RESEARCH ARTICLE

# Atmospheric Nitrogen Oxides Emissions from Global Agricultural Soils: Present and Future

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## Abstract

Global nitric oxide (NO) emissions to the atmosphere are projected to increase in the coming years with the increased use of synthetic nitrogen fertilizers and fossil fuel combustion. Here, a statistical model (NO_STAT) is developed for characterizing atmospheric NO emissions from agricultural soil sources, and it is compared to the performance of other global and regional NO emissions (e.g., EDGAR and U.S. EPA). The statistical model was developed using a multiple linear regression between NO emission and the physicochemical variables. The model was evaluated for 2012 NO emissions. In comparison to other data sets, the model provides a lower global NO estimate by 59%, (NO_STAT: 0.67 Tg N $yr^{-1}$; EDGAR: 1.62 Tg N $yr^{-1}$). We also performed a region-based analysis (U.S., India; and China) using the NO_STAT model. For the U.S., the model produces an estimate that is 47% lower in comparison to EDGAR. Meanwhile, the NO_STAT model estimate for India shows NO emissions 75% lower when compared to other data sets i.e. EDGAR (which is a comprehensive emissions inventory used in global/regional air quality modeling, and therefore, we have refered to it as 'other data sets'). A lower estimate is also seen for China, where the model estimates NO emissions 82% 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 NO emissions by utilizing a more simplified approach than those used previously. Moreover, the NO_STAT model provides an opportunity to predict future NO emissions in a changing world. We have made a prediction for future (2050) NO emissions from agricultural soils i.e. emissions from agricultural soils may rise to above ~2.3 TgN/yr, based on anticipated future applications of nitrogen to agricultural soils.


**Keywords:** Calculates global nitrogen oxides emissions from agricultural soils, Develops a statistical model for nitrogen oxides emissions from agricultural soil, Predicts future nitrogen oxides emissions from agricultural soils in a changing world.

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## Introduction

Reactive nitrogen compounds in the atmosphere are defined as nitrogen compounds which are chemically reactive, biologically active, or radiatively active by absorbing infrared radiation or other radiation. These compounds contrast with nitrogen gas ($N_2$), which is non-reactive. Reactive nitrogen compounds include chemically-oxidized inorganic nitrogen such as oxides of nitrogen ($NO_X$), nitric acid ($HNO_3$), the nitrate radical and ion ($NO_3$ and $NO_3$-), and nitrous oxide ($N_2O$); chemically-reduced inorganic nitrogen such as ammonia ($NH_3$) and ionic ammonium ($NH_4$+); and organic nitrogen such as urea, amino acids, and proteins. Sutton *et al.* (2011) have estimated that increased reactive nitrogen emissions costs the European Union between 77 billion and 354 U.S. dollars annually owing to the increased costs associated with environmental management, economic losses, substantial health risks for vulnerable human populations, etc. Moreover, Doering *et al*., (2011), Battye *et al*., (2017), Abrol *et al*., (2017), and Houlton *et al*., (2019) describe that reactive nitrogen loss to the environment is one of the major environmental challenges of the 21st century impacting climate change; energy and food security; air, water and soil quality; and human health.

$NO_X$ emissions contribute to a number of air pollution problems, including smog, tropospheric $O_3$, acid rain, and elevated levels of fine particulate matter ($PM_{2.5}$). $NO_X$ comprises nitric oxide (NO) and nitrogen dioxide ($NO_2$), which are easily interconverted. These are important trace constituents in the troposphere, where they regulate the production and consumption of photochemical oxidants, ozone ($O_3$) and hydroxyl radicals (Warneck, 2000). Tropospheric $O_3$ is a significant air pollution problem in the United States, as well as in most developed and developing countries. It is harmful to both human health and welfare. $NO_X$ is an important precursor to tropospheric ozone. $NO_2$ reacts in air to produce NO and $O_3$. When the NO concentration is below 3-8 ppt, NO reacts with $O_3$ to produce $NO_2$ and $O_2$ thus consuming $O_3$. But when the NO concentration is higher, NO catalyzes the oxidation of $CH_4$, CO and volatile organic compounds (VOC) to produce $O_3$ (Warneck, 2000). NO is recycled to $NO_2$ by free radicals. In rural environments, the reaction of NO with biogenic VOC can be a predominant source of ozone (Aneja *et al*., 1996).

$NO_X$ in the troposphere can be oxidized and react with water to form of nitric acid ($HNO_3$). This contributes to acid rain, which

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directly accelerates acidification and eutrophication processes in regional ecosystems. $HNO_3$ and nitrates in the atmosphere also contribute to elevated levels of $PM_{2.5}$ and regional haze.

The U.S. Environmental Protection Agency (USEPA) has established National Ambient Air Quality Standards (NAAQS) for $O_3$, $NO_2$, and $PM_{2.5}$. Elevated levels of these pollutants can cause irritation to the human respiratory system, aggravate respiratory problems such as asthma, and contribute to chronic obstructive pulmonary disease (COPD). Elevated levels of $PM_{2.5}$ have been associated with premature death for people with heart and lung disease (Lelieveld *et al*. 2015).

$NO_X$ emissions stem from both anthropogenic and natural sources. Nitrogen gas and oxygen combine to form $NO_X$ in lightning and during combustion processes. Microbes in soil also produce NO as they metabolize nitrogen compounds, which may be present naturally in the soil, or enhanced by nitrogen fertilizers (Aneja *et al*., 1996; Aneja *et al*., 2008; Aneja *et al*., 2009; Bray *et al*., 2019; Houlton *et al*., 2019; Schlesinger and Bernhardt, 2020). Fossil fuel combustion is the largest source of $NO_X$ emissions, contributing more than half of the global NO budget.

Soils, especially agricultural soils, are an important source of biogenic NO emissions. A recent estimate of global NO emission from soils is ~21% of the total global sources of NO to the atmosphere (57 Tg N yr-*1*) (Schlesinger and Bernhardt 2020). 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; Aneja *et al*., 1996, 1998).

$NO_X$ emissions from large combustion sources and industrial sources are regulated in the U.S. and many other countries under air pollution programs designed to ameliorate tropospheric $O_3$, acid rain, $PM_{2.5}$, and regional haze. However, emissions of NO from agriculture are unregulated.

Many factors affect the NO emission from soil. Pilegaard (2013) identifies the availability of nitrogen compounds in the soil as a key factor affecting the NO emission rate. This soil nitrogen can be derived from various sources, including nitrogen-fixing bacteria in the soil, deposition of $NH_3$, $NO_X$, acid rain, or other nitrogen compounds from the atmosphere, decay of organic material, or inputs of synthetic nitrogen fertilizer and manure. In agricultural soils, the primary sources of nitrogen are synthetic fertilizers, manure, and nitrogen-fixing crops such as soybeans. In addition to the availability of nitrogen, the NO emission rate is affected by environmental factors, including soil temperature, soil pH, and soil moisture (Sullivan *et al*., 1996). Soil water content controls the rate of $O_2$ supply, which directly affects the nitrification and denitrification. Goldberg & Gebauer (2009) observed that NO emission decreased after precipitation, but increased during 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). There is no direct relationship between the rate of NO emission and soil pH; however, microbial nitrification processes, which convert chemically reduced nitrogen compounds (such as $NH_{4+}$) to chemically oxidized compounds ($NO_3$-), are enhanced at higher soil pH. In addition to other oxidized nitrogen compounds, nitrification processes produce some NO, which can be emitted to the atmosphere.

Global and regional estimates for NO emissions are subject to considerable uncertainty, and emissions estimates cover a wide range between the lowest and highest values. Most measurements are short-term and inherently do not represent the spatial and temporal variation of NO emission. Thus, statistical models for estimating NO emissions from agricultural soil are desirable. Ideally, such models should use readily available input parameters.

Accurate estimates of NO emission are necessary for global inventories. Global and regional estimates are also important for developing better models to assess the impact of NO emission on the atmosphere and the deposition of reactive nitrogen in terrestrial ecosystems. The emissions factor approach is generally used to estimate local, regional and global NO and other species emissions. In this approach, emission factors are computed in terms of the mass of NO emissions per mass of nitrogen applied in fertilizer, for a variety of nitrogen fertilizers and for manure. To compute NO emissions, these emission factors are multiplied by the amount of nitrogen-based fertilizer applied over agricultural regions. A global emission factor of about 1% is calculated by dividing the estimate of global emission from soils (12 TgN/yr) by the turnover of nitrogen in soils (1200 TgN/yr; Schlesinger and Bernhardt 2020).

Aneja *et al*. (2019) and Aneja *et al*. (2020) published nitrous oxide and ammonia emissions respectively from global agricultural soils. This nitric oxide emissions model development and analysis will help advance research in reactive nitrogen emissions from global agricultural systems. The $NO_2$ emission from agricultural soil is negligible, so this paper focuses on the emissions of NO. The goal of this study is to develop a statistical model to predict NO emissions from agricultural soils amended with synthetic and organic fertilizers using physicochemical properties of the soils from different regions. We also analyze the spatial distribution of NO emissions from agricultural soils and compare the results with a modeled emission inventory from EDGAR v.4.3.2. Moreover, the statistical model (NO_STAT model) provides an opportunity to predict future NO emissions in a changing world. We have made a prediction for future (2050) emissions.

## Materials and Methods

The data collection and statistical analysis follow that of Aneja *et al*. (2020). Fig. 1 illustrates the methodology we adopted to estimate NO global emissions from agricultural soil. We conducted a literature review in two parts. The initial review compiled information on the physico-chemical variables controlling the NO emissions. Based on the initial literature review, we identified four variables which are readily available and which would be expected to be of importance in controlling NO emissions from soil: (1) soil moisture content, (2) soil temperature, (3) the amount of nitrogen applied to soils in the form of synthetic fertilizer or manure, and (4) soil pH. A comprehensive literature review was then conducted to gather results from field experiments which measured the NO emission rate accompanied by data for the desired input variables. More on this is discussed in section Data collection.

After compiling available data from field experiments, we performed a series of statistical analyses utilizing the

R-studio statistical software (https://github.com/rstudio/rstudio) to examine the distribution of data. We then developed an appropriate regression model (NO_STAT) with NO as the response (dependent) variable and other variables as independent predictors.

We applied the NO_STAT model to predict NO emissions on a global scale using the Integrated Land and Water Information System (ILWIS) v.3.31 Academic (https://www.itc.nl/ilwis/download/ilwis33/). ILWIS is a Geographic Information Systems (GIS) tool which provides global data sets for the model input parameters.

## Data collection

Data collection included two components: (1) identification of data sets for developing the statistical model using the variables controlling listed in Table 1a and (2) identification of global data sets for extrapolating the results to larger areas (Table 1b). For the statistical model, we conducted a comprehensive literature review regarding NO emissions from agricultural soil. In our statistical model development, we have used results published principally after 1990. For inclusion in the model, studies needed to include information on all of the variables listed in Table 1.a. In all, 94 major studies were identified which measured NO emissions from agricultural soils and also gave measured values for all of the identified controlling parameters. Most of the studies were carried out in North America and western Europe, however these are supplemented by some studies conducted in Asia, South America, and Oceania. The measurement and physico-chemical soil data used in our analysis are compiled in the supplemental data set for this paper, in Table S.

In our statistical model, we empirically relate NO emissions to the identified soil parameters. We then use the model to compute global emissions using the datasets listed in Table 1b. The soil temperature and moisture data sets listed in Table 1b are for the year 2012. Spatial maps of crop cover, fertilizer usage, manure usage, and pH are for the year 2000. However, we have adjusted the crop coverage and fertilizer application rates from 2000 to 2012 using FAO data for both years. These maps were adjusted by accounting for the changes in fertilizer inputs and

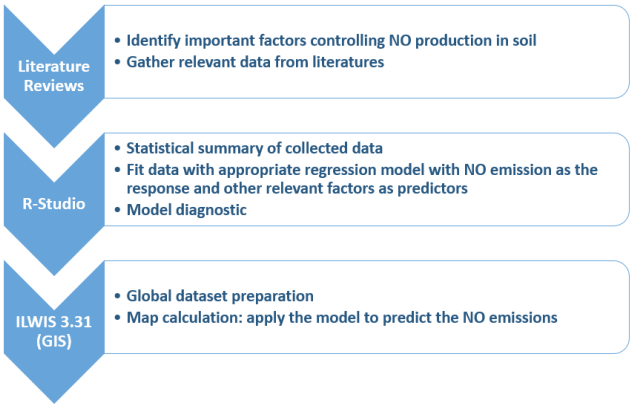


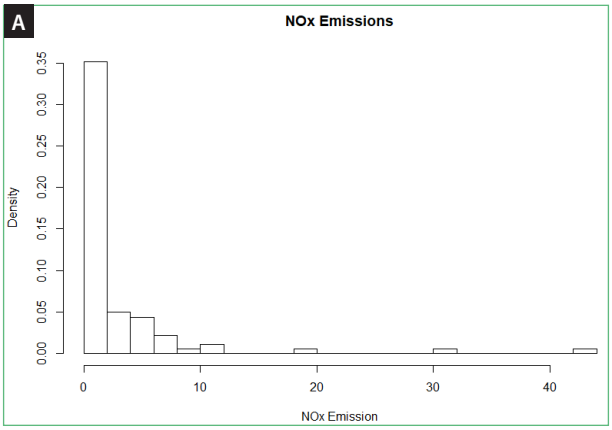

**Fig. 1:** Summary of the NO statistical model development methodology.

**Table 1a:** Data for statistical model development.

| *Parameters* | *Units* |
|---|---|
| NO emissions | kg N $ha^{-1}$ $year^{-1}$ |
| 10 cm - soil temperature | °C |
| Soil pH | - |
| 10 cm - soil moisture | % |
| Fertilizer N content usage | kg N $ha^{-1}$ $year^{-1}$ |
| Manure N content usage | kg N $ha^{-1}$ $year^{-1}$ |

**Table 1b:** Global data set used for prediction.

| **Parameter** | **Data sets** |
|---|---|
| Soil temperature; Soil moisture | ERA-Interim Global Atmospheric Reanalysis https://www.ecmwf.int/en/forecasts/datasets/archive-datasets/reanalysis-datasets/era-interim |
| Soil pH | The Global Soil Dataset for Earth System Modeling http://globalchange.bnu.edu.cn/research/soilw |
| Cropland cover, fertilizer usage, manure usage | 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/ Adjusted EarthStat's Cropland and Pasture Area http://www.earthstat.org/ |



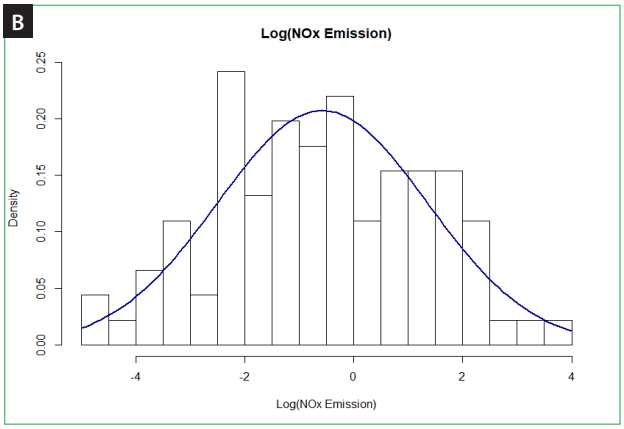




**Fig. 2:** (a) Histogram of NOx emission; and (b) log of NOx emission. (NO emission rate is expressed in kg-N $ha^{-1}$ $yr^{-1}$)

cropland area between 2000 and 2012 using the FAO global fertilizer data for both years. Data were not available to adjust for any changes in soil pH for 2012.

## Model setup

Based on the statistical analysis, we found that the data distribution, shown as the histogram of NO emissions (Fig. 2a), was skewed to the right. To normalize the data, we transformed the data to the logarithmic value (Fig. 2b), for which the data of logarithm of NO emissions appear as a normal distribution. Most data are located between -2 and +2 on the logarithmic scale. We proceeded with the logarithmically transformed dataset using the assumption of a log-normal distribution.

Then, we used a multiple linear regression model to fit the response variable (the log of NO emissions), using the physicochemical variables as predictors. The statistically-derived model (hereinafter: NO_STAT) to predict NO emissions from agricultural soils is mathematically expressed as the following:

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

Where: NO emission rate is expressed in kg-N ha-*1* yr-*1*; Tsoil refers to soil temperature, in °C; SM refers to soil moisture (%), and the coefficients A, B, C, D, E, and F are statistically-derived parameters (Table 2).

Table 2 summarizes the coefficients and p-values of each variable. Based on the p-value, soil moisture, soil pH, and fertilizer usage are statistically significant in the model. The Residual standard error is 1.57, and R-squared is 0.38 (this provides an uncertainty estimate of the NO_STAT model). The F-test shows that this multivariate linear regression model is statistically significant (90% confidence level), but, notably, the magnitude of nitrogen input did not have a significant influence on the NO emission rate. This may seem counterintuitive, because NO emissions are produced by the microbial processes of nitrification and denitrification. Nitrogen concentrations may be the limiting factor in these reactions when the levels of soil nitrogen are low; however, other factors may become limiting when soil nitrogen concentrations are at agronomic levels (Aneja *et al*., 2019; Aneja *et al*., 2020; Schlesinger and Bernhardt, 2020).

In the NO_STAT model development, we used the log of NO emissions to perform multiple regression. The NO_STAT model is a multiplicative model. The model determines the NO emission rate using the product of physicochemical parameters. To enhance the R-squared value of the model, the nitrogen input term was also transformed prior to the regression analysis. Performing this log-transformation allows the NO_STAT model to be transformed into a power law relationship between NO emissions and nitrogen input.

**Table 2:** Summary of the NO_STAT model.

| *Variable* | *Parameter* | *Coefficient* | *p-value* |
|---|---|---|---|
| A | Intercept | 3.675989 | 0.007829 |
| B | Soil temperature | 0.007431 | 0.787556 |
| C | Soil moisture | -0.006302 | 0.0449183 |
| D | Soil pH | -0.749266 | 0.000239 |
| E | Nitrogen input | 0.004374 | 0.035034 |
| F | Fertilizer type | -0.311669 | 0.550954 |

## Model Diagnostics

The efficacy of the NO emission model (NO_STAT) is shown in Fig. 3a and 3b. The model fits the data well and also performs linear regression. We conducted a two-step model diagnostic. First, we analyzed the variance and distribution of the residuals (Fig. 3a). Second, we demonstrated that the data used in this methodology for model development are normally distributed. The QQ plot shown in Fig. 3b verify that the data used in developing the model are normally distributed.

## Dataset

The total annual NO emissions for the globe and for major agricultural regions are made for soils emissions based on the NO_STAT emissions projections and compared with NO emissions from EDGAR.

EDGAR (https://edgar.jrc.ec.europa.eu/report_2020): The Emission Database for Global Atmospheric Research compiles anthropogenic global NO emissions and trends from 1970 to 2012 based on international statistics and emission factors (Janssens-Maenhout *et al*. 2017). The resolution of EDGAR data set is 0.1 deg x 0.1 deg. For $NO_X$, 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.

# Results and Discussion

The $NO_2$ emission from agricultural soil is negligible, so this paper focuses on the emission of NO. After calculating the coefficients of all variables, we applied the NO_STAT model for each grid cell (5 arc-minute x 5 arc-minute) to generate a global map for NO emissions. Fig. 4a gives the spatial distribution of global NO emission from agricultural soils calculated using NO_STAT in kg N yr-1 grid cell-1. The resolution of this map is 5

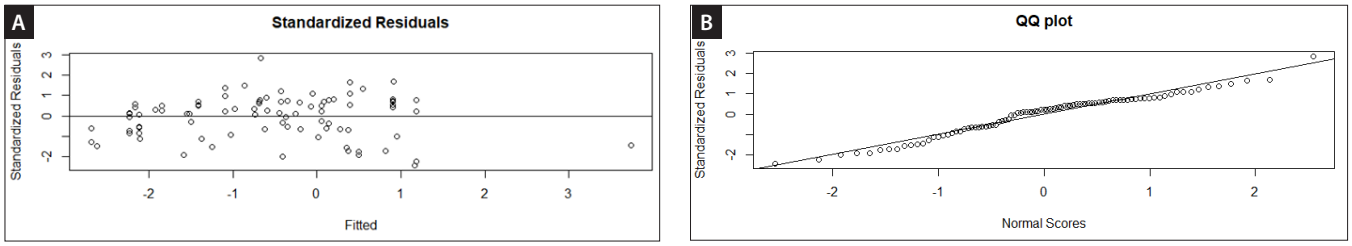



**Fig. 3:** $NO_x$ Model diagnostic: (a) Standardized residual; (b) Q-Q Plot.

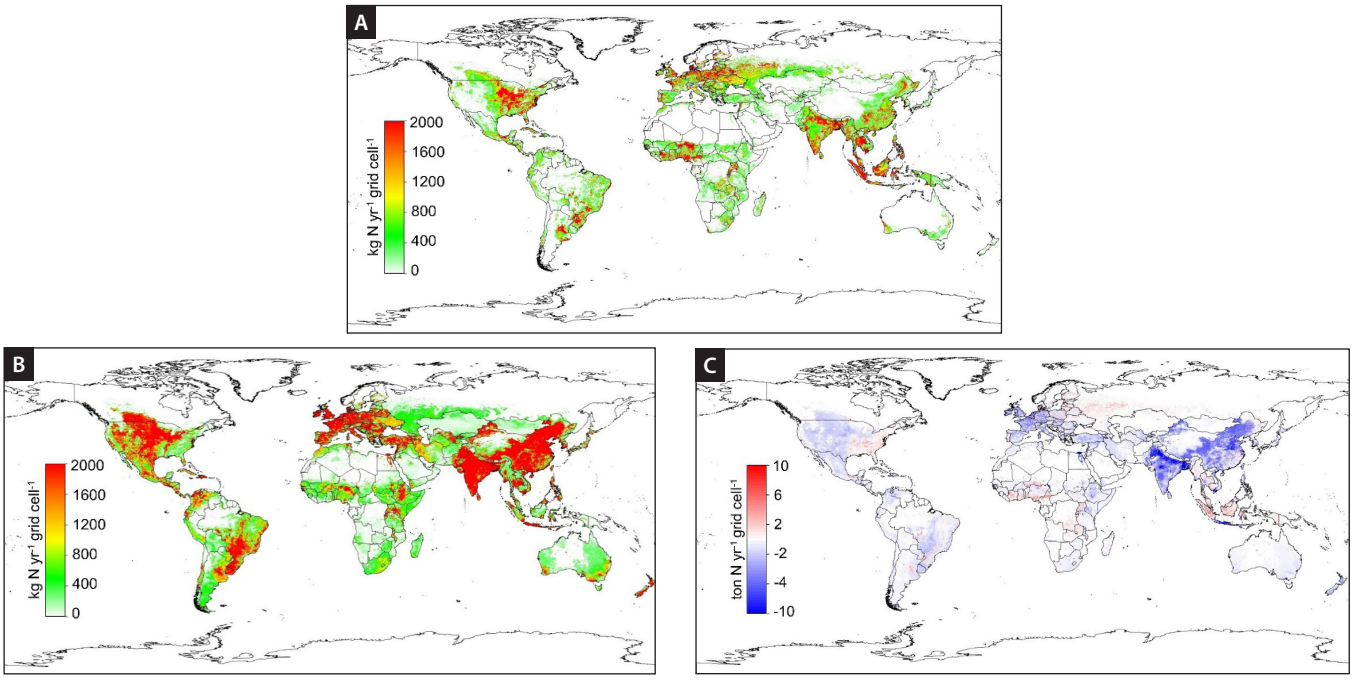



**Fig. 4:** Comparison between the results from (a) NO_STAT, (b) EDGAR, and (c) absolute difference between the two models (in ton N yr *1* grid cell-*1*).

arc-minute, which is equivalent to about 8500 ha at the equator. Figure 4b presents the spatial distribution of global NO emission from agricultural soils based on EDGAR. Total annual global NO emissions from NO_STAT and EDGAR are 0.671 Tg N yr-*1* and 1.623 Tg N yr-*1*, respectively. Based on these two values, the NO_STAT model gives a lower global NOx estimates by 59%. However, it is important to note that our model only accounts for the NO emissions from agricultural soils, which is only one of various factors considered in EDGAR. For the regional emissions, our model also gives NO emission estimates that are lower than EDGAR. Our model estimate for the continental U.S., China, and India are −47%, −82%, and −75% lower than from EDGAR, respectively.

As discussed previously, our lower values may be attributed to other sources that are excluded from our model, whereas EDGAR included these additional sources in its estimates, e.g. Enteric fermentation, Manure management, and Rice cultivation. However, NO_STAT is exclusive to emissions from agricultural soils to which fertilizer and manure are applied as fertilizer. EDGAR treats NO emissions as directly proportional to the amount of nitrogen added to soils in the form of fertilizer and animal waste. Our model suggests that, for agricultural soils, other parameters such as soil moisture and temperature may have a larger impact on nitrogen emissions than the amount of waste or fertilizer applied. Nevertheless, in general, the model shows a similar global spatial pattern in NO emissions. Fig. 4 shows areas of elevated NO emissions(northern China, northern India, and the Mid-West U.S.). Fig. 4c shows the absolute difference between NO_STAT and EDGAR. NO_STAT gives relatively lower NO emission values in all areas.

NO_STAT and EDGAR predict global emissions of NO from agricultural soils that contribute 5.6 and 13.5% to the estimated total global emission of NO from soils (12 TgN/yr) and 1.2 to 2.8% of the total emissions of NO to the atmosphere from all sources (57 TgN/yr; Schlesinger and Bernhardt 2020). The emissions from agricultural soils predicted by NO_STAT and EDGAR suggest Emissions Factors of 0.0045 to 0.0108 relative to global fertilizer applications of 150 TgN/yr to agricultural soils (Schlesinger and Bernhardt 2020). By 2050, emissions from agricultural soils may rise to above 2.3 TgN/yr, based on anticipated future applications of nitrogen to agricultural soils (Galloway *et al*. 2004).

These estimates are subject to much uncertainty. In particular, our statistical analysis was restricted to parameters that were readily available in the literature and in global data bases. Thus, the analysis did not include parameters such as soil composition and porosity, which could also influence NO emissions. We also do not take into account short term variations in moisture and temperature which may result in enhanced NO 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.

Nevertheless, previous NO emissions inventory approaches are also subject to large uncertainties. Based on a literature survey, Hudman *et al*. (2012) used a mechanistic model of global soil NO emissions to estimate that total global NO emissions from soil are 10.7 Tg N yr-*1* and those from fertilizer N input (1.5% of applied N) are 1.8Tg N yr-*1*. This exceeds the EDGAR estimate for NO from the combination of chemical fertilizer and animal wastes applied to agricultural soils.

The NO_STAT model provides a method for computing NO emissions using existing databases on soil, fertilizer usage, and animal waste production (Table 1b). The model also provides insight to importance of different soil parameters in producing NO emissions.

## Conclusion

In this work, a statistical model (NO_STAT) is developed for characterizing atmospheric NO emissions from agricultural soils. As a result of considering only one source of emissions, in comparison to other data sets, the model generates a lower global NO estimates by 59%, (NO_STAT: 0.671 Tg N yr-*1*; EDGAR: 1.6 Tg N yr-*1*). Based on these results, NO_STAT statistical model captures the spatial distribution of global NO emissions by applying a simpler modeling approach based on existing global data sets (Table 1b). However, the model gives lower estimates compared to other inventories.

While this statistical model provides an innovative and relatively simple way to estimate global NO emission in agricultural sources, some limitations exist. This model only considers physicochemical variables of the emissions, excluding the soil management practices and soil microbial activity which may also contribute to the NO emissions from soils. Moreover, differences in field NO emission experimental methodologies were not considered in the development of the NO emission from soils statistical model. Statistical analysis suggests that NO emissions and most physicochemical variables are at a high significance level (95%). Unlike other approaches, the NO_STAT model provides an opportunity to predict future NO emissions owing to global changes e.g. climate, increased use of fertilizers, etc. We estimate that by 2050, emissions from agricultural soils may rise to above ~2.3 TgN/yr, based on anticipated future applications of nitrogen to agricultural soils.

For NO_STAT, the statistical model captures the spatial distribution of global $NO_x$ emissions but the model estimate is below other model estimates and the results of 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 additional sources that EDGAR estimated, e.g. Enteric fermentation, Manure management and 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 campaigns are short-term and non-continuous. Future efforts to apply the NO_STAT statistical model approach to include more detailed descriptions of cropping practices and season variations will be beneficial.

Since nitric oxide in the atmosphere is a precursor to tropospheric ozone, some mitigation options include reduction in N fertilizer use through an increase in fertilizer use efficiency, improved timing of fertilizer application, and enhancing the fertilizer uptake efficiency of crops. This will have the potential to reduce global annual NO emissions.

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## Supplementary Information

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

| *Location* | *pH* | *Soil moisture (%)* | *Temperature (°C)* | *N_type (0: fer 1:man)* | *N_rate (kg N/ ha)* | *NO emission (kg N/ha)* | *Length of Experiment (days)* | *Reference* |
|---|---|---|---|---|---|---|---|---|
| Japan | 5.9 | 39 | 23 | 0 | 200 | 1.5 | 131 | Akiyama *et al* (2000) |
| Japan | 5.9 | 39 | 23 | 0 | 200 | 2.3 | 131 | Akiyama *et al* (2000) |
| Japan | 5.9 | 39 | 23 | 0 | 200 | 2.4 | 131 | Akiyama *et al* (2000) |
| Colorado, USA | 7.5 | 14.3 | 28 | 0 | 63 | 0.2 | 64 | Anderson *et al* (1987) |
| Virginia, USA | 6 | 25 | 15 | 0 | 40 | 1.3 | 350 | Anderson *et al* (1987) |
| Virginia, USA | 6 | 25 | 15 | 0 | 40 | 1.5 | 350 | Anderson *et al* (1987) |
| Virginia, USA | 6 | 25 | 15 | 0 | 100 | 1.9 | 360 | Anderson *et al* (1987) |
| Virginia, USA | 6 | 25 | 15 | 0 | 100 | 4.3 | 360 | Anderson *et al* (1987) |
| NC, USA | 6.2 | 28.33 | 14.9 | 0 | 68 | 0.5 | 365 | Aneja_et_al._(1996) |
| NC, USA | 6 | 35.75 | 13.7 | 0 | 168 | 2.2 | 365 | Aneja_et_al._(1996) |
| Puerto Rico | 4 | 87 | 22.6 | 0 | 150 | 0.6 | 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 | 0.5 | 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 | 0.01 | 8 | Matson_et_al._(1996) |
| Maui | 4.7 | 49.5 | 19.7 | 0 | 84 | 0.01 | 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) |
| 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 | 0.01 | 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 | 1.6 | 183 | |
| Brazil | 5.94 | 39.5 | 32.75 | 0 | 42 | 2.1 | 183 | |
| NC, USA | 5.8 | 22.7 | 14.3 | 0 | 150 | 0.23 | 4 | Roelle_et_al._(2001) |
| NC, USA | 5.8 | 22.7 | 14.3 | 0 | 150 | 0.23 | 4 | Roelle_et_al._(2001) |
| NC, USA | 5.8 | 6.9 | 15.4 | 0 | 140 | 0.01 | 4 | Roelle_et_al._(2001) |
| NC, USA | 5.8 | 9.4 | 17.1 | 0 | 150 | 0.03 | 4 | Roelle_et_al._(2001) |
| NC, USA | 5.8 | 11.3 | 23 | 0 | 45 | 0.03 | 4 | Roelle_et_al._(2001) |
| NC, USA | 5.8 | 12.4 | 25.6 | 0 | 190 | 0.03 | 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 | 0.34 | 7 | Roelle_et_al._(2001) |
| NC, USA | 5.8 | 21.1 | 24.1 | 0 | 175 | 1.06 | 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) |

Table S: *(Continued)*

| *Location* | *pH* | *Soil moisture (%)* | *Temperature (°C)* | *N_type (0: fer 1:man)* | *N_rate (kg N/ ha)* | *NO emission (kg N/ha)* | *Length of Experiment (days)* | *Reference* |
|---|---|---|---|---|---|---|---|---|
| Guárice state | 3.7 | 2.7 | 30 | 0 | 600 | 1.3 | 19 | Sanhueza_et_al._(1994) |
| Venezuela | 4.6 | 20 | 27 | 0 | 200 | 0.01 | 5 | Sanhueza_et_al._(1990) |
| Venezuela | 4.6 | 20 | 27 | 0 | 200 | 0.3 | 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 | 0.05 | 13 | Aneja_et_al._(1995) |
| NC, USA | 6 | 5.5 | 35 | 0 | 173 | 0.1 | 13 | Aneja_et_al._(1995) |
| Tennessee, USA | 5.8 | 25 | 11.5 | 1 | 100 | 0.05 | 20 | |
| Tennessee, USA | 5.5 | 25 | 27.3 | 0 | 111 | 0.33 | 9 | Valente_and_Thornton_ (1993) |
| Tennessee, USA | 5.7 | 25 | 26.2 | 0 | 111 | 0.31 | 15 | Valente_and_Thornton_ (1993) |
| 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 | 4.6 | 365 | Veldkamp_et_al._(1998) |
| Costa Rica | 5.1 | 72.9 | 25.8 | 0 | 300 | 5.4 | 365 | Veldkamp_et_al._(1998) |
| Costa Rica | 5.1 | 72.9 | 25.8 | 0 | 300 | 7.0 | 365 | Veldkamp_et_al._(1998) |
| Costa Rica | 5.1 | 72.9 | 25.8 | 0 | 300 | 7.5 | 365 | Veldkamp_et_al._(1998) |
| Costa Rica | 5.1 | 72.9 | 25.8 | 0 | 300 | 8.4 | 365 | Veldkamp_et_al._(1998) |
| NC, USA | 6 | 5.6 | 27.3 | 0 | 70 | 0.03 | 7 | Aneja_et_al._(1998) |
| NC, USA | 6 | 12.6 | 24.8 | 0 | 190 | 0.03 | 6 | Aneja_et_al._(1998) |
| NC, USA | 6 | 11.3 | 22.6 | 0 | 197 | 0.07 | 9 | Aneja_et_al._(1998) |
| Colorado, USA | 5.9 | 4.8 | 27.2 | 1 | 100 | 0.15 | 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 | 0.49 | 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) |
| Spain | 8.1 | 70 | 20 | 0 | 0 | 0.01 | 150 | Meijide *et al.* (2007) |
| Spain | 8.1 | 70 | 20 | 1 | 175 | 0.13 | 150 | Meijide *et al.* (2007) |
| Spain | 8.1 | 70 | 20 | 1 | 175 | 0.13 | 150 | Meijide *et al.* (2007) |
| Spain | 8.1 | 70 | 20 | 1 | 175 | 0.04 | 150 | Meijide *et al.* (2007) |
| Spain | 8.1 | 70 | 20 | 1 | 175 | 0.1 | 150 | Meijide *et al.* (2007) |
| Spain | 8.1 | 70 | 20 | 1 | 175 | 0.03 | 150 | Meijide *et al.* (2007) |
| Spain | 8.1 | 70 | 20 | 0 | 175 | 0.24 | 150 | Meijide *et al.* (2007) |
| Spain | 6.6 | 60 | 17.2 | 0 | 0 | 0.26 | 59 | Menendez *et al.* 2006 |
| Spain | 6.6 | 60 | 17.2 | 0 | 97 | 2.76 | 59 | Menendez *et al.* 2006 |
| Spain | 6.6 | 60 | 17.2 | 0 | 97 | 1.54 | 59 | Menendez *et al.* 2006 |
| Spain | 6.6 | 60 | 17.2 | 1 | 97 | 0.7 | 59 | Menendez *et al.* 2006 |
| Spain | 6.6 | 60 | 17.2 | 1 | 97 | 0.52 | 59 | Menendez *et al.* 2006 |
| Spain | 8.1 | 72 | 22.5 | 0 | 0 | 0.03 | 180 | Vallejo *et al.* 2005 |
| Spain | 8.1 | 72 | 22.5 | 1 | 200 | 0.06 | 180 | Vallejo *et al.* 2005 |

Table S: *(Continued)*

| *Location* | *pH* | *Soil moisture (%)* | *Temperature (°C)* | *N_type (0: fer 1:man)* | *N_rate (kg N/ ha)* | *NO emission (kg N/ha)* | *Length of Experiment (days)* | *Reference* |
|---|---|---|---|---|---|---|---|---|
| Spain | 8.1 | 72 | 22.5 | 1 | 200 | 0.05 | 180 | Vallejo *et al.* 2005 |
| Spain | 8.1 | 72 | 22.5 | 1 | 200 | 0.03 | 180 | Vallejo *et al.* 2005 |
| Spain | 8.1 | 60 | 21 | 0 | 0 | 0.01 | 180 | Vallejo *et al.* 2006 |
| Spain | 8.1 | 60 | 21 | 1 | 175 | 0.29 | 180 | Vallejo *et al.* 2006 |
| Spain | 8.1 | 60 | 21 | 1 | 175 | 0.22 | 180 | Vallejo *et al.* 2006 |
| Spain | 8.1 | 60 | 21 | 1 | 175 | 0.28 | 180 | Vallejo *et al.* 2006 |
| Spain | 8.1 | 60 | 21 | 1 | 175 | 0.22 | 180 | Vallejo *et al.* 2006 |
| Spain | 8.1 | 60 | 21 | 0 | 175 | 0.33 | 180 | Vallejo *et al.* 2006 |
| Spain | 8.1 | 60 | 21 | 0 | 175 | 0.24 | 180 | Vallejo *et al.* 2006 |
| Japan | 5.6 | 55 | 20 | 0 | 250 | 0.11 | 77 | Cheng *et al.* 2002 |
| Japan | 5.6 | 55 | 20 | 0 | 250 | 0.50 | 77 | Cheng *et al.* 2002 |
| Japan | 5.6 | 43.5 | 26.6 | 0 | 250 | 0.11 | 180 | Hou & Tsuruta 2003 |
| Japan | 5.6 | 43.5 | 26.6 | 0 | 250 | 0.09 | 180 | Hou & Tsuruta 2003 |
| Japan | 6.1 | 50 | 20 | 0 | 150 | 0.03 | 70 | Yan *et al.* 2001 |
| Japan | 6.1 | 50 | 20 | 0 | 150 | 0.4 | 70 | Yan *et al.* 2001 |