute, which is equivalent to 8464 ha. The average N2O emission from agricultural soils is ~9000 kg yr-1 grid cell-1, which is comparable to average values of 1 kg ha-1 for agricultural soils, reported by Kim et al. (2013) and Shcherbak et al. (2014). Figure 3.1(b) is spatial distribution of global N2O emission from agricultural soil based on EDGAR. Total annual global N2O emissions from N2O_STAT and EDGAR are 3.06 Tg N yr-1 and 4.49 Tg N yr-1, respectively. It is encouraging to see that, in general, the model captures the global spatial pattern in N2O emissions well. Figure 3.1 (c) shows the absolute difference between N2O_STAT and EDGAR. In comparison with EDGAR, N2O_STAT gives relatively lower N2O emission values in North America, South America, India, and China, while higher values in Eastern Europe and mid Africa.

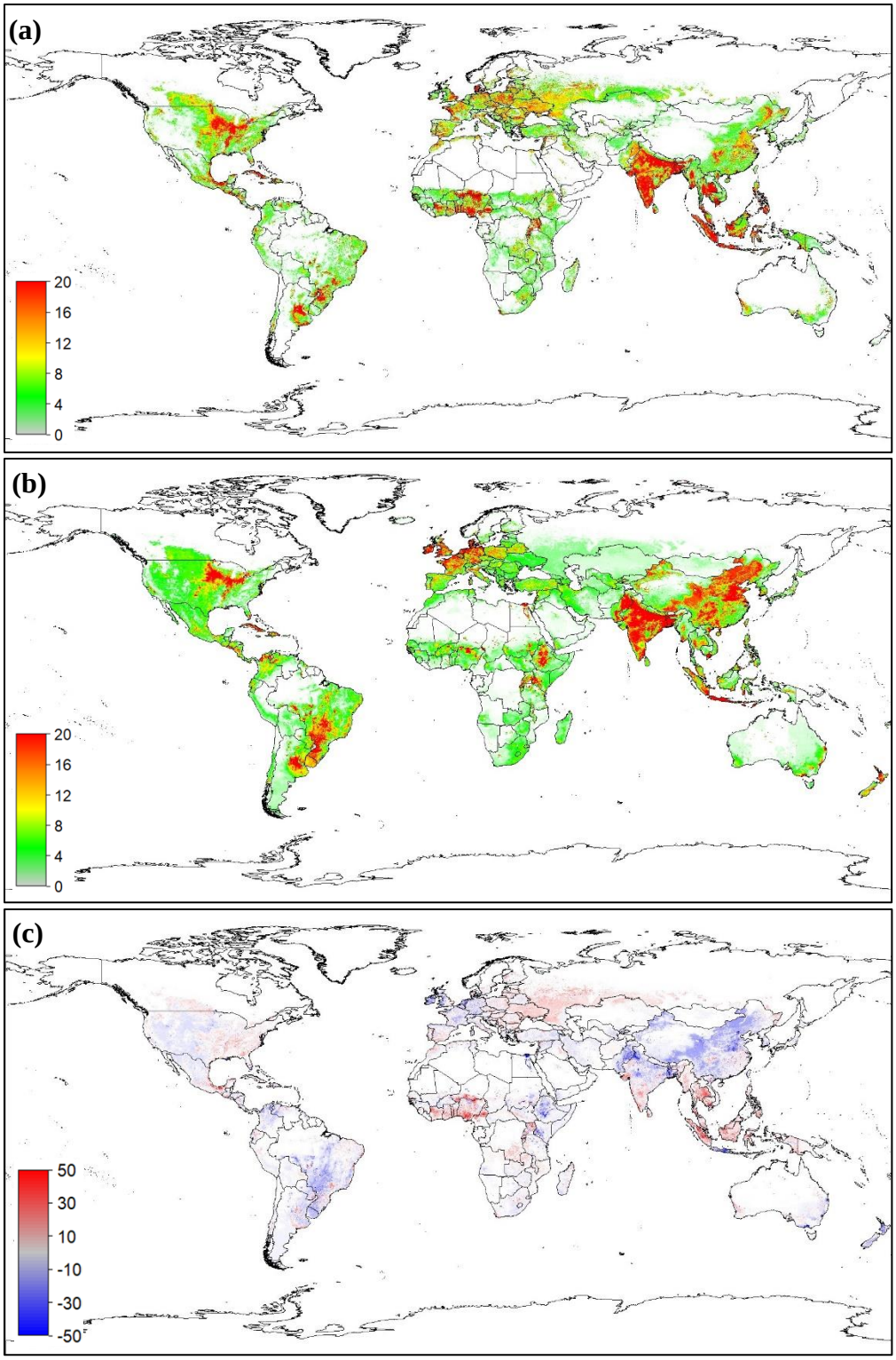

Figure 3.1. Comparison between the results from (a) N_2O_STAT, (b) EDGAR, and (c) absolute difference between the two models (in ton N_2O yr^{-1} grid $cell^{-1}$).

3.2. Regional

3.2.1. Continental U.S.

Figure 3.2 provides the comparison between the results from N2O_STAT, EDGAR, and EPA/USGS over the continental U.S. (hereafter listed as U.S). Total N2O emissions from agricultural soils in the U.S. calculated from N2O_STAT, EDGAR, and EPA/USGS are 0.35 Tg N yr-1, 0.43 Tg N yr-1, and 0.46 Tg N yr-1, respectively. Our model performs well in capturing the spatial pattern as well as the total annual emission. Figure 3.3 gives the absolute difference of N2O emissions between N2O_STAT model and EDGAR, and between N2O_STAT and EPA/USGS. Based on Figure 3.3(a), N2O_STAT underestimates N2O emission in the Midwest Region of the U.S., and overestimates emissions in the southeastern region, compared with EDGAR. Figure 3.3(b) shows that N2O_STAT gives lower N2O emission values in northern Texas, California, Florida, and states around Lake Michigan, while in other areas the values are comparable.

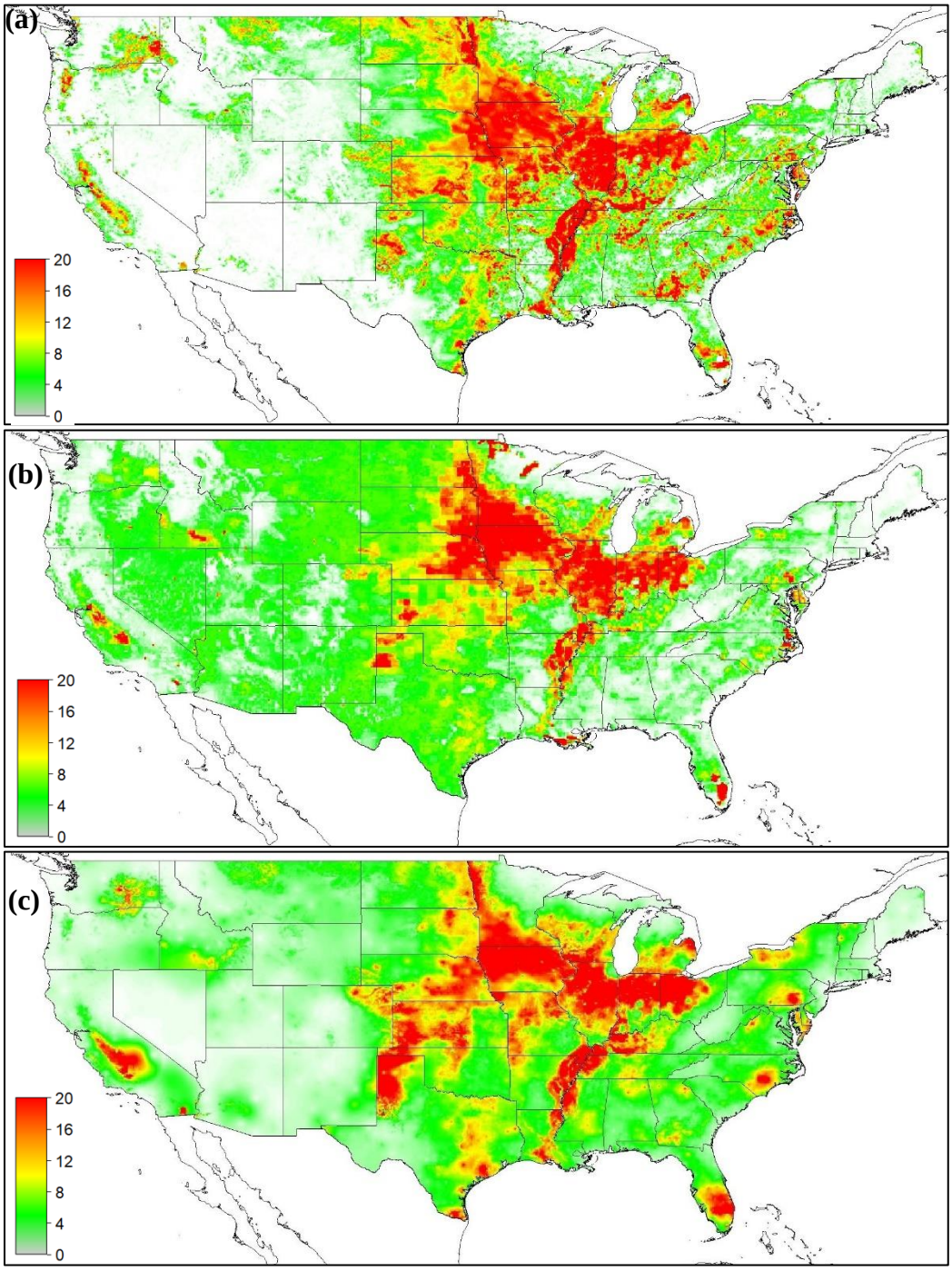

Figure 3.2. Comparison between the results from (a) N_2O_STAT, (b) EDGAR, and (c) EPA/USGS (in ton N_2O yr^{-1} grid $cell^{-1}$).

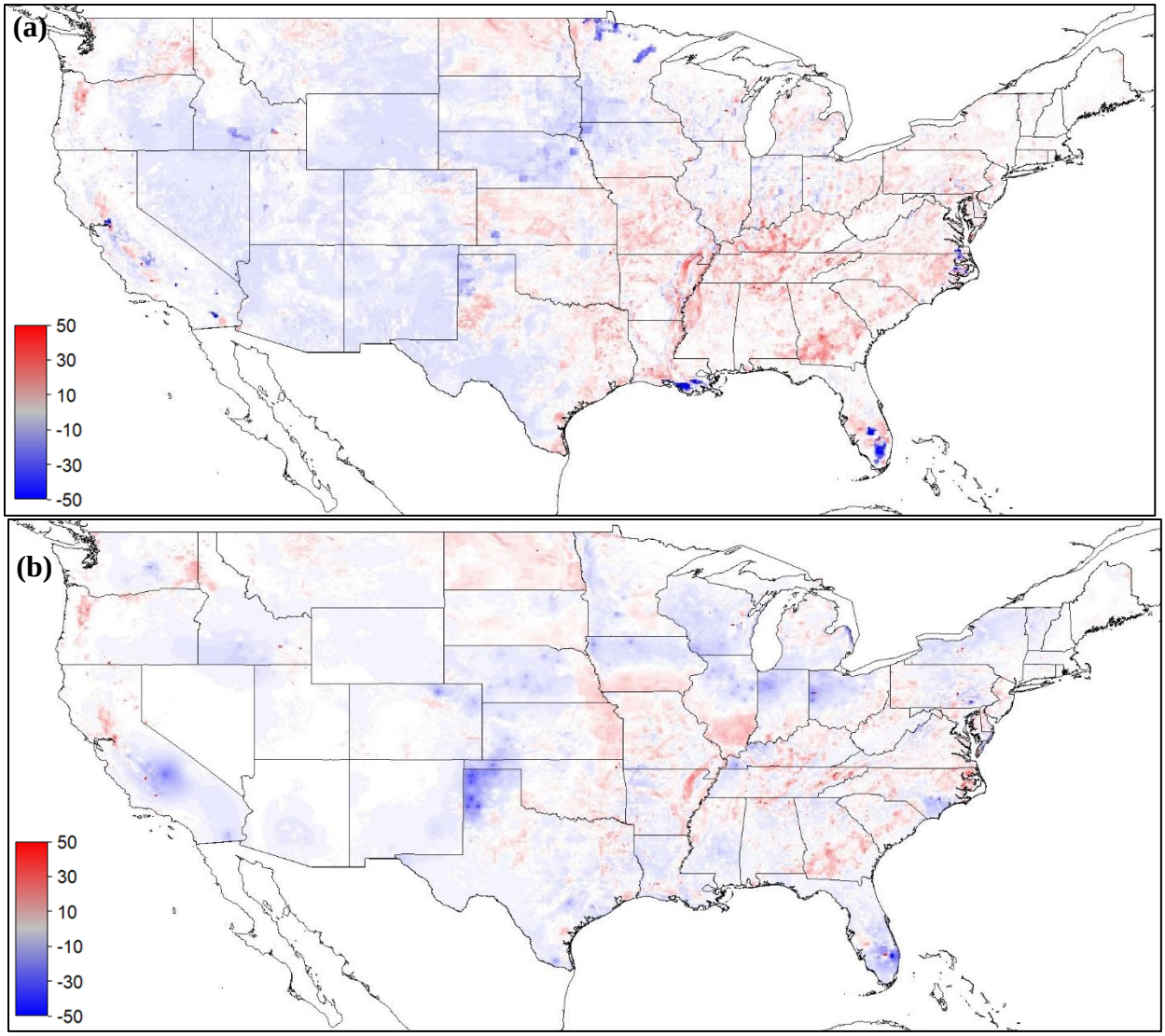

Figure 3.3. Absolute difference of N_2O emissions over CONUS between (a) N_2O_STAT and EDGAR, (b), between N_2O_STAT and EPA/USGS (in ton N_2O yr^{-1} grid $cell^{-1}$).

3.2.2. India

Figure 3.4 and Table 3.1 show close agreement between the N2O_STAT estimate of agricultural soil emissions for India when compared with EDGAR (~ -14%), the finely-resolved N2O emission estimate by Aneja et al. (2012) (~17%); and FAOSTAT (~ -7%). Moreover, Table 3.1 compares the finely-resolved N2O emission for India with other published studies. The Aneja et al. (2012) results are higher than the results of Garg et al. (2006) and Sharma et al.

(2011). The reason might be due to the selection of higher emission factors in Aneja et al. (2012) study. Overall, we believe that Aneja et al. (2012) is more appropriate in the sense that the researchers used the activity level data at district levels and chose the emission factors suitable for Asian context

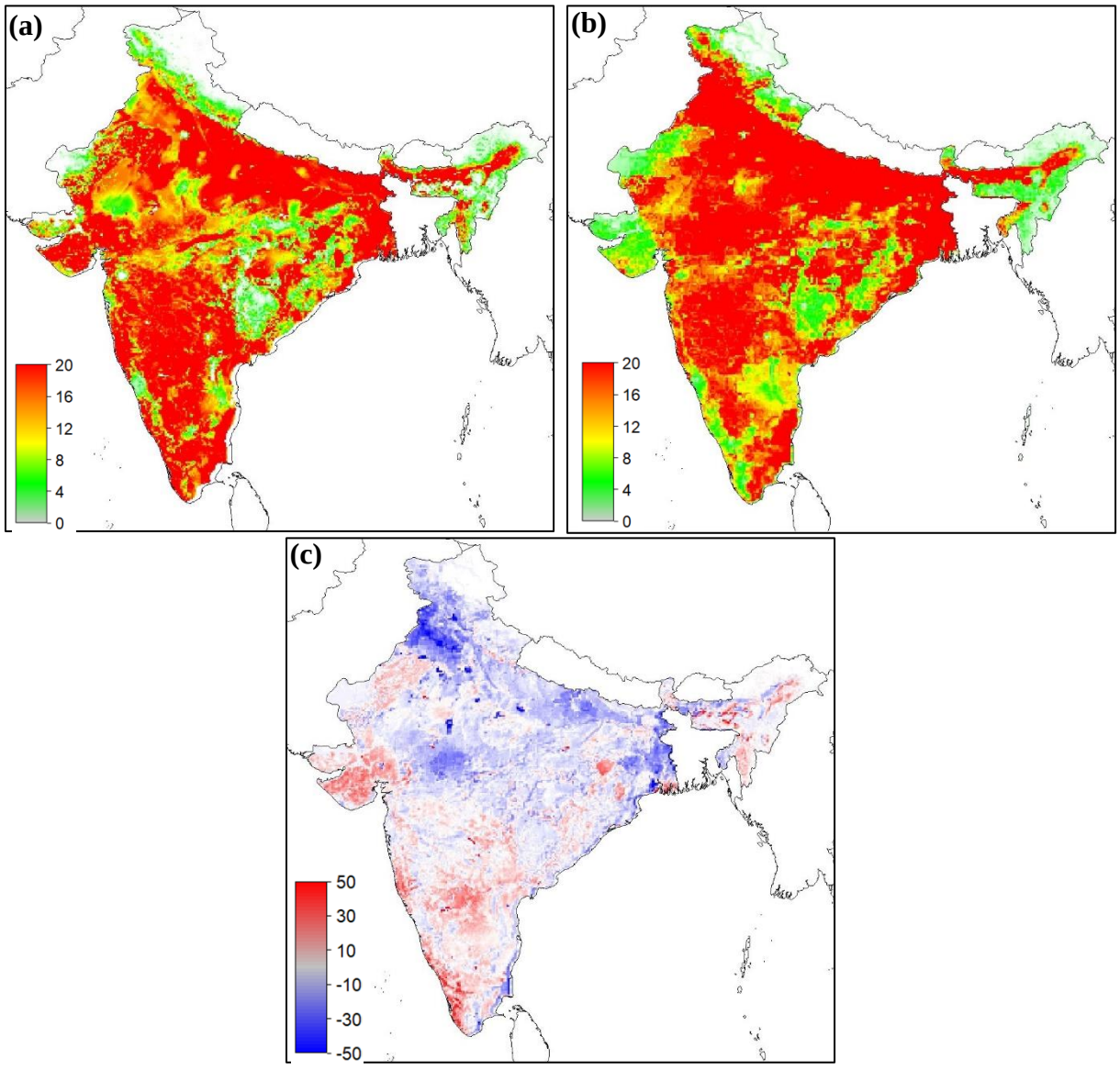

Figure 3.4. Comparison between the results from (a) N_2O_STAT, (b) EDGAR, and (c) the absolute difference between N_2O_STAT and EDGAR (in ton N_2O yr^{-1} grid $cell^{-1}$).

3.2.3. China

For China, our estimate of N2O emissions is 2.3-2.8 times lower than those estimated by EDGAR and FAO. Our N2O emission estimate, however, is in better agreement with Gao et al. (2011) and Zhou et al. (2014), being 2% higher and 38% lower, respectively. These two studies employ different methods in estimating N2O emission from Chinese agricultural soils. Gao et al. (2011) studied N2O direct emissions from croplands by using localized emission factors; while Zhou et al. (2014), in addition to using regional emission factors, utilized high-resolution activity data and localized parameters.

Table 3.1. Summary of N_2O emission from different inventories.

Emission inventory	N_2O (Tg N yr^{-1})			
	Global	**US**	**India**	**China**
N_2O_STAT	3.75	0.400	0.412	0.300
EDGAR	4.49	0.432	0.468	0.832
FAOSTAT	4.07	0.350	0.440	0.686
EPA/USGS	-	0.457	-	-
EPA Report	-	0.529	-	-
Aneja et al. (2012)	-	-	0.344	-
Garg et al. (2006)	-	-	0.181	-
Sharma et al. (2011)	-	-	0.226	-
Gao et al. (2011)	-	-	-	0.294
Zhou et al. (2014)	-	-	-	0.414

3.3. Conclusions

Table 3.1 summarizes the data comparison between N2O_STAT, EDGAR, EPA/USGS, and FAOSTAT for N2O emission from agricultural soils for the U.S., India, China, and globally. The FAOSTAT, EDGAR and EPA estimates are slightly different, although statistically consistent given the large uncertainties in the IPCC default methodologies (Tubiello et al., 2013). Tubiello et al. (2013) compared N2O emission from four databases. The results show that the

EPA 2006 and EPA 2011 provided the highest values while FAOSTAT gave the lowest, consistent with our study.

The region-based analysis (U.S., India, and China) indicates that our model gives lower estimates than other data sets. The global N2O agricultural soil emission from N2O_STAT estimate is 9-20% lower than EDGAR and FAOSTAT. The major differences, as observed in Figure 6(c), are most prominent in India and China. In India, negative differences are depicted in Figure 9(c) in the northern and central regions while the positive deviations are mainly to the west and south of the subcontinent. However, the finely-resolved agricultural emissions for N2O (Aneja et al, 2012), as well as EDGAR and FAOSTAT, agrees well with N2O_STAT (< 20% differences). It is to be noted that the estimate from Aneja et al. (2012) was calculated for 2003. Despite this difference, there are similarities found between these two data sets with respect to the areas from which high emissions are estimated. Such similarities are not observed when we compared our model estimate with EDGAR.

The underestimation of N2O emissions in comparison to other data sets and the fact that N2O_STAT may be still missing some key N inputs to its variables should be taken into account in the future work. Another component worth investigating is soil biological activity that would more fully represent the processes governing the N2O emissions.

Supplementary information

Table 1S. List of publications from where N_2O emissions and physicochemical parameters were obtained.

Soil type	Air temperature (°C)	Soil temperature (°C)	Soil moisture (%)	Soil pH	Usage (kg N ha^{-1})	Fertilizer type (0:synthetic fertilizer; 1: manure)	Annual N_2O emission (kg N_2O ha^{-1})	Reference
grassland	20	21.6	28.9	6	63.9	1	1.6	Dittert et al., 2001
grassland	12.4	14.8	79	6	100	0	1.4	Ganesan et al., 2015
grassland	11.7	14.2	64	6	100	0	0.8	Ganesan et al., 2015
grassland	2.8	6.3	82	6	100	0	0.9	Ganesan et al., 2015
grassland	10.2	12.9	75	6	100	0	2	Ganesan et al., 2015
grassland	12.3	14.7	77	6	100	0	2.8	Ganesan et al., 2015
grassland	2.1	5.7	91	6	100	0	0.3	Ganesan et al., 2015
paddy	35	34.9	23.1	7.6	120	0	1	Ghosh et al., 2003
upland	28	28.7	30.4	4.6	90	0	6.9	Hadi et al., 2008
upland	27	15	50	5.7	90	0	1.9	Jumadi et al., 2008
grassland	14.8	15.6	35	4.9	1000	1	6.25	Kelly et al., 2008
grassland	18.3	20.2	40	4.9	1000	1	4.99	Kelly et al., 2008
paddy	35	34.9	24.1	7.8	140	0	0.16	Kumar et al., 2000
grassland	10.7	11.4	80	5.1	270	1	11.5	Merino et al., 2005
grassland	20.4	16.7	60	5.1	194	1	9.1	Merino et al., 2005
paddy	35	34.9	60	7.9	140	0	0.06	Majumdar et al., 2000
paddy	35	34.9	45	8.1	120	0	0.76	Malla et al., 2005
paddy	35	34.9	66.7	8	120	0	0.66	Malla et al., 2005
grassland	18.2	20	50	6.6	97	0	4.4	Menendes etl al., 2006
grassland	18.2	20	60	6.6	181	1	15.5	Menendes etl al., 2006
grassland	15.9	17.9	80	7	80	0	1.2	Merino et al., 2002
grassland	15.9	17.9	80	7	85	1	1.2	Merino et al., 2002
winter wheat	9	9.2	28.9	5.1	160	0	0.5	Misselbrook et al., 2014
silage	9.7	10.9	28.9	5.1	120	0	1.5	Misselbrook et al., 2014
silage	11.1	7.9	28.9	5.8	120	0	1.1	Misselbrook et al., 2014
winter wheat	9.1	9.9	28.9	5.8	200	0	3.1	Misselbrook et al., 2014
winter barley	8.3	8.6	28.9	5.1	160	0	1.7	Misselbrook et al., 2014
grassland	9.9	16	28.9	5.1	625	1	2.4	Misselbrook et al., 2014
grassland	9.9	14.2	28.9	5.1	488	1	1.9	Misselbrook et al., 2014
grassland	10.3	8.7	28.9	5.1	470	1	1.6	Misselbrook et al., 2014

Table 1S. (continued).

grassland	9.5	14.5	28.9	5.1	365	1	2.8	Misselbrook et al., 2014
grassland	9.6	14	28.9	5.1	181	1	2.7	Misselbrook et al., 2014
grassland	10.7	8.3	28.9	5.1	167	1	0.8	Misselbrook et al., 2014
grassland	9.8	16.3	28.9	5.1	106	1	0.8	Misselbrook et al., 2014
grassland	9.1	6.8	28.9	5.1	146	1	0.8	Misselbrook et al., 2014
maize	16.7	20	39	6.5	224	0	3.6	Adviento-Borbe et al., 2010
maize	16.7	20	39	6.5	225	1	5.5	Adviento-Borbe et al., 2010
vegetable	15	15	31.8	5.9	150	0	0.4	Baruah et al., 2010
vegatable	15	15	31.8	5.9	150	1	1.8	Baruah et al., 2010
vegetable	15	15	31.8	5.9	150	1	0.6	Baruah et al., 2010
rapeseed	13.8	15	27.6	7	120	0	2.48	Asgedom et al., 2014
rapeseed	13.8	15	27.6	7	129	1	1.19	Asgedom et al., 2014
maize	4.2	7.6	60	6.3	171	1	8.2	Chantigny et al., 2012
maize	4.2	7.6	60	6.3	146	1	4	Chantigny et al., 2012
maize	4.2	7.6	60	6.3	130	1	6.4	Chantigny et al., 2012
maize	4.2	7.6	60	6.3	109	1	2.4	Chantigny et al., 2012
maize	4.2	7.6	60	6.3	149	1	2.5	Chantigny et al., 2012
maize	4.2	7.6	60	6.3	137	1	3.8	Chantigny et al., 2012
maize	4.2	7.6	60	6.3	124	1	1.7	Chantigny et al., 2012
maize	4.2	7.6	55	6.7	151	1	4	Chantigny et al., 2012
maize	4.2	7.6	55	6.7	148	1	4.7	Chantigny et al., 2012
maize	4.2	7.6	55	6.7	136	1	3.5	Chantigny et al., 2012
maize	4.2	7.6	55	6.7	151	1	2.3	Chantigny et al., 2012
maize	4.2	7.6	55	6.7	149	1	1.8	Chantigny et al., 2012
maize	4.2	7.6	55	6.7	156	1	2.8	Chantigny et al., 2012
maize	4.2	7.6	55	6.7	156	1	1.3	Chantigny et al., 2012
maize	1.5	5.2	40	5.9	150	1	4.4	Chen et al, 2014
maize	1.5	5.2	40	5.9	150	1	3.14	Chen et al, 2014
maize	1.5	5.2	40	5.9	150	0	2.82	Chen et al, 2014
maize	19.5	21.1	14.4	6.7	224	0	0.7	Collin et al, 2011
maize	18.9	20.6	15.6	6.7	224	0	1.4	Collin et al, 2011
maize	19.5	21.1	14.4	6.7	224	1	1.1	Collin et al, 2011
maize	18.9	20.6	15.6	6.7	224	1	1.4	Collin et al, 2011

Table 1S. (continued).

wheat and maize	13.9	15.7	44.9	8	150	0	4.4	Ding et al, 2013
wheat and maize	13.9	15.7	44.9	8	150	0	3.46	Ding et al, 2013
wheat and maize	13.9	15.7	44.9	8.1	150	1	3.77	Ding et al, 2013
maize	19.7	21.3	51.1	6.2	230	0	3	Guo et al, 2013
maize	19.5	21.1	46.7	6.2	230	0	1.8	Guo et al, 2013
maize	20	21.1	37.2	5.9	120	0	1.7	Li et al., 2013
grassland	5.6	8.8	39	5.8	98	0	0.3	Shimizu et al., 2013
grassland	6.1	9.2	39	5.8	100	0	0.6	Shimizu et al., 2013
grassland	6.2	9.3	39	5.8	100	0	1	Shimizu et al., 2013
grassland	6.1	9.2	39	5.8	162	0	0.5	Shimizu et al., 2013
grassland	6.2	9.3	39	5.8	157	0	1	Shimizu et al., 2013
grassland	6.5	9.6	39	5.8	160	0	5.5	Shimizu et al., 2013
grassland	5.6	8.8	39	5.8	208	1	0.6	Shimizu et al., 2013
grassland	6.1	9.2	39	5.8	271	1	1.8	Shimizu et al., 2013
grassland	6.2	9.3	39	5.8	242	1	9.5	Shimizu et al., 2013
grassland	6.1	9.2	39	5.8	162	1	0.7	Shimizu et al., 2013
grassland	6.2	9.3	39	5.8	203	1	0.7	Shimizu et al., 2013
grassland	6.5	9.6	39	5.8	121	1	0.7	Shimizu et al., 2013
grassland	8.4	11.3	58	5.3	164	0	2.8	Shimizu et al., 2013
grassland	8.1	11	58	5.3	183	0	2.9	Shimizu et al., 2013
grassland	9.1	11.9	58	5.3	74	0	1.5	Shimizu et al., 2013
grassland	8.3	11.2	58	5.3	74	0	2.1	Shimizu et al., 2013
grassland	8.6	11.5	58	5.3	91	0	1.2	Shimizu et al., 2013
grassland	8.4	11.3	58	5.3	236	1	3.6	Shimizu et al., 2013
grassland	8.1	11	58	5.3	310	1	4.9	Shimizu et al., 2013
grassland	9.1	11.9	58	5.3	331	1	2.2	Shimizu et al., 2013
grassland	8.3	11.2	58	5.3	308	1	0.9	Shimizu et al., 2013
grassland	8.6	11.5	58	5.3	491	1	1.4	Shimizu et al., 2013
grassland	8.4	11.3	49	5.5	90	0	2.5	Shimizu et al., 2013
grassland	8.5	11.4	49	5.5	100	0	2.4	Shimizu et al., 2013
grassland	8.5	11.4	49	5.5	89	0	5.2	Shimizu et al., 2013
grassland	8.4	11.3	49	5.5	205	1	4.8	Shimizu et al., 2013
grassland	8.5	11.4	49	5.5	241	1	1.7	Shimizu et al., 2013
grassland	8.5	11.4	49	5.5	152	1	4.9	Shimizu et al., 2013
grassland	12.3	14.7	48	5.3	209	0	4.7	Shimizu et al., 2013
grassland	11.7	14.2	48	5.3	198	0	9.1	Shimizu et al., 2013
grassland	12.6	15	48	5.3	186	0	10.1	Shimizu et al., 2013
grassland	12	14.5	48	5.3	190	0	10	Shimizu et al., 2013

Table 1S. (continued).

grassland	12.2	14.7	48	5.3	190	0	3.8	Shimizu et al., 2013
grassland	12.4	14.8	48	5.3	184	0	5.7	Shimizu et al., 2013
grassland	12.3	14.7	48	5.3	76	1	7	Shimizu et al., 2013
grassland	11.7	14.2	48	5.3	195	1	11	Shimizu et al., 2013
grassland	12.6	15	48	5.3	123	1	11.9	Shimizu et al., 2013
grassland	12	14.5	48	5.3	253	1	5.4	Shimizu et al., 2013
grassland	12.2	14.7	48	5.3	219	1	1.9	Shimizu et al., 2013
grassland	12.4	14.8	48	5.3	173	1	6.6	Shimizu et al., 2013
grassland	16.4	18.4	39	6.1	164	0	1.8	Shimizu et al., 2013
grassland	16.1	18.1	39	6.1	233	0	3.1	Shimizu et al., 2013
grassland	16.8	18.7	39	6.1	218	0	5.1	Shimizu et al., 2013
grassland	16	18	39	6.1	194	0	15.2	Shimizu et al., 2013
grassland	16.4	18.4	39	6.1	160	0	4.9	Shimizu et al., 2013
grassland	16.4	18.4	39	6.1	119	1	11.2	Shimizu et al., 2013
grassland	16.1	18.1	39	6.1	284	1	5.3	Shimizu et al., 2013
grassland	16.8	18.7	39	6.1	242	1	6.1	Shimizu et al., 2013
grassland	16	18	39	6.1	272	1	10.7	Shimizu et al., 2013
grassland	16.4	18.4	39	6.1	310	1	7.8	Shimizu et al., 2013
rapeseed	18.6	20.3	37.5	5.2	32.5	1	5.4	Nyamadzawo et al, 2014
rapeseed	18.6	20.3	37.5	5.2	60	0	12.6	Nyamadzawo et al, 2014
rapeseed	18.6	20.3	37.5	5.2	65	1	6.8	Nyamadzawo et al, 2014
rapeseed	18.6	20.3	37.5	5.2	120	0	12	Nyamadzawo et al, 2014
rapeseed	18.6	20.3	37.5	5.2	97.5	1	19.3	Nyamadzawo et al, 2014
rapeseed	18.6	20.3	37.5	5.2	240	0	112	Nyamadzawo et al, 2014
maize	4.4	7.7	50	4.6	90	0	7.4	Pelster et al, 2012
maize	4.4	7.7	50	4.6	90	1	6.7	Pelster et al, 2012
maize	4.4	7.7	50	4.6	92.2	1	6.4	Pelster et al, 2012
maize	4.4	7.7	50	4.6	88.2	1	6.9	Pelster et al, 2012
Rye	18	20	25	4.3	90	0	0.6	Sosulski et al, 2014
Rye	18	20	25	4.3	100	1	0.7	Sosulski et al, 2014
rice	23	24.2	25.2	7.9	180	0	0.2	Wang et al, 2013
rice	23	24.2	25.2	7.9	180	1	0.2	Wang et al, 2013
vegetable	17.7	22.8	15.2	4.6	144	0	4.4	Watanabe et al, 2014
wheat	12.9	10.9	17.1	5.7	90	1	2.51	Zhai et al, 2011
wheat	12.9	10.9	17.1	5.7	90	0	1	Zhai et al, 2011
wheat	12.9	10.9	17.1	5.7	90	0	1	Zhai et al, 2011
wheat	12.9	10.9	17.1	5.7	90	0	0.53	Zhai et al, 2011
maize	21.1	23.1	12.4	5.7	90	1	4.46	Zhai et al, 2011
maize	21.1	23.1	12.4	5.7	210	0	1.16	Zhai et al, 2011

Table 1S. (continued).

maize	21.1	23.1	12.4	5.7	210	0	0.84	Zhai et al, 2011
maize	21.1	23.1	12.4	5.7	210	0	0.56	Zhai et al, 2011
maize and wheat	17.3	15	60	8.3	280	0	3.14	Zhou et al, 2014
maize and wheat	17.3	15	60	8.3	280	1	5.97	Zhou et al, 2014

CHAPTER 4 Characteristic of NH3 Emission

4.1. Global

After calculating the coefficients of all variables, we applied the NH3_STAT model for each grid cell (5 arc-minute x 5 arc-minute) and generated a global map for NH3 emissions. Figure 4.1(a) gives the spatial distribution of global NH3 emission from agricultural soils calculated using NH3_STAT in kg NH3 yr-1 grid cell-1. The resolution of this map is 5 arc-minute, which is equivalent to 8464 ha. Figure 4.1(b) is spatial distribution of global NH3 emission from agricultural soil based on EDGAR. Total annual global NH3 emissions from NH3_STAT and EDGAR are 14.2 Tg N/yr and 33.3 Tg N/yr, respectively. There is a large difference between our estimate and EDGAR. This difference can be attributed to sources that are excluded in our model. Nevertheless, it is encouraging to see that, in general, the model captures the global spatial pattern in NH3 emissions well. This is understandable since NH3_STAT is exclusive to emissions from fertilizer and manure applied as fertilizer, whereas EDGAR has additional sources in their estimate. Based on our literature survey, NH3 emission from fertilizers ranges from 9 to 12 Tg N/yr, a range that is similar to our estimate.

Figure 4.1 (c) shows the absolute difference between NH3_STAT and EDGAR. In comparison with EDGAR, NH3_STAT gives relatively lower NH3 emission values in North America, South America, India, and China, while higher values in Eastern Europe.

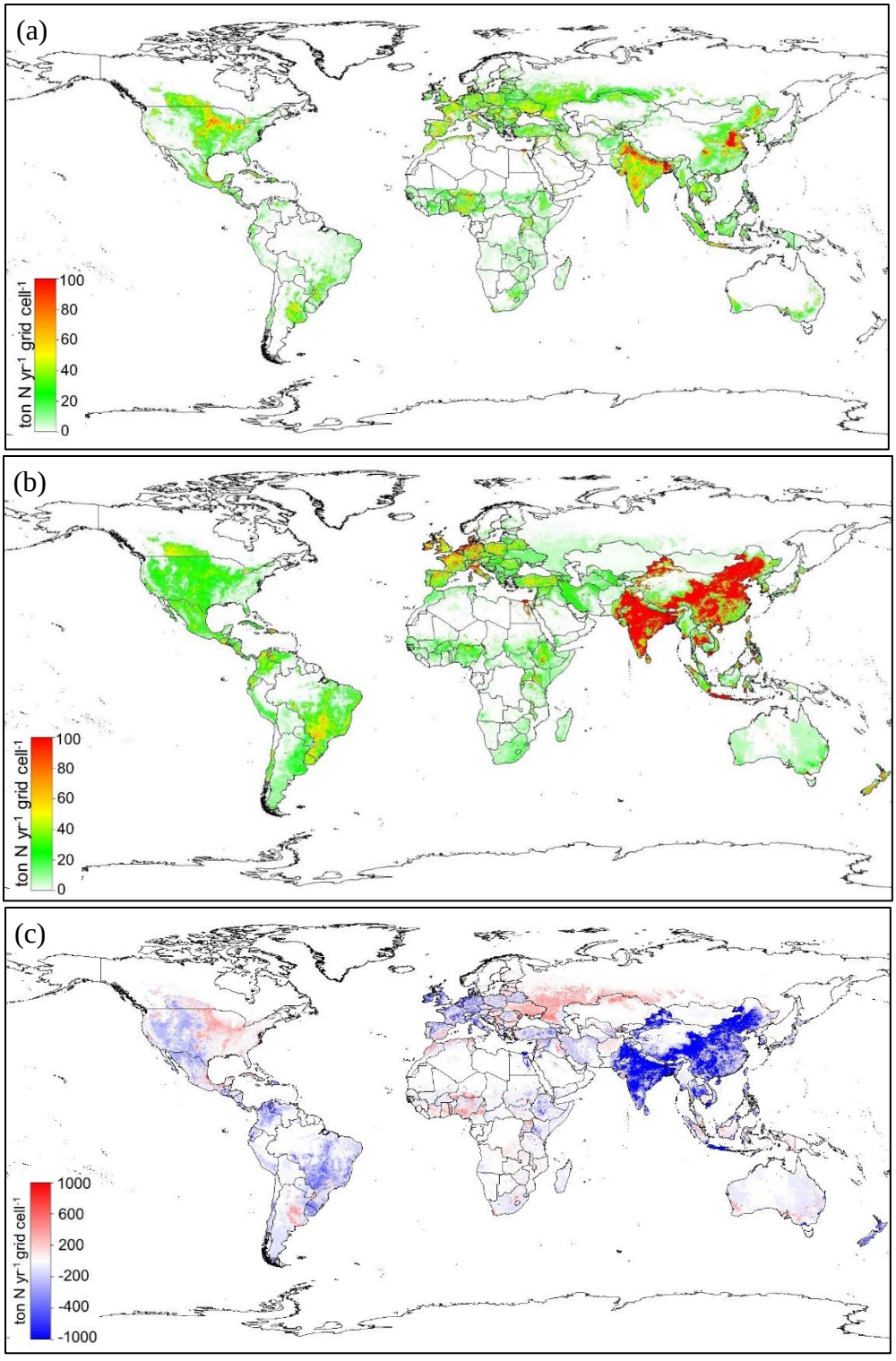

Figure 4.1. Comparison between the results from (a) NH3_STAT, (b) EDGAR, and (c) absolute difference between the two models (in ton N yr ^{1} grid cell^{-1}).

4.2. Regional

4.2.1. Continental U.S.

Figure 4.2 provides the comparison between the results from NH3_STAT over U.S. Total U.S. NH3 emission from agricultural soil calculated from NH3_STAT and EPA are 1.7 Tg N/yr and 0.7 Tg N/yr, respectively. Our model produces an estimate that is 143% higher in comparison to EPA. Our model performs well in capturing the spatial trend as well as the total annual emission. Figure 8(c) gives the absolute difference of NH3 emissions between NH3_STAT model and EDGAR, and between NH3_STAT and EPA/USGS. Based on Figure 8(a), NH3_STAT underestimates N2O emission in California and some parts in Arizona, New Mexico, and Texas, while shows overestimations for most of the U.S., compared with EPA.

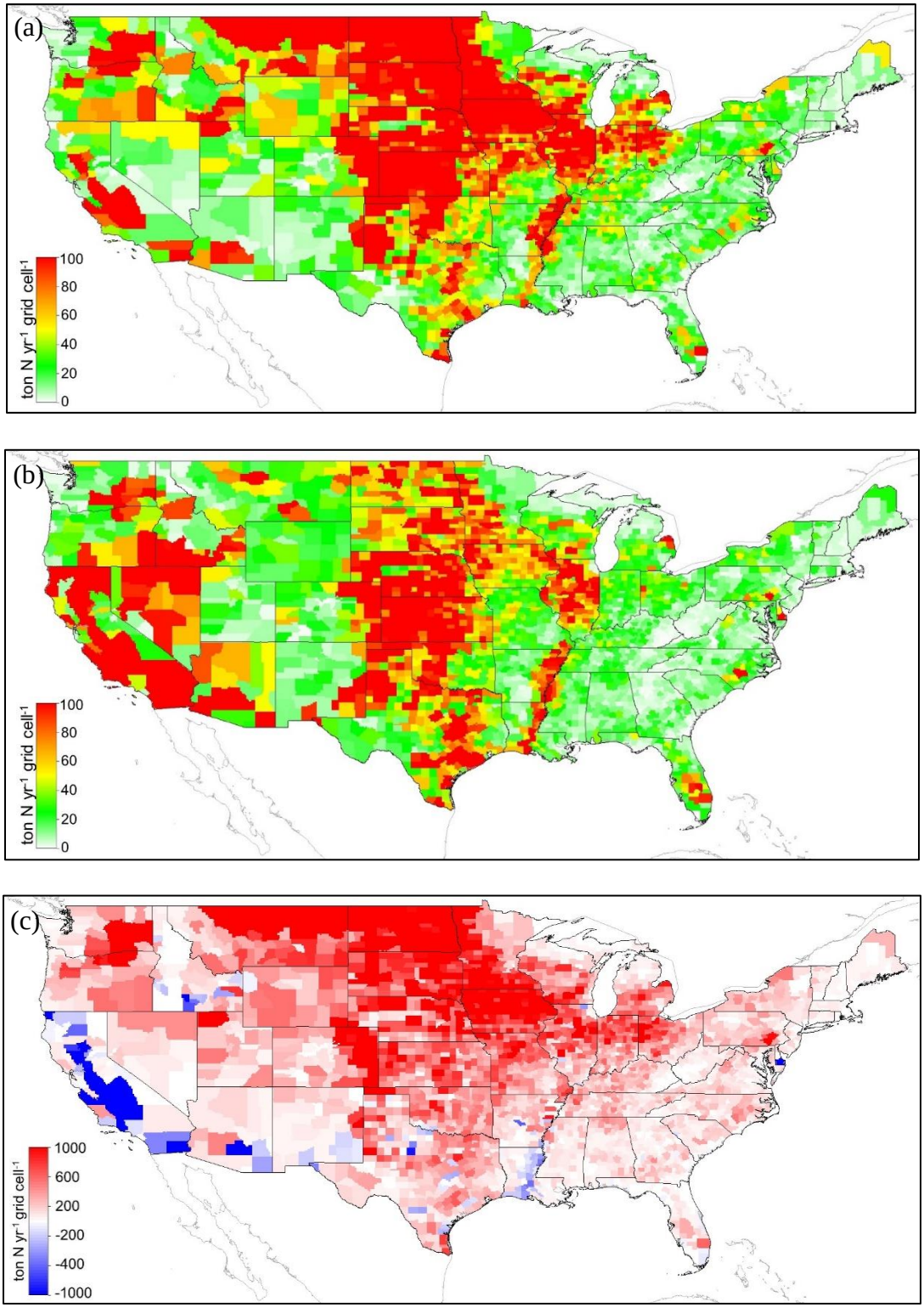

Figure 4.2. Comparison between the results from (a) NH3_STAT, (b) EDGAR, and (c) absolute difference between the two models (in ton N yr^{1} grid $cell^{-1}$).

4.2.2. Indian and China

We also performed a region-based analysis in India, and China) using the NH3_STAT model. The NH3_STAT model estimate for India shows NH3 emissions between -141% and -77% when compared to other data sets. A lower estimate is also seen for China, where the model estimates NH3 emissions 36-482% lower than other data sets.

4.3. Conclusions

The statistical model captures the spatial distribution of global NH3 emissions by utilizing a more simplified approach than those used by other readily available data sets.

From global scale, the results indicate that, in comparison to other data sets, the model generates a lower global NH3 estimate by 57%, (NH3_STAT: 14.2 Tg N yr-1; EDGAR: 33.0 Tg N yr-1).

The region-based analysis (U.S., India, and China) using the NH3_STAT model indicates that there are variations in developed country and developing country. For the U.S., our model produces an estimate that is 143% higher in comparison to EPA. Meanwhile, the NH3_STAT model estimate for India shows NH3 emissions between -141% and -77% when compared to other data sets. A lower estimate is also seen for China, where the model estimates NH3 emissions 36-482% lower than other data sets. The difference in the global estimates is attributed to the lower estimates in major agricultural countries like China and India.

The underestimation of NH3 emissions in comparison to EDGAR can be attributed to other sources that are excluded in our model. This is understandable that EDGAR has additional sources in their estimate, e.g. enteric fermentation, manure management, rice cultivation, whereas NH3_STAT is exclusive to emissions from fertilizer and manure applied as fertilizer.

Supplementary Information

Table 2S. List of publications from where NH3 emissions and physicochemical parameters were obtained.

Location	soil_ph	soil_moisture	soil_temperature	total_N	fertilizer_type	Nh3 emission	Reference
CA	5.3	17.0	19.4	872.1	1	268.9	ALFAM2
CA	5.3	17.0	19.4	872.1	1	264.2	ALFAM2
CA	5.3	17.0	19.4	703.8	1	237.5	ALFAM2
CA	5.3	17.0	19.4	872.1	1	224.7	ALFAM2
CA	5.3	17.0	19.4	703.8	1	194.6	ALFAM2
CA	5.3	17.0	19.4	703.8	1	193.1	ALFAM2
DK	6.3	57.0	15.1	139.7	1	166.2	ALFAM2
DK	5.5	47.0	6.7	181.9	1	151.3	ALFAM2
DK	5.5	47.0	6.7	167.8	1	149.0	ALFAM2
DK	6.3	57.0	15.2	139.7	1	135.3	ALFAM2
DK	5.5	47.0	6.7	181.9	1	112.6	ALFAM2
DK	5.5	47.0	6.7	181.9	1	104.1	ALFAM2
UK	7.9	32.4	15.4	190.4	1	103.3	ALFAM2
UK	7.9	32.4	15.4	190.4	1	74.5	ALFAM2
UK	7.9	32.4	15.4	190.4	1	63.4	ALFAM2
UK	7.9	32.4	15.4	190.4	1	58.7	ALFAM2
DK	5.6	41.0	14.0	160.4	1	54.6	ALFAM2
UK	8.0	25.9	5.8	221.4	1	54.1	ALFAM2
UK	7.1	24.7	14.4	150.0	1	54.0	Webb et al.,2004
UK	7.1	24.7	14.4	180.0	1	51.0	Webb et al.,2004
CA	6.6	30.0	13.4	159.1	1	50.3	ALFAM2
CA	6.4	24.0	9.7	150.2	1	47.5	ALFAM2
CA	6.6	30.0	13.4	101.2	1	46.2	ALFAM2
CA	6.4	24.0	9.7	139.7	1	43.4	ALFAM2
CA	6.4	24.0	9.7	139.7	1	42.3	ALFAM2
CA	6.4	24.0	9.7	150.2	1	41.6	ALFAM2
CA	6.6	30.0	13.4	159.1	1	41.3	ALFAM2
CN	8.3	40.0	18.6	119.3	0	40.6	Yang et al., 2013
CA	6.4	24.0	9.7	111.7	1	39.5	ALFAM2
CA	6.4	24.0	9.7	139.7	1	39.3	ALFAM2
UK	7.9	32.4	15.4	190.4	1	39.1	ALFAM2
CA	6.6	30.0	13.4	159.1	1	38.1	ALFAM2
CA	6.6	30.0	13.4	115.5	1	37.2	ALFAM2
CN	8.3	22.0	23.2	174.0	0	36.8	Yang et al., 2017
CA	6.6	30.0	13.4	115.5	1	36.7	ALFAM2
CA	6.4	24.0	9.7	150.2	1	36.1	ALFAM2

Table 2S. (continued).

IT	8.2	18.9	10.2	107.0	1	35.9	ALFAM2
UK	7.9	32.4	15.4	190.4	1	35.4	ALFAM2
DK	5.6	41.0	14.0	160.4	1	34.8	ALFAM2
DK	5.6	41.0	14.0	160.4	1	34.5	ALFAM2
FR	7.6	17.9	24.5	76.0	1	34.5	ALFAM2
CA	6.4	24.0	9.7	111.7	1	33.9	ALFAM2
DE	6.5	31.0	5.8	120.9	1	33.7	ALFAM2
CA	6.6	30.0	13.4	95.7	1	33.7	ALFAM2
CA	6.6	30.0	13.4	85.9	1	33.1	ALFAM2
CA	6.6	30.0	13.4	101.2	1	33.1	ALFAM2
UK	8.0	25.1	17.4	164.8	1	33.1	ALFAM2
CN	8.3	22.0	23.2	174.0	0	32.8	Yang et al., 2017
CA	6.4	24.0	9.7	142.8	1	32.7	ALFAM2
CA	6.4	24.0	9.7	139.4	1	32.3	ALFAM2
DK	5.5	47.0	6.7	167.8	1	31.7	ALFAM2
CA	6.5	24.0	16.4	152.4	1	31.7	ALFAM2
IT	6.4	14.5	12.6	223.4	1	31.6	ALFAM2
DK	5.5	47.0	6.7	204.8	1	31.4	ALFAM2
IT	6.4	14.5	12.9	223.4	1	31.2	ALFAM2
IT	6.4	21.0	18.5	109.0	1	31.1	Carozzi et al., 2013
UK	8.0	25.1	17.4	164.8	1	31.1	ALFAM2
CA	6.6	30.0	13.4	95.7	1	30.9	ALFAM2
DK	8.1	100.0	13.4	131.4	1	30.4	ALFAM2
IT	8.2	22.0	12.3	68.0	1	30.2	Carozzi et al., 2013
DE	6.5	31.0	5.8	120.9	1	30.0	ALFAM2
UK	7.1	24.7	8.4	110.0	1	30.0	Webb et al.,2004
CA	6.6	30.0	13.4	85.9	1	29.6	ALFAM2
CA	6.6	30.0	13.4	101.2	1	29.1	ALFAM2
DE	6.5	19.0	18.6	170.0	1	28.7	ALFAM2
DE	5.8	31.0	15.2	208.1	1	28.4	ALFAM2
CA	6.4	24.0	9.7	111.7	1	28.2	ALFAM2
DK	7.9	100.0	18.0	125.1	1	28.1	ALFAM2
CA	6.5	24.0	16.4	142.6	1	27.9	ALFAM2
CA	6.5	24.0	16.4	142.6	1	27.6	ALFAM2
CA	6.6	30.0	13.4	95.7	1	27.6	ALFAM2
CA	6.4	24.0	9.7	142.8	1	27.3	ALFAM2
CL	5.4	56.2	17.3	100.0	0	26.7	Salazar et al., 2012
CA	6.5	24.0	16.4	142.6	1	26.7	ALFAM2
CA	6.5	24.0	16.4	151.7	1	26.6	ALFAM2

Table 2S. (continued).

IT	7.1	13.8	12.8	188.3	1	26.4	ALFAM2
DE	6.5	19.0	18.6	170.0	1	26.4	ALFAM2
IT	7.1	17.0	11.2	95.0	1	26.4	Carozzi et al., 2013
DE	5.8	31.0	16.2	199.9	1	26.2	ALFAM2
CA	6.6	30.0	13.4	85.9	1	26.1	ALFAM2
CA	6.5	24.0	16.4	145.2	1	26.1	ALFAM2
UK	7.1	24.7	11.3	145.0	1	26.0	Webb et al.,2004
UK	7.1	24.7	11.3	105.0	1	26.0	Webb et al.,2004
DE	6.5	31.0	5.9	120.9	1	25.8	ALFAM2
CA	6.6	30.0	13.4	115.5	1	25.8	ALFAM2
DE	5.8	31.0	16.2	153.2	1	25.8	ALFAM2
CL	6.1	50.0	10.6	100.0	0	25.7	Salazar et al., 2014
CA	6.4	24.0	9.7	139.4	1	25.7	ALFAM2
CL	6.1	50.0	14.3	100.0	0	25.3	Salazar et al., 2014
CN	8.3	22.0	23.2	174.0	0	24.3	Yang et al., 2017
UK	8.0	25.9	5.8	221.4	1	24.2	ALFAM2
CA	6.1	20.0	23.3	150.0	1	24.2	ALFAM2
CA	6.4	24.0	9.7	139.4	1	24.0	ALFAM2
DE	6.6	45.0	9.1	180.0	1	24.0	Wolf et al., 2014
CA	6.5	24.0	16.4	152.4	1	24.0	ALFAM2
UK	8.0	25.1	17.4	164.8	1	23.7	ALFAM2
CN	8.3	40.0	18.6	90.5	0	23.5	Yang et al., 2013
UK	7.9	32.4	15.4	190.4	1	23.4	ALFAM2
CA	6.5	24.0	16.4	152.4	1	23.2	ALFAM2
DE	5.8	31.0	15.8	167.5	1	22.9	ALFAM2
SW	7.1	30.0	25.0	88.4	1	22.7	Hani et al., 2016
DK	7.0	100.0	12.9	108.4	1	22.6	ALFAM2
CA	6.4	24.0	9.7	142.8	1	22.5	ALFAM2
IT	7.8	10.6	25.8	240.0	0	22.4	Ferrara et al., 2014
UK	7.1	32.5	7.7	115.6	1	22.0	ALFAM2
DE	6.5	19.0	18.6	170.0	1	21.3	ALFAM2
DE	6.5	19.0	18.4	218.9	1	21.2	ALFAM2
DE	6.5	19.0	18.5	218.9	1	21.2	ALFAM2
DK	6.8	14.5	11.9	129.0	1	20.9	ALFAM2
UK	8.0	25.1	17.3	92.3	1	20.9	ALFAM2
CL	6.1	50.0	16.3	100.0	1	20.5	Salazar et al., 2014
DK	5.6	41.0	14.0	160.4	1	20.5	ALFAM2

Table 2S. (continued).

SW	7.1	30.0	11.0	90.0	1	20.4	Hani et al., 2016
CL	5.6	50.0	13.9	100.0	1	20.1	Martinez-Lagos et al., 2013
UK	7.9	32.4	15.4	190.4	1	19.9	ALFAM2
CA	6.5	23.0	12.5	144.7	1	19.9	ALFAM2
CA	6.5	24.0	16.4	151.7	1	19.3	ALFAM2
DK	5.5	47.0	6.7	167.8	1	19.2	ALFAM2
DE	5.8	31.0	16.1	134.0	1	19.2	ALFAM2
DE	6.5	31.0	6.2	143.8	1	19.1	ALFAM2
DE	6.5	19.0	18.2	182.8	1	19.0	ALFAM2
DK	6.4	16.2	11.9	110.9	1	18.9	ALFAM2
SW	7.1	30.0	10.0	93.0	1	18.9	Hani et al., 2016
CA	6.5	24.0	16.4	145.2	1	18.6	ALFAM2
UK	7.9	32.4	15.4	190.4	1	18.6	ALFAM2
SW	6.9	30.0	20.0	78.0	1	18.6	Hani et al., 2016
IT	7.1	13.8	12.8	188.3	1	18.4	ALFAM2
DE	6.5	19.0	18.2	182.8	1	18.3	ALFAM2
DK	6.6	17.0	11.9	105.3	1	18.0	ALFAM2
UK	8.0	25.9	5.8	221.4	1	17.8	ALFAM2
DE	5.8	10.0	17.5	193.0	1	17.7	ALFAM2
DE	6.5	31.0	6.2	143.8	1	17.4	ALFAM2
UK	8.0	29.6	6.2	160.2	1	17.3	ALFAM2
DE	5.8	31.0	16.1	134.0	1	17.0	ALFAM2
DE	6.5	19.0	18.2	182.8	1	16.8	ALFAM2
DE	5.8	10.0	17.1	105.9	1	16.6	ALFAM2
UK	7.9	32.4	15.4	190.4	1	16.6	ALFAM2
UK	7.2	32.5	7.8	115.6	1	16.5	ALFAM2
DE	6.5	19.0	18.5	218.9	1	16.4	ALFAM2
DK	5.5	47.0	6.7	204.8	1	16.3	ALFAM2
UK	7.2	32.5	7.9	115.6	1	16.1	ALFAM2
DE	5.8	31.0	16.1	152.8	1	16.0	ALFAM2
IT	8.1	60.0	15.5	120.0	0	16.0	Badagliacca et al., 2018
CL	6.1	50.0	16.3	100.0	0	15.9	Salazar et al., 2014
CN	8.2	23.0	13.0	140.0	0	15.8	Huo et al., 2015
CL	5.7	50.0	9.7	100.0	1	15.8	Martinez-Lagos et al., 2013
DE	5.8	31.0	15.8	192.0	1	15.8	ALFAM2
UK	8.0	28.5	9.8	126.1	1	15.7	ALFAM2
UK	8.0	29.6	6.2	160.2	1	15.7	ALFAM2

Table 2S. (continued).

DE	5.8	10.0	17.4	172.1	1	15.5	ALFAM2
CA	6.5	23.0	12.5	143.6	1	15.5	ALFAM2
DE	5.8	31.0	16.1	142.8	1	15.4	ALFAM2
UK	8.0	29.6	6.2	160.2	1	15.3	ALFAM2
DE	5.8	10.0	17.5	114.7	1	15.3	ALFAM2
DE	5.8	31.0	16.1	197.5	1	15.2	ALFAM2
SW	6.9	30.0	20.0	58.9	1	15.2	Hani et al., 2016
UK	8.0	25.1	17.3	92.3	1	15.2	ALFAM2
DE	5.8	23.0	9.7	161.4	1	15.1	ALFAM2
DE	5.8	10.0	17.5	114.7	1	15.1	ALFAM2
DE	5.8	31.0	16.2	134.0	1	15.0	ALFAM2
UK	8.0	25.9	5.8	94.7	1	14.9	ALFAM2
CA	6.1	20.0	23.3	144.7	1	14.8	ALFAM2
SW	7.2	30.0	10.0	93.0	1	14.8	Hani et al., 2016
DE	6.5	31.0	6.2	132.0	1	14.7	ALFAM2
DE	5.8	10.0	12.1	124.6	1	14.6	ALFAM2
DE	5.8	31.0	16.2	184.6	1	14.4	ALFAM2
UK	8.0	25.9	5.8	94.7	1	14.4	ALFAM2
CA	6.1	20.0	23.3	143.6	1	14.2	ALFAM2
DE	5.8	10.0	17.4	172.1	1	14.2	ALFAM2
DE	5.8	10.0	17.5	114.7	1	14.1	ALFAM2
DK	8.0	100.0	17.9	126.4	1	14.1	ALFAM2
DE	6.5	31.0	6.1	132.0	1	14.1	ALFAM2
DE	6.5	19.0	18.6	170.0	1	14.1	ALFAM2
DE	6.1	40.0	16.0	140.0	1	14.0	Gericke et al., 2011
UK	8.0	28.5	9.8	126.1	1	13.9	ALFAM2
UK	7.2	20.3	17.5	130.6	1	13.9	ALFAM2
DE	5.8	10.0	17.1	105.9	1	13.8	ALFAM2
CA	6.1	20.0	23.3	140.4	1	13.8	ALFAM2
DE	5.8	31.0	15.2	238.6	1	13.7	ALFAM2
DE	6.5	31.0	6.2	143.8	1	13.3	ALFAM2
DE	6.1	40.0	16.0	140.0	1	13.3	Gericke et al., 2011
UK	8.0	29.6	6.2	66.3	1	13.3	ALFAM2
UK	7.2	32.5	7.8	115.6	1	13.2	ALFAM2
DE	5.8	10.0	17.5	172.1	1	13.2	ALFAM2
CA	6.1	30.0	19.2	142.0	1	13.2	ALFAM2
DK	5.6	41.0	14.0	160.4	1	13.1	ALFAM2
DE	5.8	10.0	17.0	158.9	1	13.1	ALFAM2
DE	5.8	31.0	16.1	174.9	1	13.0	ALFAM2
DE	5.8	10.0	17.2	158.9	1	12.9	ALFAM2

Table 2S. (continued).

SE	7.1	24.7	15.3	67.5	1	12.9	Rodhe et al., 2006
SW	6.6	30.0	19.0	65.0	1	12.9	Hani et al., 2016
DE	5.8	31.0	16.1	197.5	1	12.9	ALFAM2
DE	5.8	31.0	15.8	167.5	1	12.7	ALFAM2
DE	6.5	40.0	15.0	140.0	1	12.6	Ni et al., 2015
DE	5.8	10.0	17.0	52.9	1	12.6	ALFAM2
DE	6.5	17.0	21.0	191.8	1	12.6	ALFAM2
CA	6.1	20.0	23.3	143.4	1	12.6	ALFAM2
DE	5.8	10.0	17.5	114.7	1	12.6	ALFAM2
DE	6.5	31.0	6.1	132.0	1	12.5	ALFAM2
DE	5.8	31.0	16.2	142.8	1	12.4	ALFAM2
DE	6.5	17.0	21.0	191.8	1	12.4	ALFAM2
DE	5.8	31.0	16.2	142.8	1	12.4	ALFAM2
DE	5.8	23.0	9.7	184.5	1	12.3	ALFAM2
CL	5.6	63.3	16.8	100.0	0	12.2	Salazar et al., 2012
DE	5.8	10.0	16.9	105.9	1	12.1	ALFAM2
UK	7.1	24.7	8.4	115.0	1	12.0	Webb et al.,2004
DE	5.8	10.0	17.5	172.1	1	11.9	ALFAM2
DK	6.1	24.9	10.7	116.0	1	11.8	ALFAM2
DE	6.5	17.0	20.7	149.5	1	11.8	ALFAM2
UK	8.0	25.2	12.0	136.6	1	11.7	ALFAM2
DK	5.6	41.0	14.0	160.4	1	11.6	ALFAM2
DE	5.8	23.0	19.8	109.6	1	11.6	ALFAM2
DE	5.8	10.0	12.1	127.8	1	11.6	ALFAM2
CA	6.5	21.0	23.0	150.0	1	11.5	ALFAM2
DE	5.8	10.0	12.1	73.0	1	11.5	ALFAM2
CA	6.5	23.0	12.5	143.4	1	11.4	ALFAM2
UK	8.0	20.2	16.9	164.0	1	11.4	ALFAM2
CA	6.5	24.0	16.4	97.4	1	11.4	ALFAM2
DE	5.8	31.0	15.3	143.3	1	11.4	ALFAM2
UK	7.1	28.3	16.1	102.0	1	11.3	ALFAM2
UK	7.2	30.6	10.5	110.1	1	11.3	ALFAM2
CA	6.5	21.0	23.0	144.7	1	11.3	ALFAM2
DE	6.5	17.0	20.8	149.5	1	11.3	ALFAM2
DE	5.8	10.0	17.4	193.0	1	11.2	ALFAM2
FR	7.6	30.7	9.2	141.4	1	11.2	ALFAM2
CN	8.2	23.0	13.0	117.0	0	11.1	Huo et al., 2015
DE	5.8	10.0	12.2	157.4	1	11.1	ALFAM2
DE	5.8	23.0	9.7	184.5	1	11.0	ALFAM2

Table 2S. (continued).

DE	5.8	10.0	12.1	127.8	1	11.0	ALFAM2
DE	6.5	17.0	20.9	191.8	1	10.9	ALFAM2
UK	7.1	32.5	7.7	115.6	1	10.9	ALFAM2
DE	5.8	31.0	16.1	184.6	1	10.9	ALFAM2
DE	5.8	10.0	12.2	157.4	1	10.8	ALFAM2
DE	6.5	17.0	20.9	149.5	1	10.7	ALFAM2
UK	8.0	25.9	5.8	94.7	1	10.7	ALFAM2
DK	6.1	22.8	10.7	109.2	1	10.7	ALFAM2
CA	6.5	21.0	23.0	143.6	1	10.7	ALFAM2
UK	7.2	20.3	17.5	130.6	1	10.7	ALFAM2
UK	8.0	20.2	16.8	89.4	1	10.6	ALFAM2
DE	5.8	31.0	15.9	192.0	1	10.6	ALFAM2
DE	5.8	23.0	9.7	161.4	1	10.6	ALFAM2
DE	5.8	23.0	9.7	184.5	1	10.5	ALFAM2
CA	6.5	23.0	12.5	150.0	1	10.5	ALFAM2
UK	7.1	32.5	7.7	115.6	1	10.4	ALFAM2
DE	5.8	23.0	9.7	184.5	1	10.4	ALFAM2
DE	5.8	10.0	12.1	124.6	1	10.3	ALFAM2
DE	5.8	23.0	19.5	174.3	1	10.3	ALFAM2
UK	7.2	26.5	16.1	102.0	1	10.3	ALFAM2
UK	7.2	20.4	21.6	130.3	1	10.3	ALFAM2
DE	6.5	17.0	20.8	149.5	1	10.2	ALFAM2
DE	5.8	23.0	19.9	109.6	1	10.2	ALFAM2
DK	6.1	24.9	10.7	116.0	1	10.2	ALFAM2
CA	6.5	21.0	23.0	140.4	1	10.2	ALFAM2
DE	5.8	10.0	17.5	193.0	1	10.1	ALFAM2
DK	7.9	100.0	10.5	135.9	1	10.1	ALFAM2
UK	7.1	26.6	12.5	130.7	1	10.1	ALFAM2
GE	6.5	40.0	15.0	140.0	1	10.1	Ni et al., 2015
GE	6.5	40.0	8.0	140.0	1	10.1	Ni et al., 2015
IT	8.1	60.0	15.5	80.0	0	10.0	Badagliacca et al., 2018
GE	6.5	40.0	15.0	140.0	1	10.0	Ni et al., 2015
DK	6.1	26.9	9.9	110.9	1	9.9	ALFAM2
DE	5.8	10.0	12.1	54.8	1	9.9	ALFAM2
DE	5.8	10.0	12.1	124.6	1	9.9	ALFAM2
UK	7.2	20.3	17.5	130.6	1	9.8	ALFAM2
CA	6.5	24.0	16.4	151.7	1	9.8	ALFAM2
UK	8.0	28.5	10.0	149.1	1	9.7	ALFAM2
DE	5.8	23.0	9.7	161.4	1	9.6	ALFAM2
UK	8.0	20.2	16.9	164.0	1	9.5	ALFAM2

Table 2S. (continued).

DE	5.8	10.0	12.1	54.8	1	9.5	ALFAM2
UK	8.0	25.1	17.4	92.3	1	9.5	ALFAM2
UK	7.1	26.6	12.5	130.7	1	9.5	ALFAM2
DE	5.8	10.0	12.1	127.8	1	9.4	ALFAM2
DE	5.8	23.0	19.9	109.6	1	9.4	ALFAM2
DE	6.5	28.0	2.2	124.2	1	9.4	ALFAM2
UK	7.2	20.4	21.6	130.3	1	9.4	ALFAM2
UK	8.0	25.2	12.0	136.6	1	9.4	ALFAM2
DE	5.8	23.0	19.5	174.3	1	9.4	ALFAM2
UK	8.0	20.2	16.8	89.4	1	9.3	ALFAM2
DE	5.8	23.0	19.5	174.3	1	9.3	ALFAM2
DE	5.8	10.0	12.3	157.4	1	9.3	ALFAM2
DK	8.2	100.0	16.7	108.9	1	9.3	ALFAM2
DK	6.1	23.0	10.7	157.6	1	9.3	ALFAM2
DE	6.5	28.0	2.2	124.2	1	9.2	ALFAM2
CA	6.5	23.0	12.5	140.4	1	9.2	ALFAM2
SW	7.0	30.0	20.0	70.2	1	9.2	Hani et al., 2016
DE	6.5	28.0	2.2	124.3	1	9.1	ALFAM2
UK	7.1	28.8	12.1	110.1	1	9.1	ALFAM2
DE	5.8	23.0	9.8	161.4	1	9.1	ALFAM2
SW	6.8	30.0	20.0	69.6	1	9.0	Hani et al., 2016
DK	8.2	56.6	5.1	115.1	1	9.0	ALFAM2
DK	5.6	41.0	14.0	160.4	1	9.0	ALFAM2
DE	6.5	30.0	5.1	111.2	1	9.0	ALFAM2
UK	8.0	20.2	16.9	164.0	1	8.9	ALFAM2
DE	5.8	23.0	19.8	109.6	1	8.9	ALFAM2
DE	6.1	40.0	2.7	120.0	1	8.9	Gericke et al., 2011
DK	6.1	26.9	9.9	110.9	1	8.9	ALFAM2
DE	6.5	17.0	20.8	115.3	1	8.8	ALFAM2
UK	8.0	29.6	6.2	66.3	1	8.8	ALFAM2
DE	6.1	40.0	2.7	120.0	1	8.8	Gericke et al., 2011
UK	8.0	20.2	16.8	89.4	1	8.7	ALFAM2
DE	5.8	10.0	12.1	127.8	1	8.7	ALFAM2
CL	6.1	50.0	14.3	100.0	1	8.7	Salazar et al., 2014
DE	5.8	31.0	5.8	167.4	1	8.6	ALFAM2
UK	7.2	20.4	21.6	130.3	1	8.6	ALFAM2
DE	5.8	23.0	8.8	188.4	1	8.6	ALFAM2
DE	6.5	30.0	5.1	111.2	1	8.6	ALFAM2
UK	7.1	28.3	16.1	102.0	1	8.6	ALFAM2
GE	6.5	40.0	8.0	140.0	1	8.5	Ni et al., 2015

Table 2S. (continued).

UK	8.0	28.5	9.8	126.1	1	8.5	ALFAM2
SW	7.1	30.0	15.0	44.0	1	8.5	Hani et al., 2016
DE	5.8	10.0	12.2	157.4	1	8.4	ALFAM2
CA	6.5	21.0	23.0	143.4	1	8.4	ALFAM2
DK	6.1	23.0	10.7	157.6	1	8.4	ALFAM2
DE	6.5	17.0	20.4	95.5	1	8.4	ALFAM2
DE	5.8	23.0	19.9	73.0	1	8.4	ALFAM2
DK	6.1	28.3	9.9	121.8	1	8.4	ALFAM2
SW	7.1	30.0	16.0	54.0	1	8.3	Hani et al., 2016
UK	7.1	26.6	12.5	130.7	1	8.3	ALFAM2
DE	5.8	10.0	12.1	54.8	1	8.2	ALFAM2
DK	6.1	24.2	10.7	116.3	1	8.2	ALFAM2
DE	5.8	28.0	14.6	177.5	1	8.1	ALFAM2
CA	6.1	30.0	19.2	140.1	1	8.1	ALFAM2
SW	6.9	30.0	19.0	59.8	1	8.1	Hani et al., 2016
DE	6.5	17.0	20.8	115.3	1	8.1	ALFAM2
UK	8.0	25.2	12.0	87.3	1	8.0	ALFAM2
DK	7.0	10.8	12.8	76.8	1	8.0	ALFAM2
DK	6.1	24.2	10.7	116.3	1	8.0	ALFAM2
UK	7.2	30.6	10.5	110.1	1	7.9	ALFAM2
GE	6.5	40.0	17.0	110.0	0	7.8	Ni et al., 2015
DE	6.5	17.0	20.8	191.8	1	7.8	ALFAM2
DE	5.8	31.0	15.3	178.3	1	7.8	ALFAM2
CA	6.1	30.0	19.2	140.5	1	7.8	ALFAM2
SW	7.3	30.0	14.0	64.0	1	7.7	Hani et al., 2016
CL	6.4	42.6	10.0	100.0	0	7.7	Salazar et al., 2012
DE	5.8	10.0	12.1	73.0	1	7.7	ALFAM2
DE	6.5	30.0	5.1	111.2	1	7.7	ALFAM2
DE	5.8	23.0	19.9	91.3	1	7.7	ALFAM2
DE	5.8	23.0	19.9	91.3	1	7.7	ALFAM2
GE	6.5	40.0	8.0	140.0	1	7.7	Ni et al., 2015
DE	5.8	10.0	12.1	124.6	1	7.6	ALFAM2
DE	5.8	10.0	16.9	52.9	1	7.6	ALFAM2
DE	5.8	31.0	5.3	170.7	1	7.6	ALFAM2
DE	6.1	40.0	5.3	120.0	1	7.6	Gericke et al., 2011
CL	5.3	50.0	9.4	100.0	1	7.6	Martinez-Lagos et al., 2013
DE	6.5	28.0	2.2	124.2	1	7.6	ALFAM2
DE	5.8	10.0	12.3	74.7	1	7.6	ALFAM2
DE	6.5	30.0	4.4	130.0	1	7.6	ALFAM2

Table 2S. (continued).

DE	5.8	23.0	8.9	188.4	1	7.6	ALFAM2
UK	7.1	28.8	12.1	110.1	1	7.5	ALFAM2
DE	6.1	40.0	18.5	80.0	1	7.5	Gericke et al., 2011
DK	7.0	12.0	12.8	83.6	1	7.5	ALFAM2
DK	7.8	100.0	0.3	109.1	1	7.5	ALFAM2
DE	6.5	30.0	4.6	89.0	1	7.4	ALFAM2
DE	5.8	10.0	12.3	74.7	1	7.4	ALFAM2
SW	7.3	30.0	2.0	67.5	1	7.3	Hani et al., 2016
DE	6.5	30.0	5.0	111.2	1	7.3	ALFAM2
GE	6.1	40.0	5.3	120.0	1	7.3	Gericke et al., 2011
CL	6.1	50.0	10.6	100.0	1	7.3	Salazar et al., 2014
DE	6.5	30.0	4.7	89.0	1	7.3	ALFAM2
DE	5.8	23.0	19.8	73.0	1	7.3	ALFAM2
GE	6.5	40.0	17.0	110.0	0	7.3	Ni et al., 2015
DE	6.5	30.0	4.5	130.0	1	7.2	ALFAM2
DE	5.8	10.0	12.1	73.0	1	7.2	ALFAM2
DK	5.6	41.0	14.0	160.4	1	7.2	ALFAM2
DE	6.1	40.0	18.5	80.0	1	7.1	Gericke et al., 2011
CA	6.5	24.0	16.4	145.2	1	7.0	ALFAM2
DE	5.8	31.0	5.8	167.4	1	7.0	ALFAM2
UK	8.0	28.5	9.8	149.1	1	7.0	ALFAM2
SW	7.3	30.0	21.0	55.1	1	7.0	Hani et al., 2016
DK	8.0	100.0	17.3	112.7	1	6.9	ALFAM2
UK	8.0	28.5	9.8	149.1	1	6.9	ALFAM2
IE	7.0	15.0	13.1	119.6	1	6.9	Meade et al., 2011
UK	8.0	29.6	6.2	66.3	1	6.9	ALFAM2
DE	6.5	17.0	20.6	95.5	1	6.8	ALFAM2
DK	7.4	100.0	10.2	106.5	1	6.8	ALFAM2
DE	5.8	31.0	5.3	170.7	1	6.8	ALFAM2
DK	6.1	27.4	9.9	118.4	1	6.8	ALFAM2
DE	5.8	23.0	19.9	36.5	1	6.8	ALFAM2
CA	6.1	30.0	19.2	139.2	1	6.8	ALFAM2
DK	5.5	47.0	6.7	204.8	1	6.8	ALFAM2
GE	6.5	40.0	17.0	110.0	0	6.7	Ni et al., 2015
DK	6.1	27.8	9.9	111.4	1	6.7	ALFAM2
DE	5.8	28.0	14.6	165.4	1	6.5	ALFAM2
DK	6.1	22.8	10.7	109.2	1	6.5	ALFAM2

Table 2S. (continued).

DE	6.5	28.0	1.6	116.7	1	6.5	ALFAM2
DK	6.1	29.5	9.9	115.9	1	6.5	ALFAM2
CA	6.5	24.0	16.4	97.4	1	6.3	ALFAM2
DK	5.6	41.0	14.0	160.4	1	6.3	ALFAM2
DE	5.8	23.0	19.8	73.0	1	6.3	ALFAM2
UK	7.2	30.6	10.5	110.1	1	6.2	ALFAM2
SW	7.7	30.0	2.0	49.4	1	6.2	Hani et al., 2016
DE	6.5	17.0	20.5	95.5	1	6.2	ALFAM2
CA	6.1	30.0	19.2	139.9	1	6.2	ALFAM2
DE	6.5	30.0	4.5	130.0	1	6.2	ALFAM2
DK	6.1	27.4	9.9	118.4	1	6.2	ALFAM2
DK	6.1	24.5	10.7	118.3	1	6.1	ALFAM2
DE	5.8	28.0	14.6	165.4	1	6.1	ALFAM2
DE	5.8	31.0	15.3	178.3	1	6.1	ALFAM2
DE	6.5	30.0	4.5	130.0	1	6.1	ALFAM2
IE	7.0	15.0	11.7	103.2	1	6.0	Meade et al., 2011
CL	5.5	50.0	8.7	100.0	1	6.0	Martinez-Lagos et al., 2013
DE	6.5	30.0	4.8	89.0	1	6.0	ALFAM2
UK	7.2	26.5	16.1	102.0	1	5.9	ALFAM2
IT	5.5	25.0	12.2	92.0	1	5.9	Carozzi et al., 2013
SW	7.7	30.0	21.0	35.1	1	5.9	Hani et al., 2016
GE	6.5	40.0	17.0	110.0	0	5.8	Ni et al., 2015
CN	8.2	23.0	13.0	122.0	0	5.8	Huo et al., 2015
SW	7.3	30.0	23.0	46.4	1	5.8	Hani et al., 2016
DE	5.8	23.0	19.7	99.0	1	5.7	ALFAM2
DK	6.1	27.8	9.9	111.4	1	5.7	ALFAM2
SW	6.8	30.0	16.0	69.6	1	5.7	Hani et al., 2016
DE	5.8	10.0	12.2	74.7	1	5.6	ALFAM2
DE	6.5	40.0	15.0	140.0	1	5.6	Ni et al., 2015
DE	5.8	23.0	8.9	188.4	1	5.5	ALFAM2
IT	5.9	15.8	7.8	204.1	1	5.5	ALFAM2
DE	5.8	23.0	19.9	73.0	1	5.4	ALFAM2
DE	5.8	23.0	19.8	36.5	1	5.4	ALFAM2
DE	6.5	17.0	20.8	115.3	1	5.3	ALFAM2
SW	7.2	30.0	20.0	70.2	1	5.3	Hani et al., 2016
DE	5.8	23.0	19.7	99.0	1	5.3	ALFAM2
SW	7.3	30.0	13.0	56.0	1	5.2	Hani et al., 2016
SW	7.2	30.0	9.0	55.1	1	5.2	Hani et al., 2016
DK	6.1	24.5	10.7	118.3	1	5.2	ALFAM2

Table 2S. (continued).

DE	5.8	28.0	14.6	177.5	1	5.1	ALFAM2
DE	5.8	10.0	12.1	73.0	1	5.0	ALFAM2
DE	5.8	31.0	5.8	167.4	1	5.0	ALFAM2
DE	6.5	21.0	13.3	121.3	1	4.9	ALFAM2
DE	5.8	28.0	14.6	177.5	1	4.9	ALFAM2
DK	6.1	29.5	9.9	115.9	1	4.9	ALFAM2
DK	7.0	12.9	12.8	82.9	1	4.9	ALFAM2
SW	7.1	30.0	16.0	64.0	1	4.8	Hani et al., 2016
DE	6.1	40.0	16.7	120.0	1	4.8	Gericke et al., 2011
SW	7.3	30.0	17.0	49.6	1	4.7	Hani et al., 2016
CA	6.5	24.0	16.4	97.4	1	4.7	ALFAM2
SW	7.3	30.0	13.0	56.0	1	4.6	Hani et al., 2016
DK	6.4	14.7	11.9	94.4	1	4.6	ALFAM2
IT	8.0	14.9	10.2	103.9	1	4.5	ALFAM2
UK	7.2	26.5	16.1	102.0	1	4.5	ALFAM2
DE	5.8	28.0	14.6	75.4	1	4.5	ALFAM2
DE	6.1	40.0	16.7	120.0	1	4.5	Gericke et al., 2011
DE	5.8	28.0	14.6	165.4	1	4.5	ALFAM2
DE	5.8	28.0	14.6	37.7	1	4.5	ALFAM2
UK	8.0	25.2	12.0	87.3	1	4.3	ALFAM2
DE	5.8	23.0	19.9	36.5	1	4.3	ALFAM2
DE	6.5	21.0	13.3	121.3	1	4.3	ALFAM2
DK	6.1	28.3	9.9	121.8	1	4.3	ALFAM2
SW	7.6	30.0	2.0	55.0	1	4.3	Hani et al., 2016
DE	6.5	28.0	1.7	116.7	1	4.2	ALFAM2
SW	7.3	30.0	9.0	48.6	1	4.2	Hani et al., 2016
SW	7.3	30.0	16.0	46.4	1	4.2	Hani et al., 2016
CN	7.7	18.5	10.6	200.0	0	4.1	Roelcke et al., 2002
DE	5.8	23.0	19.9	36.5	1	4.0	ALFAM2
DE	5.8	10.0	12.1	54.8	1	3.9	ALFAM2
DE	6.5	21.0	13.4	121.3	1	3.9	ALFAM2
SW	7.2	30.0	15.0	59.4	1	3.9	Hani et al., 2016
SW	7.7	30.0	23.0	24.7	1	3.9	Hani et al., 2016
DE	6.5	28.0	2.0	94.8	1	3.9	ALFAM2
DE	5.8	23.0	19.8	99.0	1	3.8	ALFAM2
IT	8.0	22.0	12.3	66.0	1	3.7	Carozzi et al., 2013
DE	5.8	28.0	14.6	111.9	1	3.6	ALFAM2
DE	6.5	28.0	2.0	94.7	1	3.6	ALFAM2
IT	7.5	25.5	19.6	191.8	1	3.5	ALFAM2

Table 2S. (continued).

DE	5.8	28.0	14.6	111.9	1	3.5	ALFAM2
CN	7.8	14.7	30.2	200.0	0	3.5	Roelcke et al., 2002
DE	5.8	10.0	16.9	52.9	1	3.4	ALFAM2
DE	6.5	28.0	2.0	94.7	1	3.4	ALFAM2
IT	7.5	36.0	19.2	139.0	1	3.4	Carozzi et al., 2013
SW	7.0	30.0	15.0	44.8	1	3.4	Hani et al., 2016
CN	8.3	20.0	13.4	180.0	0	3.3	Yang et al., 2015
SW	7.1	30.0	15.0	75.4	1	3.3	Hani et al., 2016
DE	5.8	28.0	14.6	177.5	1	3.3	ALFAM2
DE	5.8	28.0	14.6	75.4	1	3.2	ALFAM2
DK	7.0	9.0	12.8	65.5	1	3.2	ALFAM2
UK	7.1	28.8	12.1	110.1	1	3.2	ALFAM2
DE	6.5	30.0	4.7	86.6	1	3.2	ALFAM2
DE	6.5	30.0	4.6	59.3	1	3.1	ALFAM2
CL	6.1	50.0	8.5	100.0	1	3.1	Salazar et al., 2014
DE	5.8	31.0	5.6	170.7	1	3.0	ALFAM2
UK	7.1	28.3	16.1	102.0	1	2.9	ALFAM2
DE	5.8	28.0	14.6	37.7	1	2.8	ALFAM2
DE	5.8	28.0	14.6	81.1	1	2.8	ALFAM2
SW	7.7	30.0	7.0	47.6	1	2.7	Hani et al., 2016
DE	6.5	30.0	4.6	43.3	1	2.7	ALFAM2
DE	6.5	28.0	2.0	94.8	1	2.7	ALFAM2
DE	5.8	28.0	14.6	81.1	1	2.7	ALFAM2
CN	8.3	16.0	22.5	180.0	0	2.7	Yang et al., 2015
DE	5.8	28.0	14.6	37.7	1	2.5	ALFAM2
SW	7.7	30.0	23.0	29.9	1	2.4	Hani et al., 2016
DE	5.8	28.0	14.6	111.9	1	2.4	ALFAM2
SW	7.7	30.0	22.0	36.4	1	2.4	Hani et al., 2016
DE	5.8	28.0	14.6	81.1	1	2.3	ALFAM2
GE	6.5	40.0	8.0	140.0	1	2.3	Ni et al., 2015
DE	6.5	21.0	13.4	119.2	1	2.3	ALFAM2
CN	7.8	14.7	30.2	100.0	0	2.3	Roelcke et al., 2002
SW	6.8	30.0	10.0	33.6	1	2.2	Hani et al., 2016
UK	8.0	25.2	12.0	87.3	1	2.1	ALFAM2
DE	5.8	28.0	14.6	75.4	1	2.1	ALFAM2
DE	5.8	28.0	14.6	37.7	1	2.0	ALFAM2
DE	6.5	30.0	4.7	29.7	1	2.0	ALFAM2
DE	5.8	28.0	14.6	111.9	1	2.0	ALFAM2
DE	6.5	21.0	13.4	119.2	1	1.9	ALFAM2

Table 2S. (continued).

SW	7.8	30.0	7.0	28.6	1	1.8	Hani et al., 2016
CL	6.1	50.0	8.5	100.0	0	1.8	Salazar et al., 2014
DE	6.5	21.0	13.4	119.2	1	1.8	ALFAM2
SW	7.2	30.0	14.0	20.9	1	1.7	Hani et al., 2016
SW	7.7	30.0	20.0	32.2	1	1.6	Hani et al., 2016
SW	7.7	30.0	20.0	24.7	1	1.6	Hani et al., 2016
UK	8.0	25.2	12.0	136.6	1	1.5	ALFAM2
SW	6.9	30.0	10.0	26.6	1	1.5	Hani et al., 2016
SW	7.2	30.0	14.0	27.0	1	1.5	Hani et al., 2016
CL	6.4	42.6	9.2	100.0	0	1.4	Salazar et al., 2012
DE	6.5	21.0	13.4	121.3	1	1.4	ALFAM2
SW	7.7	30.0	7.0	54.4	1	1.3	Hani et al., 2016
SW	7.8	30.0	24.0	31.2	1	1.2	Hani et al., 2016
CN	7.8	18.5	10.6	100.0	0	1.1	Roelcke et al., 2002
SW	6.9	30.0	20.0	33.6	1	1.0	Hani et al., 2016
SW	6.9	30.0	21.0	32.4	1	0.8	Hani et al., 2016
SW	7.3	30.0	14.0	22.0	1	0.7	Hani et al., 2016
SW	6.8	30.0	10.0	32.5	1	0.7	Hani et al., 2016
GE	6.6	40.0	9.1	180.0	0	0.6	Wolf et al., 2014

CHAPTER 5 Characteristic of NOx Emission

5.1. Global

The NO2 emission from agricultural soil is negligible, so this chapter only focus on the emission of NO. Similar to previous procedures, after calculating the coefficients of all variables, we applied the NO_STAT model for each grid cell (5 arc-minute x 5 arc-minute) and generated a global map for NO emissions. Figure 5.1(a) gives the spatial distribution of global NO emission from agricultural soils calculated using NO_STAT in kg N/yr/grid cell. The resolution of this map is 5 arc-minute, which is equivalent to 8464 ha. Figure 5.1(b) is spatial distribution of global NO emission from agricultural soil based on EDGAR. Total annual global NO emissions from NO_STAT and EDGAR are 0.208 Tg N/yr and 1.623 Tg N/yr, respectively. The NO_STAT model gives a lower global NOx estimates by 87%. Similar to NH3, there is a large difference between our estimate and EDGAR. As discussed previously, this difference can be attributed to other sources that are excluded in our model. EDGAR has additional sources in their estimate, e.g. Enteric fermentation, Manure management, Rice cultivation, whereas NO_STAT is exclusive to emissions from fertilizer and manure applied as fertilizer. Nevertheless, it is encouraging to see that, in general, the model captures the global spatial pattern in NO emissions well. The hot spots show most NO emission comes from northern China, northern India, and Middle Eastern U.S.

Our model also underestimated NO emission compare with literature survey. Based on our literature survey, Hudman et al. (2012) used a mechanistic model of global soil NO emissions to estimate that total global NO emission from soil is 10.7 Tg N yr-1 and from

fertilizer N input (1.5% of applied N) is 1.8Tg N yr-1. This might due to that most of the experiment conducted on field are short-term and non-continuous.

Figure 5.1 (c) shows the absolute difference between NO_STAT and EDGAR. In comparison with EDGAR, NO_STAT gives relatively lower NH3 emission values in all the places.

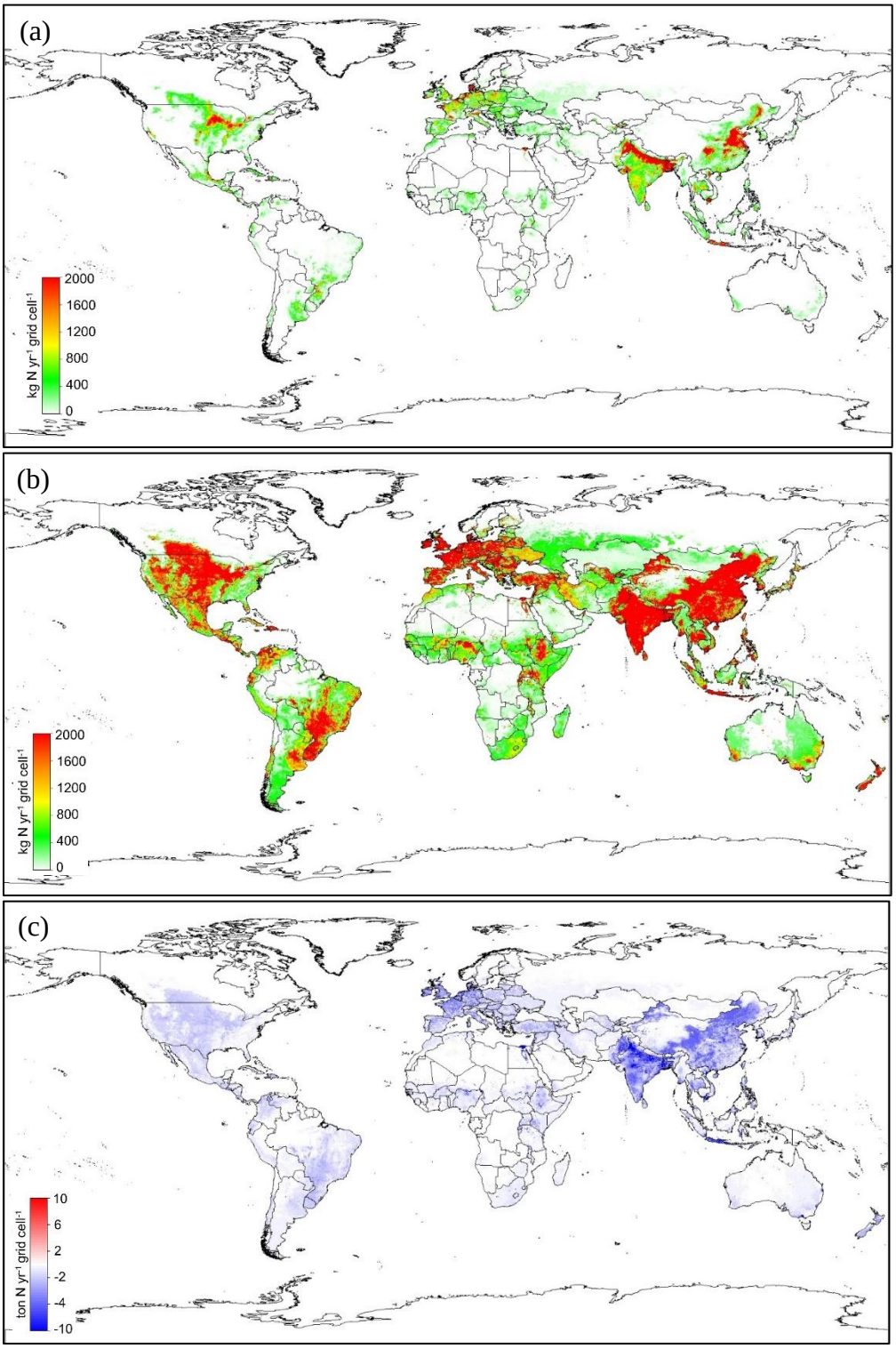

Figure 5.1 Comparison between the results from (a) NO_STAT, (b) EDGAR, and (c) absolute difference between the two models (in ton N yr ^{1} grid cell^{-1}).

5.2. Conclusions

Overall, it is encouraging to see that the statistical model captures the spatial distribution of global NOx emissions but the model estimates is far below other model estimate and literature survey. From global scale, the results indicate that, in comparison to other data sets, the model generates a lower global NO estimate by 87%, (NO_STAT: 0.208 Tg N/yr and EDGAR:1.623 Tg N/yr,). Two reasons can contribute to this underestimation. One reason is that the underestimation of NO emissions in comparison to EDGAR can be attributed to EDGAR has additional sources in their estimate, e.g. Enteric fermentation, Manure management, Rice cultivation, whereas NO_STAT is exclusive to emissions from fertilizer and manure applied as fertilizer. The other reason is that most NO field measurement campaign are short-term and non-continuous.

Supplementary Information

Table 3S. List of publications from where NO emissions and physicochemical parameters were obtained.

Location	pH	Soil moisture (%)	Temperature	N_type (0: fer 1:man)	N_rate (kg N/ha)	NO emission	Length of Experiment	Reference
Japan	5.9	39	23	0	200	1.5166656	131	Akiyama et al (2000)
Japan	5.9	39	23	0	200	2.3089536	131	Akiyama et al (2000)
Japan	5.9	39	23	0	200	2.376864	131	Akiyama et al (2000)
Colorado, USA	7.5	14.3	28	0	63	0.1714176	64	Anderson et al (1987)
Virginia, USA	6	25	15	0	40	1.33056	350	Anderson et al (1987)
Virginia, USA	6	25	15	0	40	1.48176	350	Anderson et al (1987)
Virginia, USA	6	25	15	0	100	1.897344	360	Anderson et al (1987)
Virginia, USA	6	25	15	0	100	4.292352	360	Anderson et al (1987)
NC, USA	6.2	28.33	14.9	0	68	0.536	365	Aneja_et_al._(1996)
NC, USA	6	35.75	13.7	0	168	2.208	365	Aneja_et_al._(1996)
Puerto Rico	4	87	22.6	0	150	0.637632	90	
Australia	8.2	22	20.3	0	80	0.002	8	Galbally_et_al._(1987)
France	5.2	30	12.5	0	280	32	365	Jambert_et_al._(1997)
France	4.8	30	6.5	0	280	0.001	1	Jambert_et_al._(1994)
France	4.5	30	6.5	0	150	0.002	1	Jambert_et_al._(1994)
France	6.6	30	18.5	0	150	0.473	1	Jambert_et_al._(1994)
Sweden	6.3	30	15.4	0	120	0.85	120	
Sweden	6.3	30	13.65	0	200	0.89	120	
Maui	6.8	40.5	20.3	0	34	0.004	8	Matson_et_al._(1996)
Maui	7.6	68.3	17.8	0	45	0.012	8	Matson_et_al._(1996)
Maui	4.7	49.5	19.7	0	84	0.012	8	Matson_et_al._(1996)
Maui	7	50.4	20.3	0	35	0.001	8	Matson_et_al._(1996)
Maui	6	68.3	17.8	0	22	0.008	8	Matson_et_al._(1996)

Table 3S. (continued).

Mauna Loa, Hawaii	5.4	68.8	5.6	0	94	0.71	30	Matson_et_al._(1996)
Maui	7	40.5	20.3	0	39	0.013	8	Matson_et_al._(1996)
Mauna Loa, Hawaii	4.3	57.7	5.8	0	95	1.95	15	Matson_et_al._(1996)
Brazil	5.84	57.75	32.25	0	33	1.58112	183	
Brazil	5.94	39.5	32.75	0	42	2.134512	183	
NC, USA	5.8	22.7	14.3	0	150	0.231	4	Roelle_et_al._(2001)
NC, USA	5.8	22.7	14.3	0	150	0.231	4	Roelle_et_al._(2001)
NC, USA	5.8	6.9	15.4	0	140	0.009	4	Roelle_et_al._(2001)
NC, USA	5.8	9.4	17.1	0	150	0.033	4	Roelle_et_al._(2001)
NC, USA	5.8	11.3	23	0	45	0.029	4	Roelle_et_al._(2001)
NC, USA	5.8	12.4	25.6	0	190	0.033	6	Roelle_et_al._(2001)
NC, USA	5.8	5.6	27.4	0	70	0.025	7	Roelle_et_al._(2001)
NC, USA	5.8	21.7	19.1	0	197	0.339	7	Roelle_et_al._(2001)
NC, USA	5.8	21.1	24.1	0	175	1.055	24	Roelle_et_al._(2001)
Venezuela	5.6	30	31	0	200	0.3	8	Rondon_et_al._(1993)
Venezuela	5.6	30	31	0	200	0.69	8	Rondon_et_al._(1993)
Venezuela	5.6	30	31	0	200	1.69	8	Rondon_et_al._(1993)
Guárice state	3.7	2.7	30	0	600	1.349	19	Sanhueza_et_al._(1994)
Venezuela	4.6	20	27	0	200	0.006	5	Sanhueza_et_al._(1990)
Venezuela	4.6	20	27	0	200	0.258	5	Sanhueza_et_al._(1990)
NC, USA	5.7	17.5	25.6	0	21	0.02	13	Aneja_et_al._(1995)
NC, USA	6.2	13.15	27.5	0	84	0.046	13	Aneja_et_al._(1995)
NC, USA	6	5.5	35	0	173	0.097	13	Aneja_et_al._(1995)
Tennessee, USA	5.8	25	11.5	1	100	0.04752	20	
Tennessee, USA	5.5	25	27.3	0	111	0.327	9	Valente_and_Thornton_(1993)
Tennessee, USA	5.7	25	26.2	0	111	0.311	15	Valente_and_Thornton_(1993)

Table 3S. (continued).

Costa Rica	5.1	70	25.8	0	360	10.7	365	Veldkamp_and_Keller_(1997)
Costa Rica	5.1	72.9	25.8	0	300	4.643	365	Veldkamp_et_al._(1998)
Costa Rica	5.1	72.9	25.8	0	300	5.413	365	Veldkamp_et_al._(1998)
Costa Rica	5.1	72.9	25.8	0	300	6.973	365	Veldkamp_et_al._(1998)
Costa Rica	5.1	72.9	25.8	0	300	7.483	365	Veldkamp_et_al._(1998)
Costa Rica	5.1	72.9	25.8	0	300	8.383	365	Veldkamp_et_al._(1998)
NC, USA	6	5.6	27.3	0	70	0.025	7	Aneja_et_al._(1998)
NC, USA	6	12.6	24.8	0	190	0.029	6	Aneja_et_al._(1998)
NC, USA	6	11.3	22.6	0	197	0.066	9	Aneja_et_al._(1998)
Colorado, USA	5.9	4.8	27.2	1	100	0.14688	17	Williams et al (1991)
England	7	22	12	0	200	0.79	365	Yamulki et al (1995)
England	7	22	10	0	150	0.64	365	Yamulki et al (1995)
Jiangsu, China	6.5	41	9	1	66	0.53	240	Yamulki et al (1995)
Jiangsu, China	6.5	41	9	0	66	0.485	240	Zheng et al (2003)
Jiangsu, China	6.5	34	5	0	29	0.03	240	Zheng et al (2003)
Jiangsu, China	6.5	34	17	0	96	4.22	240	Zheng et al (2003)

CHAPTER 6 Conclusions

6.1. Conclusions

Nr includes N2O, NH3, and NOx. They are biologically active, chemically reactive, and radiatively active in the Earth's atmosphere and biosphere, which are different from non-reactive nitrogen gas. In this thesis work, three statistical model (N2O_STAT, NH3_STAT, and NO_STAT) are developed for characterizing atmospheric Nr emissions from agricultural soils. The results indicate that, in comparison to other data sets, the model generates a lower global N2O estimate by 9-20% (N2O_STAT: 3.75 Tg N yr-1; EDGAR: 4.49 Tg N yr-1; FAO: 4.07 Tg N yr-1), a lower global NH3 estimates by 57%, (NH3_STAT: 14.2 Tg N yr-1; EDGAR: 33.0 Tg N yr-1), and a lower global NOx estimates by 87%, (NO_STAT: 0.2 Tg N yr-1; EDGAR: 1.6 Tg N yr-1). We also performed a region-based analysis (U.S., India, and China) using the N2O_STAT and NH3_STAT models. Based on results, statistical models can capture the spatial distribution of global Nr emissions by utilizing a more simplified approach than those used by other readily available data sets, but all three models generated lower estimates compared to other inventories.

For N2O_STAT model, we collected the measured N2O emissions reported from previous studies that correspond to the use of fertilizer and manure on particular soil properties, which does not include emissions from leaching and surface runoff as such information is not often available. Therefore, our estimate is only driven by the direct N2O emissions from agricultural soils. However, EDGAR and FAOSTAT incorporate both direct and indirect N2O emissions in estimating the global emissions. Moreover, EDGAR includes rice cultivation and crop residue in its estimate (IPCC, 1996). Another key difference between N2O_STAT and other

data sets lies in the methodology of collecting the model inputs. The N2O_STAT model uses the measured N2O emissions reported from field experiments. This is different from other data sets that derive their emissions by using intermediate data sets (e.g., fertilizer production, livestock counts) and apply measured emission factors to the corresponding data sets.

For NH3_STAT, the model generated lower estimates compared to EDGAR, but the difference varies in difference areas. For the U.S., our model produces an estimate that is 143% higher in comparison to EPA, while 77% and 141% lower in India and 36-482% lower in China compared to other datasets. The difference in the global estimates is attributed to the lower estimates in major agricultural countries like China and India. Also, the underestimation of NH3 emissions in comparison to EDGAR can be due to other sources that are excluded in our model.

For NO_STAT, statistical model captures the spatial distribution of global NOx emissions but the model estimatess is far below other model estimate and literature survey. Two reasons can contribute to this underestimation. One reason is that the underestimation of NO emissions in comparison to EDGAR can be attributed to EDGAR has additional sources in their estimate, e.g. Enteric fermentation, Manure management, Rice cultivation, whereas NO_STAT is exclusive to emissions from fertilizer and manure applied as fertilizer. The other reason is that most NO field measurement campaign are short-term and non-continuous.

6.2. Limitation and Future Work

Literature survey demonstrates that there is difficulty of using an emission factor approach to compute Nr emissions. In fact, this work provides an opportunity to estimate Nr emission factor from agricultural soils. Our model diagnostic tests indicated that Nr emissions

are a nonlinear function of N input. However, our model can be used to estimate a median value of Nr emission factor by calculating the ratio between Nr emission and applied N fertilizer in every grid cell of the map.

While statistical model provide an innovative and relatively simple way to estimate global Nr emission from agricultural sources, there are some limitations . These statistical models only considers physicochemical variables of the emissions, excluding the soil management practices that might contribute to the emissions. In addition, differences between animal types and fertilizer types, and crop types are not taken into account. The underestimation of Nr emissions in comparison to other data sets and the fact that statistical models may be still missing some key N inputs to its variables should be taken into account in the future work. Another component worth investigating is soil biological activity that would more fully represent the processes governing the Nr emissions. Therefore, future work can be focused on including other important physicochemical variables of the emissions, e.g. nitrogen content, organic carbon content, taking into account the differences due to different measure method and process, and including other key N input.

Notwithstanding the above arguments, the N2O_STAT, NH3_STAT, and NO_STAT use measured values for Nr emissions including N content in the fertilizer to develop the model. In addition, the correlations between emissions and most physicochemical variables are at a high significance level (90%), suggesting that these variables are likely affecting the emissions despite excluding other variables. These models provide an opportunity to predict the impacts of climactic Nr emissions.

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