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Key Points:
- Estimates global ammonia emissions from agricultural soils
- Predict future ammonia emissions from agricultural soils in a changing world
- Develops a statistical model for ammonia emissions from agricultural soil

Supporting Information:
- Supporting Information S1

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Characterization of the Global Sources of Atmospheric Ammonia from Agricultural Soils

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Abstract Global ammonia (NH_3) emissions to the atmosphere are projected to increase in the coming years with the increased use of synthetic nitrogen fertilizers and cultivation of nitrogen-fixing crops. A statistical model (NH_3_STAT) is developed for characterizing atmospheric NH_3 emissions from agricultural soils and compared to the performance of other global and regional NH_3 models (e.g., Emission Database for Global Atmospheric Research, Magnitude and Seasonality of Agricultural Emissions, MIX, and U.S. Environmental Protection Agency). The statistical model was developed from a multiple linear regression between NH_3 emission and the physicochemical variables. The model was evaluated for 2012 NH_3 emissions. The results indicate that, in comparison to other data sets, the model provides a lower global NH_3 estimate by 58%, (NH_3_STAT: 13.9 Tg N yr^{-1}; Emission Database for Global Atmospheric Research: 33.0 Tg N yr^{-1}). We also performed a region-based analysis (United States, India, and China) using the NH_3_STAT model. For the United States, our model produces an estimate that is a ~1.4 times higher in comparison to the Environmental Protection Agency. Meanwhile, the NH_3_STAT estimate for India shows NH_3 emissions between 0.8 and 1.4 times lower when compared to other data sets. A lower estimate is also seen for China, where the model estimates NH_3 emissions 0.4 to 5 times 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 statistical model captures the spatial distribution of global NH_3 emissions by utilizing a simplified approach compared to other readily available data sets. Moreover, the NH_3_STAT model provides an opportunity to predict future NH_3 emissions in a changing world.

1. Introduction

Reactive nitrogen compounds include chemically reduced inorganic nitrogen such as ammonia (NH_3) and ionic ammonium (NH_4^+); chemically oxidized inorganic nitrogen such as nitrogen oxides (NO_x), nitric acid (HNO_3), the nitrate ion (NO_3^-), and nitrous oxide (N_2O); and organic nitrogen such as urea, amino acids, and proteins. Many of these are biologically active, chemically reactive, and radiatively active in the Earth's atmosphere and biosphere, in contrast to nonreactive nitrogen gas (N_2).

Among all the reactive nitrogen species, NH_3 is a particularly important component of the global nitrogen cycle, as well as a large anthropogenic source of reactive nitrogen (Battye et al., 2017; Fowler et al., 2015; Galloway et al., 2004; Sutton, Howard, et al., 2011; Sutton, Oenema, et al., 2011). Its emission is primarily from agricultural sources (Aneja et al., 2009; Houlton et al., 2019; Schlesinger & Hartley, 1992; Sutton, Howard, et al., 2011; Sutton, Oenema, et al., 2011). A large loss of reactive nitrogen as ammonia (NH_3), mainly from the use of animal manure and synthetic fertilizer, is a major problem for agricultural systems and the environment (Aneja et al., 2008; Bouwman et al., 2011). Nitrogen loss into the environment represents a large economic cost in lost fertility. If the majority of the NH_3 emissions can be prevented or recovered, the efforts could save a significant amount of money. Assuming approximately 33% nitrogen-use efficiency by crops, the savings is estimated at about $16 billion annually (Raun & Johnson, 1999).

Loss of ammonia from agricultural soils impacts not only agricultural systems, but also adjacent and downwind ecosystems. NH_3 reacts with other atmospheric compounds and serves as a precursor of fine particulate matter ($PM_{2.5}$), which is listed as one of the six crucial atmospheric pollutants by U.S. Environmental Protection Agency (U.S. EPA). Exposure to high $PM_{2.5}$ 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, NH_3 is deposited in terrestrial, aquatic and marine ecosystems, where it causes ecosystem degradation (Bobbink et al., 2010). Through indirect microbial transformations, NH_3 deposition also contributes to the elevated levels of N_2O emission by increasing the rate of nitrogen cycling in soils (Davidson, 2009). N_2O is one of the most important greenhouse gases, which contributes to the global warming. From an economic perspective Sutton, Howard, et al. (2011) estimated that environmental pollution related to excess reactive nitrogen emissions costs the European Union between 77 billion and 354 U.S. dollar annually.

Sutton, Howard, et al. (2011), Sutton, Oenema, et al. (2011), Battye et al. (2017), and Houlton et al. (2019) describe that reactive nitrogen is one of the major environmental challenges of the 21st century. This is owing to the fact that most of reactive nitrogen species are not well regulated. 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. From 2002 to 2016, there have been substantial increases in atmospheric NH_3 emissions over several of the world's major agricultural regions, for example, US (2.61% yr^{-1}), the European Union (1.83% yr^{-1}), and China (2.27% yr^{-1}) (Warner et al., 2017). Y. Li et al. (2016) report an increasing proportional deposition of NH_4^+ from the atmosphere over the United States, reflecting the increasing emissions of NH_3. Similar trends are seen in China (Zhan et al., 2015).

NH_3 emitted from agricultural activities has the largest contribution to the atmospheric budget of reactive N (Galloway et al., 2004; Reis et al., 2009). There are other sources of NH_3, including biomass burning, industrial processes, vehicular emissions, and volatilization from soils and oceans. Currently, the estimated global NH_3 emission is about 54 Tg N yr^{-1} (Bouwman et al., 1997). Anthropogenic sources (primarily from animal and crop agriculture) contribute about 80% of global NH_3 (Aneja et al., 2008). 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's energy is used to produce the annual 120 Mt 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 provides more food for the growing human population; but enhanced N-fixation due to human activities also impacts the nitrogen cycle at global, regional, and local levels.

Nitrification in agricultural soils has the effect of reducing the release of NH_3 to the atmosphere, and some of the NO_3^- produced is later denitrified. However, the contribution and increases of reactive nitrogen by the Haber-Bosch process are not fully offset by denitrification. From the global perspective, Battye et al. (2017) estimate that total denitrification flux (terrestrial ecosystems, fresh water and ground water, and marine ecosystems) ranges from ~210 to ~ 720 Tg N yr^{-1}. The upper end of this range 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 NH_3 emission are necessary for global inventories. Global estimates are also important to develop better models to assess the impact of NH_3 emission on the atmosphere and the deposition of ammonia in terrestrial ecosystems. However, the NH_3 emissions estimates used in current modeling efforts are subject to considerable uncertainties (Battye et al., 2003). Recent efforts to provide regional and global estimates of NH_3 emissions are based on emission "factors" that are associated with different types of fertilizer, which can be multiplied by the fertilizer applied over agricultural regions. The estimates from one such database (Emission Database for Global Atmospheric Research, EDGAR) compare well to satellite-based estimates from the Infrared Atmospheric Sounding Interferometer (van Damme et al., 2018).

This paper develops a statistical model (NH_3_STAT) to predict NH_3 emissions using the physicochemical properties of agricultural soils from different regions and analyzes the spatial distribution of NH_3 emissions from agricultural soils. Our model is different from other models following a metamodeling approach, such as Ramanantenasoa et al. (2019) who utilized emission simulations from a process-based model to generate

its NH_3 emissions. Rather, our model uses measured NH_3 emissions from field experiment as the inputs. In addition, our model did not employ parameterizations of biosphere-atmosphere exchange (e.g., Flechard et al., 2013), since our model was developed from a multilinear regression of various physicochemical properties of agricultural soils. In order to assess the performance of our model, we compare the results with current emissions inventories (e.g., EDGAR, Magnitude and Seasonality of Agricultural Emissions (MASAGE), and MIX) and other regional estimates (e.g., U.S. EPA) of ammonia emissions.

2. Materials and Methods

Figure 1 is a flowchart that demonstrates the methodology we adopted to estimate the global emissions of NH_3 emissions from agricultural soils. Many factors affect NH_3 emission from soil (Flechard et al., 2013; Freney et al., 1983). For example, ammonia emission increases with increasing nitrogen content in fertilizer and pH in soils. It also increases exponentially with soil temperature (Riddick et al., 2016). NH_3 emission also requires a minimum level of soil moisture for microbial activities (Warner et al., 2017).

Based on an initial literature review, we found four variables of importance in controlling NH_3 emissions from soil: (1) soil moisture, (2) soil and air temperature, (3) synthetic and organic fertilizer usage, and (4) soil pH (Granli & Bockman, 1994). We performed a series of statistical analyses utilizing the R-studio v.1.2.1335 (https://www.rstudio.com/) to examine the distribution of data and to fit an appropriate regression model, using NH_3 as the response (dependent) variable and other variables as independent predictors. In running the R-studio, we did not utilize any specific library to perform the statistical analysis. We used the built-in multilinear regression model to determine the parameter coefficients and the p values. Lastly, we used the Integrated Land and Water Information System v.3.31 Academic (https://www.itc.nl/ilwis/download/ilwis33/), a GIS tool, to provide gridded global data sets of the independent variables and apply the statistical model to predict NH_3 emissions.

3. Data

3.1. Data Collection

Data were derived from two sources: (1) field data for developing the statistical model, and (2) global data for extrapolating the results to larger areas. Table 1 lists the parameters used in the regression analysis, and Table 2 delineates the sources of global data. For the field data, we rely extensively on the ammonia emissions gathered by ALFAM2, which is based on a comprehensive literature review regarding agricultural soil NH_3 emission (Hafner et al., 2018). We supplemented that data set with additional emission measurements published after 2000 that we obtained from the literature. We collected 520 sets of data to develop our model. In order to be included in the model, these data sets have to provide all variables listed in Table 1. From the collected data sets, variables taken from ALFAM2 are the results from field experiments conducted in North America and western Europe. These data sets are supplemented other field experimental results from China and India, as well as additional results from North America and Europe. Results reported in these papers were obtained from field experiments conducted in different timeframes. However, our statistical model is designed to be independent of time, as reflected by the equation later shown in the next section. Our model explored the measured physicochemical variables of the agricultural soils as the inputs in the equation to determine the NH_3 emissions empirically.

Prior to using these data, we synchronized the units for all parameters by converting them into the units summarized in Table 1. This is particularly important for NH_3 emissions and N inputs from fertilizer and manure as the values given in the literatures were reported using different units. The complete data set used in this analysis is compiled in Table S1 in the supporting information. For the parameters in Table 2, soil temperature and soil moisture are based on the year 2012. The original maps for cropland and manure fractions are based on the year 2000. However, these maps were adjusted by accounting for the changes in fertilizer inputs and cropland area between 2000 and 2012 using the FAO global fertilizer data for both years. The only parameter that was not based on 2012 is soil pH because there is no global map of soil pH available for that particular year.

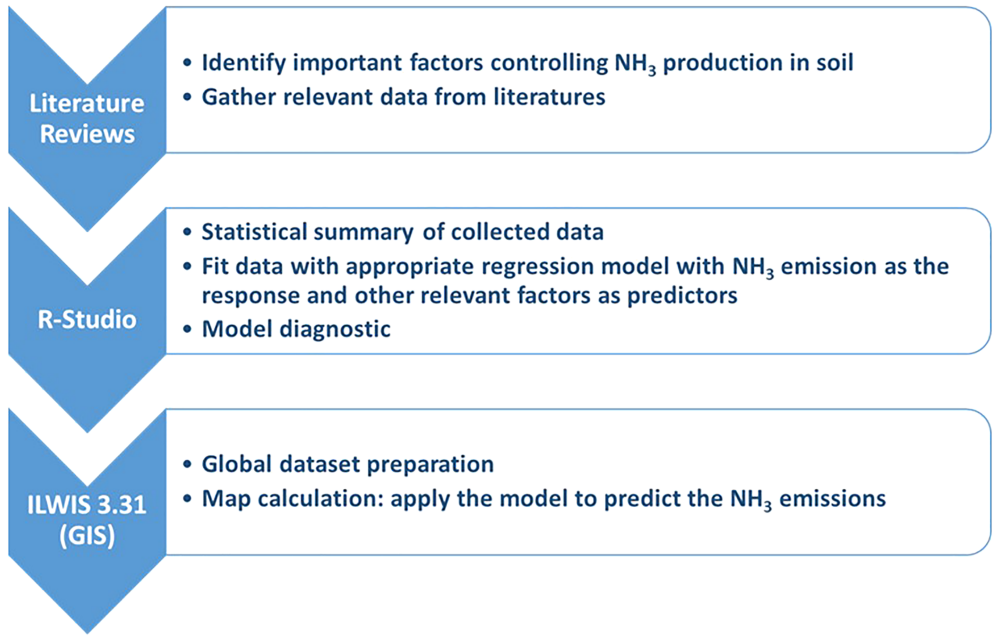


Figure 1. Flowchart of the methodology for the development of NH_3 global emissions.

3.2. Model Development

Based on the statistical analysis, we found that the data distribution, shown as the histogram of NH_3 emissions (Figure 2a), was skewed to the right. To normalize the data, we transformed each to its logarithmic value (Figure 2b) and determined that the data were then close to a normal distribution. Then, we used a multiple linear regression analysis to fit the response variable (the log of NH_3 emissions), using the physicochemical variables as predictors. The statistically derived model (hereinafter: NH_3_STAT) to predict NH_3 emissions from agricultural soils is mathematically expressed as the following:

$$\text{NH3 emission} = (\exp\,[\text{A} + \text{B}\times\text{T}_{\text{soil}} + \text{C}\times\text{SM} + \text{D}\times\text{pH}_{\text{soil}} + \text{E}\times\log(\text{N input}) + \text{F}\times\text{Fertilizer type}])\times\frac{14}{17} \quad (1)$$

where T_{soil} refers to soil temperature (°C), *SM* soil moisture (%), and the coefficients *A*, *B*, *C*, *D*, *E*, and *F* are statistically derived parameters (Table 3). We did not specify the types of N input used in the field experiments since they vary from one experiment to another. We rather used a binary code 0 and 1 to generalize the types of N input. N input is differentiated by synthetic fertilizer (labeled 0) or manure/organic fertilizer (labeled 1) and is expressed as kg N ha^{-1} yr^{-1}. The way the equation is calculated depends on whether each of the paired data used synthetic or manure fertilizer; if synthetic fertilizer is used as the N input, the equation is reduced to only account for five variables excluding the fertilizer type. The units for predicted NH_3 emission are kg N ha^{-1} yr^{-1}.

Table 3 summarizes the coefficients and *p* values of each variable. Based on the *p* value, 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. An *F* test shows that this multivariate linear regression model is statistically significant (95% confidence level).

Table 1
Data for Statistical Model Development

Parameter	Unit
NH_3 emissions	kg N ha^{-1} yr^{-1}
Soil temperature	°C
Soil pH	—
Soil moisture	%
Fertilizer/manure N content usage	kg N ha^{-1} yr^{-1}

Because the multiple regression was performed using the log of NH_3 emissions, NH_3_STAT is a multiplicative model, in which the predicted emission rate is given as a product of parameter functions. The nitrogen input term was also transformed prior to the regression analysis, resulting in an improvement of *R*-squared value. With this log transformation, NH_3_STAT takes the form of a power law relationship between

Table 2
Global Data Set Used for Prediction

Parameter	Data sets
Soil temperature; Soil pH	Harmonized World Soil Database v 1.2 (2012) http://www.fao.org/soils-portal/soil-survey/soil-maps-and-databases/harmonized-world-soil-database-v12/en/ (2000)
Soil moisture	The Global Soil Dataset for Earth System Modeling (2012) http://globalchange.bnu.edu.cn/research/soilw
Cropland cover, fertilizer usage, manure usage	EarthStat's Cropland and Pasture Area (cover from 2000, fertilizer and manure usage and 2012) http://www.earthstat.org/

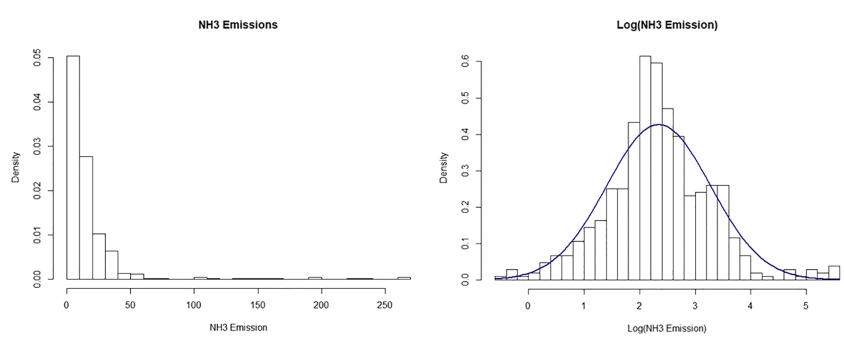
emissions and nitrogen input, where the predicted NH_3 emission rate is proportional to the term (N input)$^{1.1665}$. It is notable that the exponent of the nitrogen input term is greater than 1.0. An exponent of 1.0 would correspond to a standard emission factor where emissions are directly proportional to the amount of nitrogen applied. A higher exponent is compatible with the premise that emission rates increase when nitrogen fertilizer is applied in amounts larger than needed by the crops being fertilized.

3.3. Model Validation

From the comprehensive data set we have collected for this study, we selected eight data sets to be used for validating our model. These data represent results from the field experiments undertaken in different countries in Europe. We chose European countries because we did not intend to perform any regional analysis for Europe. Figure 3 shows the scatterplot between the measured NH_3 (i.e., results from the field experiment) and the predicted NH_3 (i.e., calculated emissions from NH_3_STAT). From this figure, the predicted NH_3 emissions from our model are, in general, in agreement with the measured NH_3, with $R^2 = 0.68$. The normalized mean bias is −8.3%, indicating the our model is slightly underestimating the emissions. Overall, the model validation suggests that our model performance is reasonably good for use in estimating NH_3 emissions from agricultural soils.

3.4. Model Diagnostics

After running the regression analysis, we verified that NH_3_STAT works well for fitting the data and complies with the assumptions of linear regression. Figures 4a and 4b collectively give the results of the model diagnostics. A two-step model diagnostic was performed by analyzing the variance and distribution of the residuals. The QQ plot shown in Figure 4a shows that most of the data follow a straight line, suggesting that these data have a normal distribution. Meanwhile, the equal distribution with no distinct patterns of residuals around the horizontal (Figure 4b) provides a good indication of a linear relationship. Additionally, the majority of standardized residuals are clustered between −1 and 1, suggesting that the data are normally distributed. The outliers indicated by residuals that are far away from the line were excluded in further analysis.



(a) (b)

Figure 2. Histograms of NH_3 emissions representing (a) original values and (b) logarithmic values. The latter resembles a normally distributed data that is more appropriate for the statistical modeling.

Table 3
Summary of the Statistical Model

Coefficient	Parameter	Coefficient value	P value
A	Intercept	−4.6414	$<2 \times 10^{-16}$
B	Soil temperature	0.0150	0.021
C	Soil moisture	0.0066	0.009
D	Soil pH	0.0863	0.049
E	Nitrogen input	1.1665	$<2 \times 10^{-16}$
F	Fertilizer type	0.5086	0.001

3.5. Other NH_3 Emission Inventories

EDGAR: The EDGAR compiles anthropogenic global NH_3 emissions and trends from 1970 to 2012 based on international statistics and emission factors (Janssens-Maenhout et al., 2017). For comparison purposes, we use the 2012 global NH_3 data set for agricultural soils (subsector 4C + 4D). EDGAR also includes emissions from sources ancillary to direct additions of nitrogen fertilizer to the soil, such as emissions from manure management and the cultivation of rice.

EPA National Emission Inventory 2014 Version 2: We calculated NH_3 emission from agricultural soils over the continental United States based on the county-level emission data. (EPA, 2018) We selected the emission from the source classification codes (SCCs) listed in Table 4. The information consists of NH_3 emissions from different types of fertilizer, manure from different types of animals, and fractions from unclassified sources.

MASAGE: We also compare the results from NH3_STAT to results from the MASAGE model (Paulot et al., 2014). In MASAGE inverse modeling is performed with wet deposition fluxes of NH_4^+ for the period 2005 through 2008 in order to create an optimized emission inventory for NH_3. The inventory is global in scope with a grid resolution of 2° latitude by 2.5° longitude.

MIX: The MIX emissions inventory is mosaic of regional emissions inventories for Asian countries (M. Li et al., 2017). For NH_3 emissions in China, MIX uses a high-resolution inventory developed by Peking University, PKU-NH3, which is derived using a process model. For NH_3 emissions in India, MIX uses the Regional Emission Inventory in Asia, Version 2.1 (REAS2).

4. Results

4.1. Global

After calculating the coefficients of all variables, we applied the NH_3_STAT model for each grid cell (5 arcmin × 5 arcmin) and generated a global map for NH_3 emissions in kg NH_3-N yr^{-1} per grid cell (Figure 5a). The resolution of this map is 5 arcmin, which is equivalent to 8,464 ha. Figure 5b is spatial distribution of global NH_3 emission from agricultural soil based on EDGAR. NH_3_STAT estimates total annual global NH_3 emissions from agricultural soils 13.9 Tg N yr^{-1}, compared with 33.3 Tg N yr^{-1} for EDGAR. This difference can be partially attributed to the other sources included in EDGAR but excluded from our model. These sources include agricultural waste burning, livestock housing, and livestock manure storage, treatment, and handling prior to land application (Crippa et al., 2018), which also contribute to NH_3 emission from the agricultural sector (Schlesinger & Bernhardt, 2013). Most emissions inventories do not give a breakdown of animal waste emissions between housing, waste storage, handling and application, even though these emissions can be substantial. In the case of swine, for instance, combined emissions from housing, waste storage, and waste handling have been estimated at 3 to 4 times the magnitude of emissions from waste application (McQuilling & Adams, 2015).

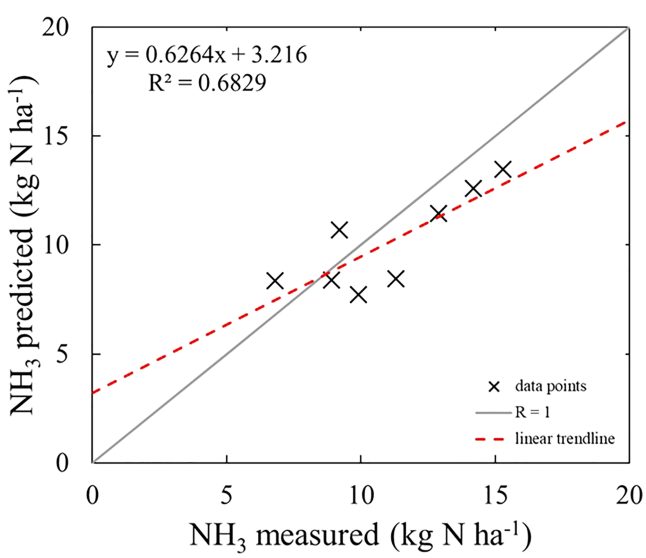
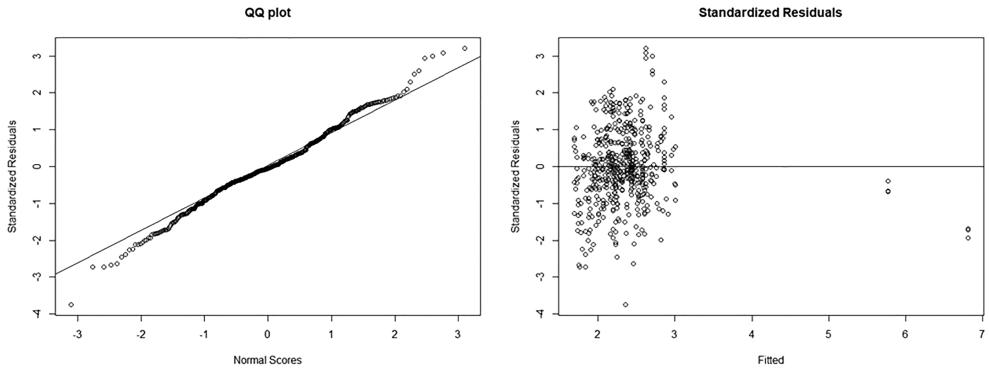


Figure 3. Model validation for NH_3_STAT against NH_3 emissions from field experiments.

It is encouraging to see that, in general, the model captures the global spatial pattern in NH_3 emissions well. Existing global estimates of NH_3 emission from fertilizers ranges from 9 to 12 Tg N yr^{-1}, a range that is slightly lower than our estimate (Bouwman et al., 1997; Schlesinger & Bernhardt, 2013). The National Research Council (2003) estimate for NH_3 from global agriculture and natural land is 12.6 Tg N, which is similar to our estimate. In comparison with, NH_3_STAT, the EDGAR database gives slightly higher NH_3 emissions in western North America, Brazil, India, and China, and lower values for Eastern Europe and the Great Plains of the United States (Figure 5c).



(a) (b)

Figure 4. Model diagnostic for (a) Q-Q Plot and (b) standardized residual.

As noted above, our model includes only soil emissions, while most other NH_3 emissions inventories include other sources associated with animal husbandry, such as animal housing and waste storage. This makes direct comparisons difficult for animal waste emissions. However, our estimates of emissions from mineral fertilizers can be directly compared with other inventories. Our model does not incorporate differences

Table 4
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 (multigrade 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)

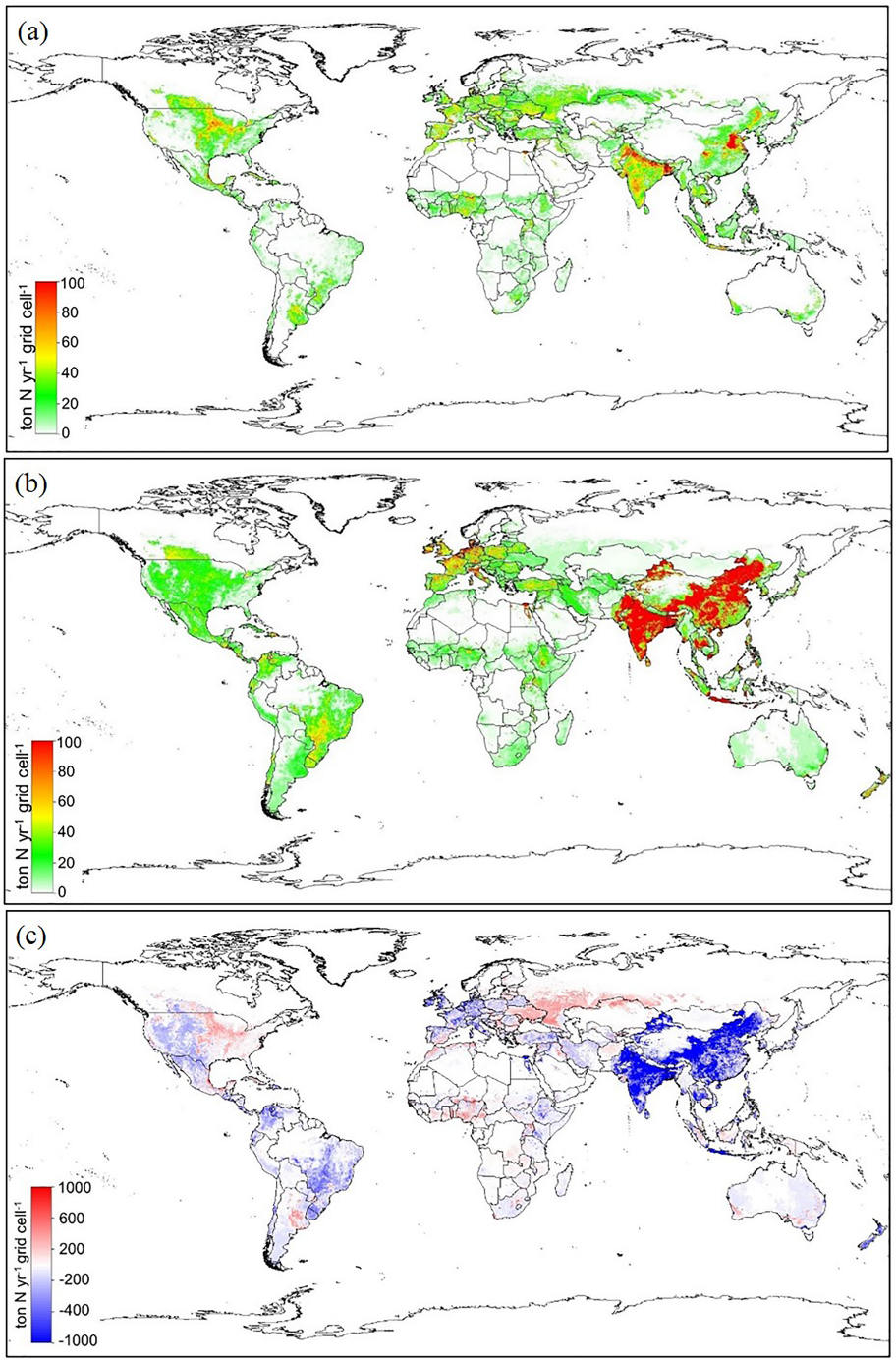


Figure 5. Comparison between the results from (a) NH_3_STAT, (b) EDGAR, and (c) absolute difference between the two models (in t N yr^{-1} per grid cell).

between crop systems, or variations in agricultural practices, such as the cultivation of more than one crop per year, or the use of multiple fertilizer applications in a year. In addition, the data used for NH_3_STAT is drawn heavily from measurements made in the United States. This may account for some of the difference between our estimates and other estimates for China and India, where crop profiles and agricultural practices differ from those in the United States. NH_3_STAT estimates global emissions of NH_3 from mineral fertilizer at 5.8 Tg N yr^{-1}, compared with 24 Tg N yr^{-1} in EDGAR, and 9.4 Tg N yr^{-1} from MASAGE (Paulot et al, 2014).

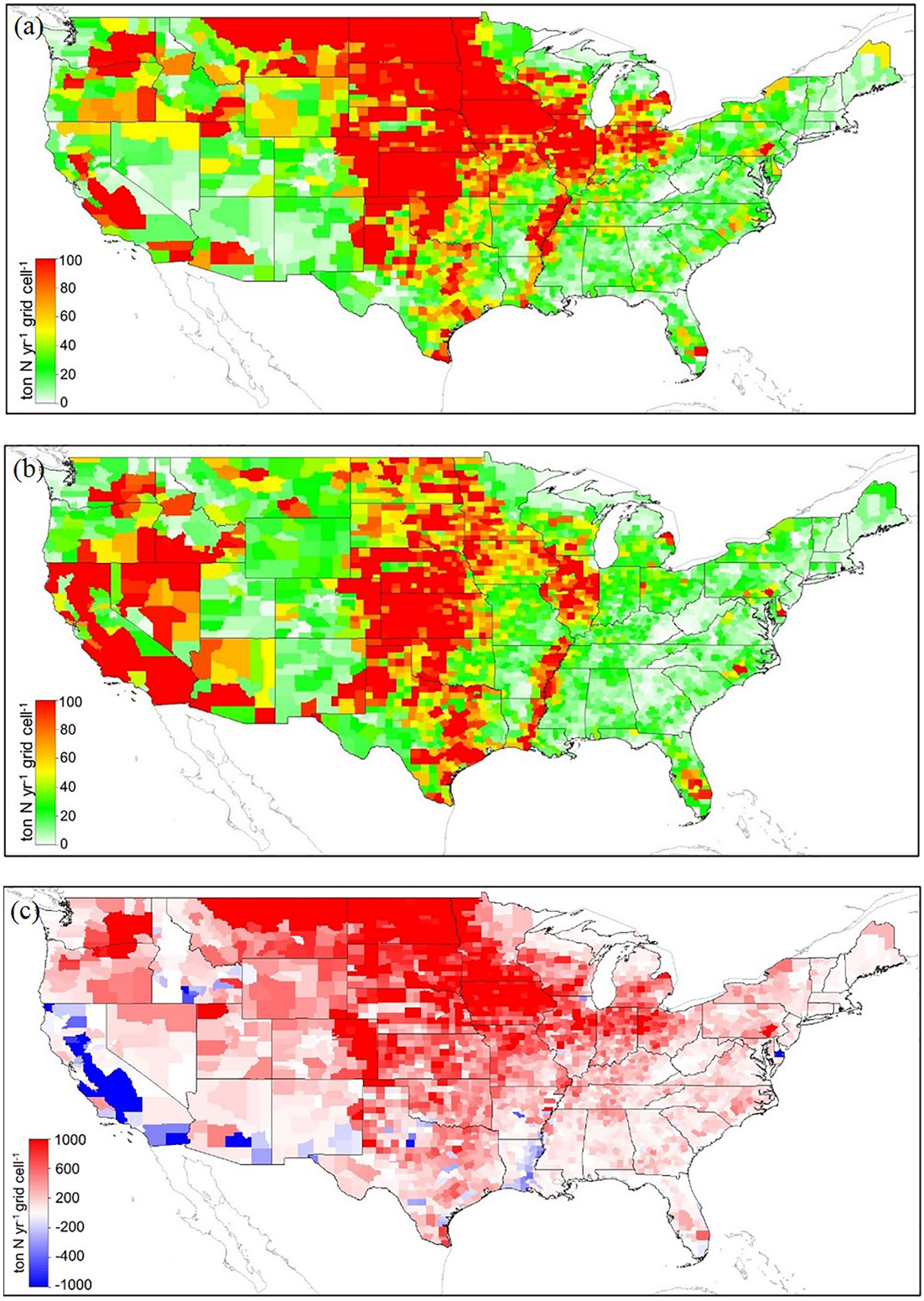


Figure 6. Comparison between the results from (a) NH_3_STAT, (b) EDGAR, and (c) absolute difference between the two models (in t N yr^{-1} per grid cell).

Table 5
Global and Regional NH_3 Emissions (in Tg N) From Agricultural Soils Summarized From Different Emission Inventories

		Global			Continental United States			India			China		
Inventory	Year	Mineral fertilizer	Manure	Total	Mineral fertilizer	Manure	Total	Mineral fertilizer	Manure	Total	Mineral fertilizer	Manure	Total
NH_3_STAT	2012	5.9	8.0	13.9	0.8	1.0	1.8	0.6	0.9	1.5	0.7	1.0	1.7
MASAGE	2008	9.4	24.0	33.4	0.5	1.7	2.2				3.0	4.8	7.8
MIX	2010									6.1			7.4
Beusen et al. (2008)	2000	10–12	16–27	27–38		2.1	2.1						
EDGAR v4.2	2008	24.0	8.6	32.6	1.6	1.0	2.6				6.8	1.9	8.7
Goebes et al. (2003)	1995				0.7								
U.S. EPA (2006)	2002					1.7							
Huang et al. (2012)	2006										2.6	4.4	7.0
Streets et al. (2003)	2000										5.5	4.1	9.6

4.2. Continental United States

Figures 6a–6c show the emission of NH_3 from fertilizer use in the United States using NH_3_STAT. NH_3 emission from agricultural soils calculated from NH_3_STAT and the EPA are 1.8 and 0.7 Tg N yr^{-1}, respectively. Meanwhile, we estimate emissions of 1.8 Tg N yr^{-1}, compared with 2.2 Tg N yr^{-1} estimated by MASAGE (Paulot et al., 2014) and 1.0 Tg N yr^{-1} by the National Research Council (2003).

Our model performs well in capturing the spatial patterns as well as the total annual emission. Figure 5c gives the absolute difference of NH_3 emissions between NH_3_STAT model and EDGAR, and between NH_3_STAT and EPA. Based on Figure 6a, NH_3_STAT underestimates NH_3 emissions in California and some parts in Arizona, New Mexico, and Texas, while it shows higher estimations for most of the U.S., compared with EPA (Figure 6b).

4.3. India and China

We also performed a region-based analysis for India, and China using the NH_3_STAT model. The NH_3_STAT model estimate for India (2.2 Tg N yr^{-1}) shows lower NH_3 emissions than EDGAR (5.3 Tg N yr^{-1}) and Aneja et al. (2012) (3.9 Tg N yr^{-1}), respectively. For China, the model estimates NH_3 emissions from fertilizer (1.7 Tg N yr^{-1}) that are lower than other data sets (e.g., Huang et al. (2012) (3.2 Tg N yr^{-1}), Cui et al. (2013) (10 Tg N yr^{-1}), and Gu et al. (2015) (7.7 Tg N yr^{-1})). NH3_STAT predicts emissions of 0.7 Tg N yr^{-1} from mineral fertilizers in China, compared with 3.0 Tg N yr^{-1} in MASAGE (Paulot et al., 2014).

The statistical model captures the spatial distribution of global NH_3 emissions by utilizing a simplified approach compared to those estimated by other readily available data sets. At the global scale, the statistical model generates a lower global NH_3 estimate than other data sets, by 49% to 63%.

4.4. Emission Analysis

The regional analyses (United States, India, and China) using the NH_3_STAT model indicate that there are variations in ammonia emissions between developed and developing countries. For the United States, our model produces an estimate that is 1.4 times higher in comparison to EPA. Meanwhile, the NH_3_STAT model estimate for India shows NH_3 emissions that are 1.4 times to 0.8 times lower when compared to other data sets. A lower estimate is also seen for China, where the model estimates NH_3 emissions between 0.4 times and 5 times 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. Table 5 shows the summary of NH_3 emissions from different data inventories for both global and regional emissions.

The underestimation of NH_3 emissions in comparison to EDGAR can be attributed to other sources that are excluded in our model. EDGAR includes additional sources of NH_3, for example, enteric fermentation, manure management, and rice cultivation, whereas NH_3_STAT is exclusive to emissions from fertilizer and manure applied as fertilizer to agricultural soils.

The NH_3_STAT uses measured values for NH_3 emissions, considering the N content in applied fertilizer. While this statistical model provides an innovative and relatively simple way to estimate global NH_3 emission from agricultural sources, some limitations exist. This model only considers physicochemical variables of the emissions, excluding the soil management practices that might contribute to the emissions. In addition, the methods used to measure NH_3 emissions in the field were different from one study to another, and these differences are not taken into consideration in developing the statistical model. Nor are differences between animal types, fertilizer types, and crop types taken into account. The correlations between NH_3 emissions and most physicochemical variables are at a high significance level (95%), suggesting that these variables are affecting the emissions despite excluding other variables. Moreover, the NH_3_STAT model provides an opportunity to predict future NH_3 emissions in a changing world, unlike other approaches.

5. Conclusions

The statistical model captures the spatial distribution of global NH_3 emissions by utilizing a simplified approach versus those used by other readily available data sets. At the global scale, the results indicate that the statistical model provides a lower global NH_3 estimate than other data sets, by 57%, (NH_3_STAT: 13.9 Tg N yr^{-1} versus EDGAR: 33.0 Tg N yr^{-1}).

The region-based analysis (United States, India, and China) using the NH_3_STAT model indicates that there are variations in ammonia emissions between developed and developing countries. For the U.S., our model produces an estimate that is 1.4 times higher in comparison to US EPA. Meanwhile, the NH3_STAT model estimate for India shows NH_3 emissions that are 0.7–1.4 times lower when compared to other data sets. A lower estimate is also seen for China, where the model estimates NH_3 emissions between 0.4 and 5 times 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.

Our approach is subject to considerable uncertainty. In particular, our statistical analysis was restricted to parameters, which were available in the measurement literature and in global data bases. Thus, the analysis did not include parameters such as soil composition and porosity, which could also influence NH_3 emissions, or the influence of crop type on emissions. We also do not take into account short-term changes in moisture and temperature, which may result in enhanced NH_3 emissions. Further, data were not available to systematically incorporate differences in agricultural practices, such as the cultivation of more than one crop per year, or the use of multiple fertilizer applications in a year. This may account for some of the difference between our estimates and other estimates for China and India, where two crops are grown per year in many places.

Previous NH_3 emissions inventory approaches are also subject to large uncertainties. In a recent analysis of emission factors from different inventory components by McQuilling and Adams (2015), normalized mean errors ranged from 24 to 61%. The underestimation of NH_3 emissions in comparison to EDGAR can be attributed to other sources that are excluded in our model. This is understandable since EDGAR includes additional sources of NH_3 in their estimate, e.g. enteric fermentation, manure management, and rice cultivation, whereas NH_3_STAT is exclusive to emissions from fertilizer and manure applied as fertilizer to the soil.

The NH_3_STAT uses measured values for NH_3 emissions, including N content in the fertilizer, to develop the model. The correlations between NH_3 emissions and most physicochemical variables are at a high significance level (95%), suggesting that these variables are likely affecting the emissions despite excluding other variables. Moreover, the NH_3_STAT model provides an opportunity to predict future NH_3 emissions in a changing world, unlike other approaches.

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