Agriculture - Estimation of Direct Nitrous Oxide (N_2O) from Synthetic Fertilizers at Country-level

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1. Introduction

Nitrous Oxide (N_2O) is a highly potent greenhouse gas (GHG), with a global warming potential (GWP) of 298 that of CO_2 on a 100-year timescale (EPA, 2018). The concentration of atmospheric N_2O has increased by more than 20%, from 270 parts per billion (ppb) in 1750 to 331 ppb in 2018 (Tian et al., 2020). This dramatic shift is primarily driven by increased anthropogenic sources that raise current total global emissions to ~17 teragram (Tg) nitrogen (N) (Syakila and Kroeze et al., 2011; Thompson et al., 2019; Tian et al., 2020). Approximately 52% of anthropogenic emissions come from the direct emissions from Nitrogen (N) addition in the agricultural sector (Tian et al., 2020). Direct N_2O emissions from soils are primarily produced during two microbial-driven biological processes, nitrification and denitrification. Nitrification is the aerobic microbial oxidation, in which ammonium ion is oxidized into nitrate and N_2O is released as a byproduct.

On the other hand, denitrification is the anaerobic microbial (mainly bacterial) reduction of nitrate to nitrite and then to the gasses NO, N_2O, and N_2. N_2O production depends on the amount of mineral N substrates in the soil, i.e., ammonium and nitrate. Therefore, additions of mineral N fertilizers and other sources of N (manures, residue) to agricultural soil are considered the primary drivers of N_2O emissions and higher atmospheric concentrations. Lassaletta et al. (2014) estimated that only 47% of the reactive nitrogen added globally onto cropland is converted into harvested products, and the rest is lost into the environment. For sustainable agriculture, it is essential to quantify these emissions with more confidence and find ways to mitigate climate change by reducing them. Northup et al. (2021) have shown in their recently published work that there is a potential to reduce 71% (1,744 kg CO_2e/ha) of greenhouse gas emissions through a combination of innovations in digital agriculture, crop and microbial genetics, and electrification (using electrical farm equipment instead of fuel-based) in the next 15 years. According to this study, process-based models guided via high-resolution monitoring systems can be used to optimize the N fertilizer use for the crop, which can further reduce the N_2O emissions. Similarly, customizing crop genetics and root traits can help improve the plant's nitrogen use efficiency.

The addition of N-fixing microbes (in addition to those that form root nodules) in the soil can reduce the need for chemical fertilizer, eventually leading to reduced emissions.

Various efforts have been made in the past to quantify N_2O emissions at the regional and global levels from the agricultural sector. The FAOSTAT emissions database of the Food and Agriculture Organization of the United Nations (FAO) covers emissions of N_2O from agriculture by country and globally from 1961 to 2018 (Tubiello et al., 2021). The United Nations Framework Convention on Climate Change (UNFCCC) provides N_2O emission data from agricultural managed soils reported by countries for a period 1990-2019. Another inventory, the Emission Database for Global Atmospheric Research version 4 (EDGAR v4.3.2), a product of the Joint Research Center and the PBL Netherlands Assessment Agency, contains global N_2O emission inventories(Janssens-Maenhout et al., 2019). EDGAR applies the Intergovernmental Panel on Climate Change (IPCC) guidelines mostly at Tier-1 (using emission factors and activity data). Still, EDGAR integrates higher tier information based on available country reporting, mostly from Annex I countries. EDGAR provides data from 1970 to 2012. The Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model (Winiwarter et al., 2018) is an extension of the Regional Air Pollution Information and Simulation (RAINS) model. This extended version (GAINS) estimates emissions of 10 air pollutants and 6 GHGs (including N_2O) for each country based on data from international energy and industrial statistics, emission inventories, and data supplied by countries. GAINS provides N_2O emissions data every five years (i.e., 1990, 1995, 2000, 2005, 2010, 2015) and uses the IPCC emission factor as a default option for estimation in the absence of more detailed information available for a country.

While there are databases that provide emissions information, each inventory reports N_2O emissions differently. For example, FAOSTAT and UNFCCC provide direct N_2O emissions for the category of synthetic N fertilizers applied to managed soils. In comparison, GAINS and EDGAR report the combined direct N_2O soil emissions from synthetic N fertilizers and manure as fertilizers and crop residues. This difference in emission reporting makes it difficult to compare the estimates of direct N_2O emissions (from synthetic N fertilizers) across different inventories. Additionally, all inventories described incorporate the IPCC Tier 1 guideline for calculating direct N_2O soil emissions, which mainly relies on the self-reported N fertilizer data. Reported data is prone to errors and might be purposely misreported leading to significant uncertainties in estimating N_2O direct emissions in the current approach (Seto et al., 2000). To reduce these uncertainties and provide greater temporal and country coverage, a new approach is needed that considers measurements that are more reflective of synthetic fertilizer usage.

This research aims to develop an innovative method that can estimate country-level N_2O emissions independently from the highly uncertain self-reported N fertilizer data. Our technique relies on crop productivity data, which is a more reliable source of information as it indicates raw material going for direct consumption or into the food systems and supply chain. The method developed in this work predicts yearly direct N_2O emissions due to N fertilizer used at the country-level by accumulating crop-specific emissions (considering the primary crops within a country). Crop yield, area, and nitrogen use efficiency (NUE) within the country are used to calculate crop-specific emissions. NUE for a prediction year in a country is estimated using NUE from the previous year (calibrated with IFASTAT data) and percent change in productivity for a crop within the two consecutive years.

2. Materials and Method

The approach utilized here primarily relies on crop productivity data to estimate N_2O emissions, which is considered a reliable source of information as crop productivity indicates raw materials going for direct consumption or into the food systems and supply chain. We hypothesized that crop productivity could be used as a proxy for estimating N_2O emissions for a country. To investigate this, we selected a subset of high-income, upper-middle-income, lower-middle-income, and low-income countries and classified them by their development level as measured by per capita gross national income (GNI) by the United Nations(UNESCAP, 2020).

To understand the relationship between the average crop productivity and direct N_2O emissions within these selected countries, two variables were used. First, we estimated average crop productivity within the country by accumulating total annual production and harvested area for all the primary crops as defined in FAOSTAT data from 1961-2018 and extracted country-level annual direct N_2O emission data from FAOSTAT for the same period. Then, we compared crop productivity (tons/ha) to direct N_2O emission for each selected country (Figure 1).

Table 1 Pearson correlation coefficients and their significance level between average crop productivity and direct N_2O emission for each country. Correlation coefficients with an asterisk are statistically significant ($p<0.01$).

GNI level	Country	Correlation coefficient (ϱ)
High-income	Australia	0.69*
	France	0.80*
	Germany	0.18
	U.S.A	0.82*
Upper-middle-income	Argentina	0.92*
	China	0.97*
	Brazil	0.95*

Lower-middle-income	India	0.99*
	Nigeria	0.73*
	Pakistan	0.96*
Low-income	Uganda	-0.09
	Tanzania	0.17

Our analysis found that the average crop productivity and direct N_2O emissions from synthetic fertilizers data were highly correlated for countries in the upper-middle-income and lower-middle-income countries (Table 1). For example, India, China, Argentina, Brazil, and Pakistan have correlation coefficients greater than 0.9. In the high-income countries group, the U.S.A, one of the significant synthetic nitrogen fertilizer users (after China and India), shows a correlation of 0.82 (Figure 1). On the other hand, this relationship was not significant in low-income countries like Uganda and Tanzania. The reason behind this is the minimal use of nitrogen fertilizer in cropland; less than 1% of farmers in most African countries apply fertilizers (Nkonya et al., 2011). One exception was Germany, which belongs to the high-income group and shows no significant relationship between productivity and direct N_2O emissions for reasons unknown to us. In our initial analysis, the major synthetic nitrogen fertilizer used countries, like China, India, U.S.A, Brazil, Pakistan, France, and Australia, which represent 66% of total global nitrogen fertilizer use (FAOSTAT, 2019) shows a strong correlation between crop productivity and direct N_2O emissions from synthetic fertilizers. Overall, this analysis supports our hypothesis that crop productivity has a strong correlation with direct N_2O emissions from synthetic fertilizers and it can be used as a proxy to estimate direct N_2O emissions.

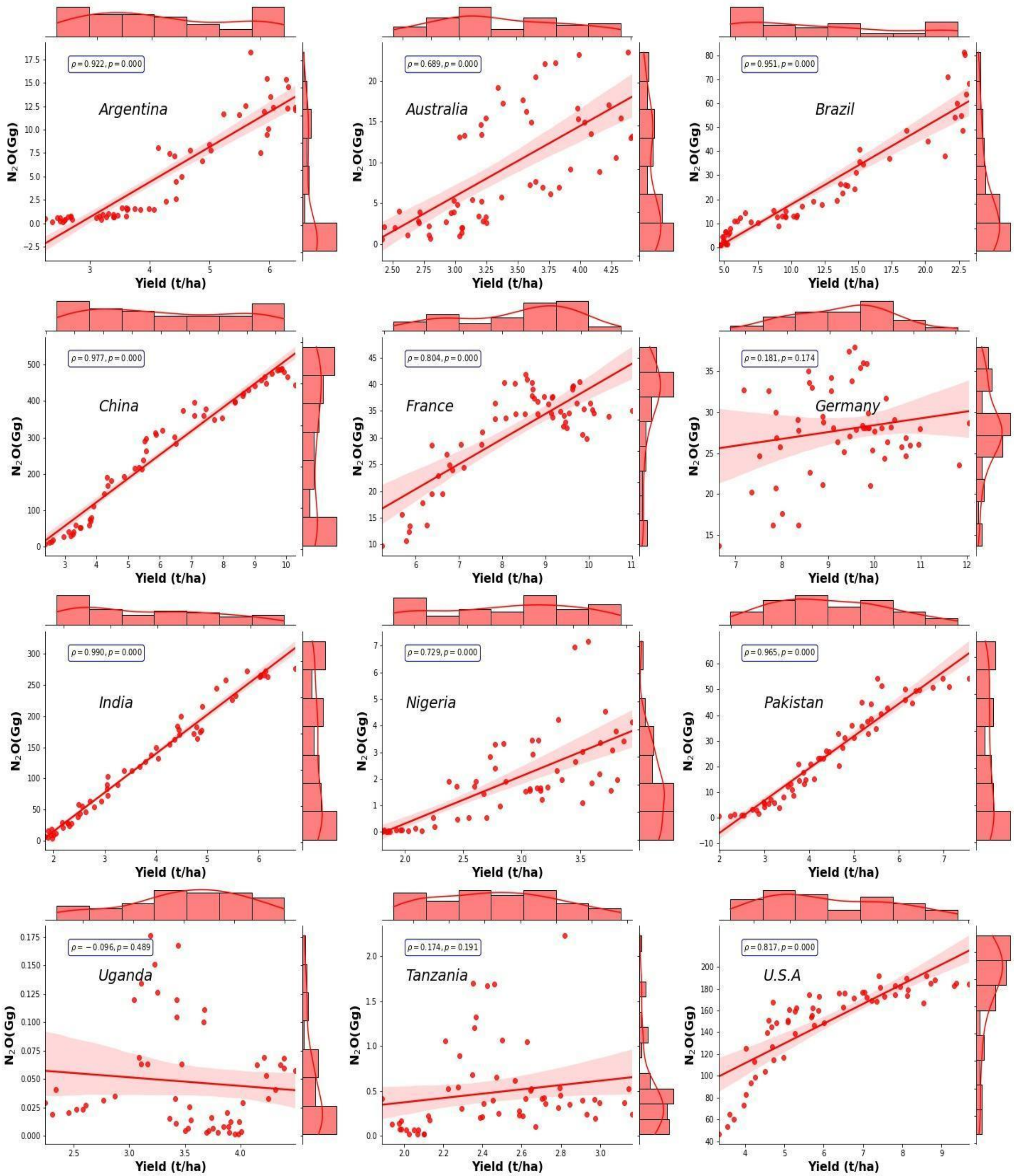


Figure 1 Correlation graph between country-level direct N_2O emissions due to synthetic N fertilizer and crop yield using FAOSTAT data from 1961-2018. (Units- y-axis: gigagram, x-axis: tonnes/ha).

2.1 Data

The following inputs were used to estimate country-level direct N2O emissions in Table 2. These data sets were used in three parts, model development, calibration and validation.

Table 2 Model development, validation data used for estimation of N_2O emissions.

Model development	***Unit***	***Source***
Crop-specific Yield	Mg/ha	FAOSTAT
Crop-specific Harvest area	ha	FAOSTAT
Crop-specific N content	Kg N/ton	Lassaletta *et al.,* 2014
Model calibration	***Unit***	***Source***
Total N fertilizer at country scale	Kton	IFASTAT
Validation	***Unit***	***Source***
Direct N_2O emission at country scale	Gg	FAOSTAT
Direct N_2O emission at country scale	Kton	UNFCCC
Output	***Unit***	***Source***
Direct N_2O emission at country scale	tCO_2eq /Gg	Model generated

2.1.1 FAOSTAT & UNFCCC

FAOSTAT is a data center for FAO (Agency of United Nations), which provides free access to food and agriculture statistics (including crop, livestock, and forestry sub-sectors) for over 245 countries and covers time periods from 1961 to the most recent year available. At the time of this writing, the most recent year for reported synthetic fertilizer emissions per country is 2019. FAOSTAT compiles this dataset using annual questionnaires sent to country focal points within the national statistical systems. The following datasets were used for this study:

a) Crop-specific yield and harvest area data were extracted for primary crops for 94 countries. Primary crops include > 150 types; however, 12 nitrogen-fixing (i.e., soybean) crops were excluded from this study as they do not require synthetic N fertilizer application. Instead, these nitrogen-fixing crops obtain N from the atmosphere via the nitrogen fixation process. This information was used in model development (Table 2)

b) To validate our country-scale emissions, direct N_2O emissions data were extracted from FAOSTAT & UNFCCC for years 2015 to 2019. FAOSTAT calculates direct N_2O emission using

Tier 1 approaches, based on generalized emission factors and other parameter values that are specified either globally or regionally. The Tier 1 method employs the default (simplest) method described in the IPCC Guidelines and the default emission factors and other parameters provided by the IPCC. UNFCCC direct N_2O emission data (available via FAOSTAT) was reported by the countries using Tier 2 and 3 methods suggested by IPCC. Both tier estimates were used to validate the modeled approach (Table 2).

2.1.2 IFASTAT

IFASTAT is a data center for the International Fertilizer Association (IFA; https://www.ifastat.org/). This is the only global fertilizer association and has a membership of some 400 entities, encompassing companies across the fertilizer value chain from producers through traders and distributors and service providers to advisors, research organizations and NGOs. Country scale N fertilizer data was extracted from IFASTAT, which is based on a survey conducted every year, sent to country correspondents, including fertilizer associations, fertilizer companies, consultants, experts, and university researchers.

2.2 Method

This novel method is based on an integrated approach using country-level crop harvest area, yields, and crop-specific NUE, to predict the amount of synthetic N fertilizer applied to the primary crops in a country for a particular year. Country-level estimates of N_2O emissions are based on the IPCC (2006) emission factors for synthetic N fertilizer use (Figure 2). Using this approach, we estimated the emissions for 94 countries which represent approximately 98% of total synthetic N fertilizer use globally based on 2019 FAOSTAT data.

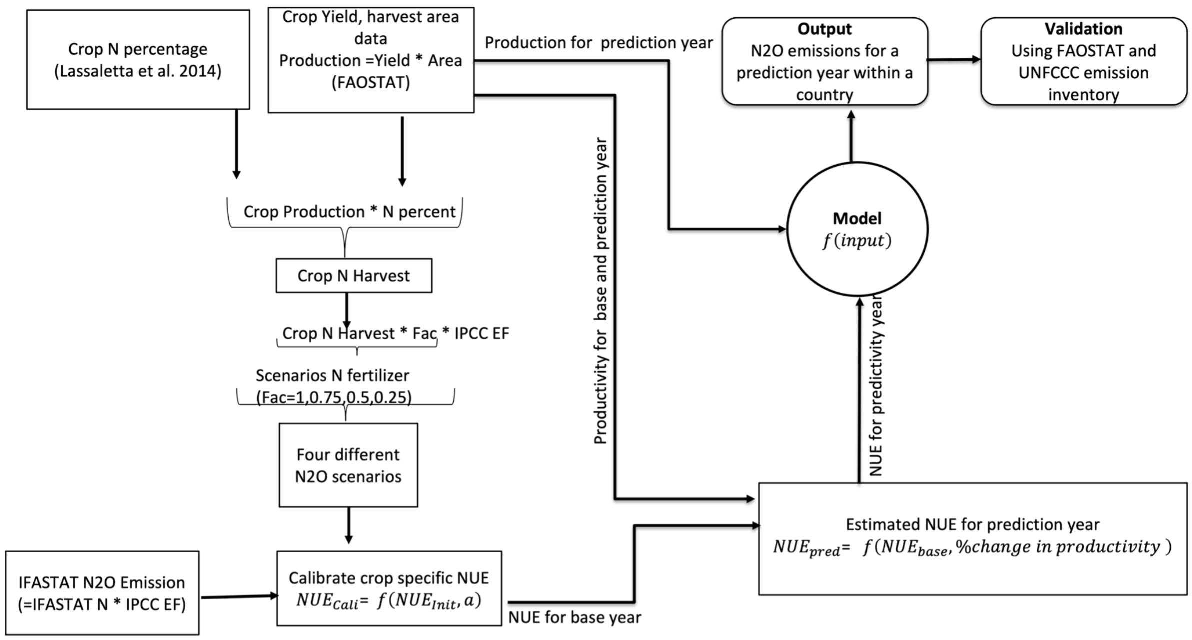


Figure 2 Schematic representation of estimating N_2O emissions from crop productivity at country scale.

2.2.1 Scenario development for N fertilizer amount

The first step in this method was to estimate N fertilizer applied in a country. To do this, four scenarios were created to estimate N fertilizer applied as the fraction of N uptake into crops. To develop each scenario, first, we used FAOSTAT crop-specific yield and harvest area data, and nitrogen content ($N_{CONTENT}$; Eq. 1; Table 2) to calculate the amount of N removed from the field during the harvest process, denoted as $N_{HARVEST}$. For this calculation, it was assumed that $N_{HARVEST}$ is the actual N uptake from the soil ($\sim N_{UPTAKE}$), and avoided N left in as the crop residue (e.g., roots) part. The reason for not accounting the residues N content is that they are assumed to be returned to the soil, thus cycled in, a common procedure in N mass balance (Basso et al., 2019). The total N uptake in the crops during the growing season can be from many N sources like Synthetic N fertilizers, Manure N, N deposit, and N mineralization from the soil. However, Synthetic N fertilizers are recognized as the most important factor contributing to direct N_2O emissions from agricultural soils. Based on our assumption that the $N_{HARVEST}$ is proportional to the amount of synthetic N fertilizers applied, four scenarios were designed for N fertilizer (N_{FERT}) as shown in Figure 3.

$$N_{FERT} = \underbrace{\text{Yield} \times \text{Area} \times N_{CONTENT}}_{N_{HARVEST}\ (\sim N_{UPTAKE})} \times \text{Fac (Eq. 1)}$$

Where, "Fac" is the proportionality factor for each scenario (1, 0.75, 0.5, 0.25). Then each scenario is converted into emissions using the emission factor (EF) from IPCC (2006) in Eq. 2:

$$N_2O = N_{FERT} \times EF \ \text{(Eq. 2)}$$

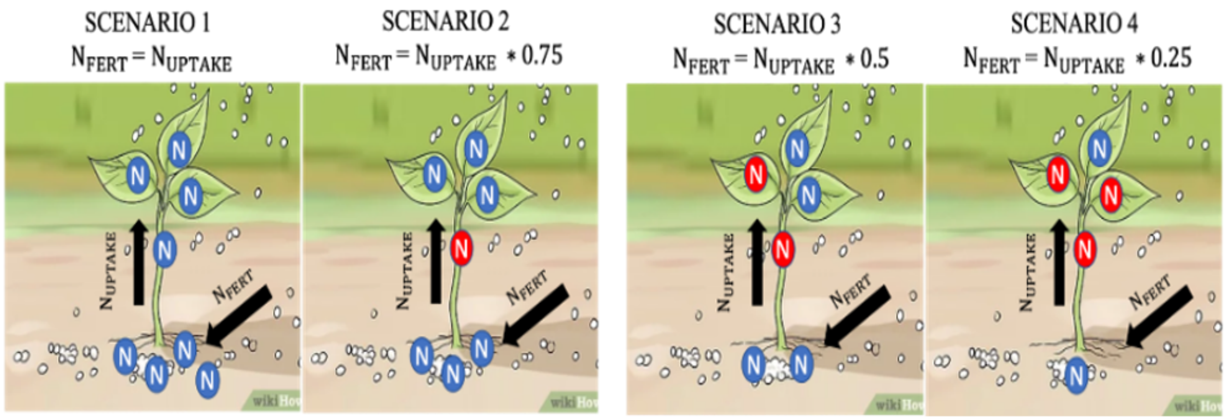


Figure 3 Overview of N fertilizer amount as a proportion of N uptake for four scenarios. For Scenarios 1, 2, 3, 4, the N fertilizer amount is 100%, 75%, 50%, and 25% of N uptake by the crops, respectively.

Scenario 1 represents the ideal case where all the N taken up by the plants is supplied from the synthetic N fertilizer applied and both are equal in amount. In scenario 2, 3, and 4 we considered 75%, 50%, and 25%, of the N taken up by the plants is supplied from the synthetic N fertilizer applied, respectively. For example, suppose a case where actual emissions (calculated based on IFASTAT fertilizer data) in three countries are higher than, equal to, and less than scenario 1. Then, in the first country, where emissions are greater than scenario 1, it suggests that the N fertilizer amount applied is more than the required amount needed by crops for their growth. But, on the other hand, the second country, where emissions are equal (or closer) to scenario 1 (ideal case), represents the adequate application of fertilizers, matching the amount needed for crop growth. For the third country, where the emissions are less than scenario 1, this suggests a lower fertilizer application rate than the crops require. We further divided the last case into three more scenarios 2, 3, and 4, which represent the case where the amount of fertilizer applied is 25% (75% of N uptake is from synthetic fertilizer), 50% (50% of N uptake is from synthetic fertilizer), and 75% (25% of N uptake is from synthetic fertilizer), less than the required amount, respectively.

Our aim in creating these "Scenarios" is to understand how the actual emissions data compares to our assumed scenarios, and it might or might not represent the real-time conditions. These scenarios were compared to real-time emissions to see how well each scenario relates, which provides insight into the N fertilizer application amount to the crops within a country. This can help determine if the amount applied is higher, equal, or lower from the required amount needed for the crops.

2.2.2 Estimation of crop-specific NUE at a country scale

This step provides insight into how efficiently the primary crops take up the amount of N fertilizer applied within the country and its temporal variability. Country-scale nitrogen use efficiency (NUE) was calculated using N fertilizer amount in scenario 1 which is equal to N uptake for the crops (Fac =1; Eq 1) and observed N fertilizer from IFASTAT (Eq. 3):

$$\mathrm{NUE} = \frac{N_{FERT}\,(Scenario\ 1)}{N_{FERT}(IFASTAT)} \qquad \text{(Eq. 3)}$$

Our calculated NUE is more in context to Synthetic N fertilizers, ignoring other N inputs (i.e., N manure, deposits). However, as our target was to estimate direct N_2O from Synthetic N fertilizers, it is relevant to our final calculations.

Based on the understanding acquired about the country-specific NUE, we scaled up or down the crop-specific N fertilizer amount from scenario 1 to a new value. Also, we make sure that the scaling factor for the individual crop is weighed by its contribution to total N_2O emissions in that scenario. Using this approach, we calculated the NUE for each crop within a country.

Our aim here was to develop an independent estimate of N fertilizer amount for the estimation of N_2O emission. Therefore, the predicted NUE for the prediction year and temporal variability can be explained by the changes in crop yield from base year to prediction year within a country.

2.2.3 Prediction of N_2O emissions

In the final step, we estimated annual emissions for a country. First, we calculated the emission for each crop using yield, harvest area, and NUE (Eq. 4). Then, accumulated crop-specific N_2O for all the primary crops within the country to determine the total emission (Eq. 5). To estimate direct N_2O emissions for a country, the following data as an input for each crop was needed:

1. Yield, harvest area, Nitrogen content for target year (Table 2)
2. Estimated crop-specific NUE for the target year (estimated in section 2.2.2)

The inputs from 1 and 2 above were combined in Eq.4 to estimate N_2O emissions:

$$N_2O_{i,j} = \left(\frac{Yield_{i,j}\ x\ Area_{i,j}\ x\ Ncontent_i}{NUE_i}\right)\ \text{X EF (Eq. 4)}$$

Where EF converts N to N_2O. The final, total, calculation for a country,

$$TotO_j = \sum_{i=1}^{n} N_2O_{ij}\ \text{(Eq. 5)}$$

Where i is the type of crop and j is the target year for the country. N_2O_{ij} is the direct emission predicted for the particular crop (i) in a target year (j), and n is the total number of primary crops defined in FAOSTAT data within a country. $TotN_2O_j$ is the total direct emission for a country in a target year (j).

2.2.4 Downscaling from country scale to grid-level

Once country-level emissions estimates were generated, we downscaled the country-level N fertilizer amount to grid level at a spatial resolution of 0.083° (~10 km) in latitude by longitude, using country scale crop-specific N fertilizer rate (calculated in earlier steps) and gridded harvest area produced by Global Agro-Ecological Zones (GAEZ) Version 3 model, which is based on the FAO crop production data. GAEZ crop classification is grouped into 26 crop-types which includes all the 160 unique crops from FAOSTAT (Table S3). This was done to understand the spatial distribution of N_2O emissions within a country and to identify areas with significant N_2O emissions (fertilizer application).

Our study has excluded the nitrogen fixing crops (discussed in section 2.1.1) from FAOSTAT data. Therefore, to harmonize our crop type to GAEZ classification we distributed our crop type into 23 crop groups from GAEZ data, excluding soybeans and pulses. Then, we aggregated country scale crop-specific N fertilizer data (estimated in our study) and harvest area (from FAOSTAT) for each group of crops. Next, using these two data sets we calculated the country scale N fertilizer rate for each crop group. Further, GAEZ data is only available for circa 2015, to obtain gridded harvest area for year 2015 to 2020, crop-specific FAOSTAT country scale harvest areas were used to upscale or downscale the gridded values. Then, the crop-specific gridded harvest area was multiplied with N fertilizer rate (country-specific) to estimate crop-specific N fertilizer amount at 0.083° (~10 km spatial resolution) and aggregated for all the crops within the grid to derive the total N fertilizers applied (Figure S2). Finally, to calculate direct N_2O emission from Synthetic N fertilizers, the aggregated N fertilizer amount within the grid was multiplied with the IPCC emission factor.

An example of 2020 grid-level data is displayed in Figure S2. For other years, the grid-level data is hosted on the Climate TRACE website, https://climatetrace.org/.

2.3 Verifying modeled emissions estimates

To verify the modeled country-level N_2O emission estimates, FAOSTAT and UNFCCC direct N_2O emissions data for years 2015 to 2019 were used. A time-series analysis was performed for 24 countries, representing the significant portion of N_2O emissions globally, and a direct modeled emissions to FAOSTAT or UNFCCC comparison was performed. The spatially gridded N_2O emissions were not compared to other emissions estimates (i.e., *in-situ* measurements) due to a lack of globally distributed data at the time of this work.

3. Results & Discussion

We compared N_2O derived from the N_{FERT} amounts in different scenarios to the IFASTAT observation data at the country-level for 2014-2019 (Figure 4a). IFASTAT only provides N fertilizer value at the country scale and to convert it to N_2O emissions the IPCC (2006) emission factor is applied. Figure 4b, shows the country scale NUE (calculated in section 2.1.2). For Asian countries, like India, China, Pakistan, and Indonesia, the direct emissions (or N fertilizer) observed is much higher than scenario 1, suggesting primary crops take up a very low percentage of N fertilizer applied (low Nitrogen Use Efficiency: NUE<<1). On the other hand, direct emissions for African countries like Tanzania, Kenya, and Nigeria, are closer to scenario 4 for most years, suggesting a small fraction of N uptake comes from synthetic N fertilizer (lower N application) in these countries (N mining: NUE >1). Soil N mining or fertility lost results from over exploitation of agricultural land without giving back the lost N to the soil. Due to limited access of synthetic N fertilizer and increasing population pressure led to severe loss of soil nutrient fertility in these African countries. This problem of nutrient mining and loss of soil fertility in African countries is highlighted in previous studies (Vitousek et al., 2009, Liu et al., 2010).

In countries like the U.S.A, France, Canada, Germany, Australia, Italy, and South Africa, observed emissions are closer to scenario 1, suggesting these countries have better nutrient efficiency (Moderate Nitrogen Use Efficiency: NUE < 1) as compared to Asian countries. One of the reasons might be due to more technological advancement and better management practices in these countries. According to our analysis, in the last two decades, NUE for most countries have not varied significantly, except for African countries like Nigeria and Tanzania, where synthetic fertilizer is not the primary source of crop N uptake, and extra N comes from the agricultural soil nutrient storage. Lassaletta et al. (2014) showed the NUE trends for the past 50 years for 124 countries and found higher crop yield than fertilization (NUE >1; N mining) for 18 countries such as Canada, Morocco, Algeria, Iraq, and Mozambique in the 1960–1980 period (Lassaletta et al., 2014). Also, in recent years of his study period, NUE higher than 1 have been observed in 10 African countries and former Soviet Union countries, Afghanistan and Paraguay. Argentina is the only country with NUE higher than 1 for the whole period of his study.

Although our calculated NUE is more in context to Synthetic N fertilizers, ignoring other N inputs (i.e., N manure, deposit), our study and results shown in Figure 4b suggests NUE is greater than 1 for countries like 1) Argentina, 2) Nigeria and Tanzania (African countries), 3) Russia and Ukraine (former Soviet Union countries), aligning well with analysis from Lassaletta et al. (2014).

a)

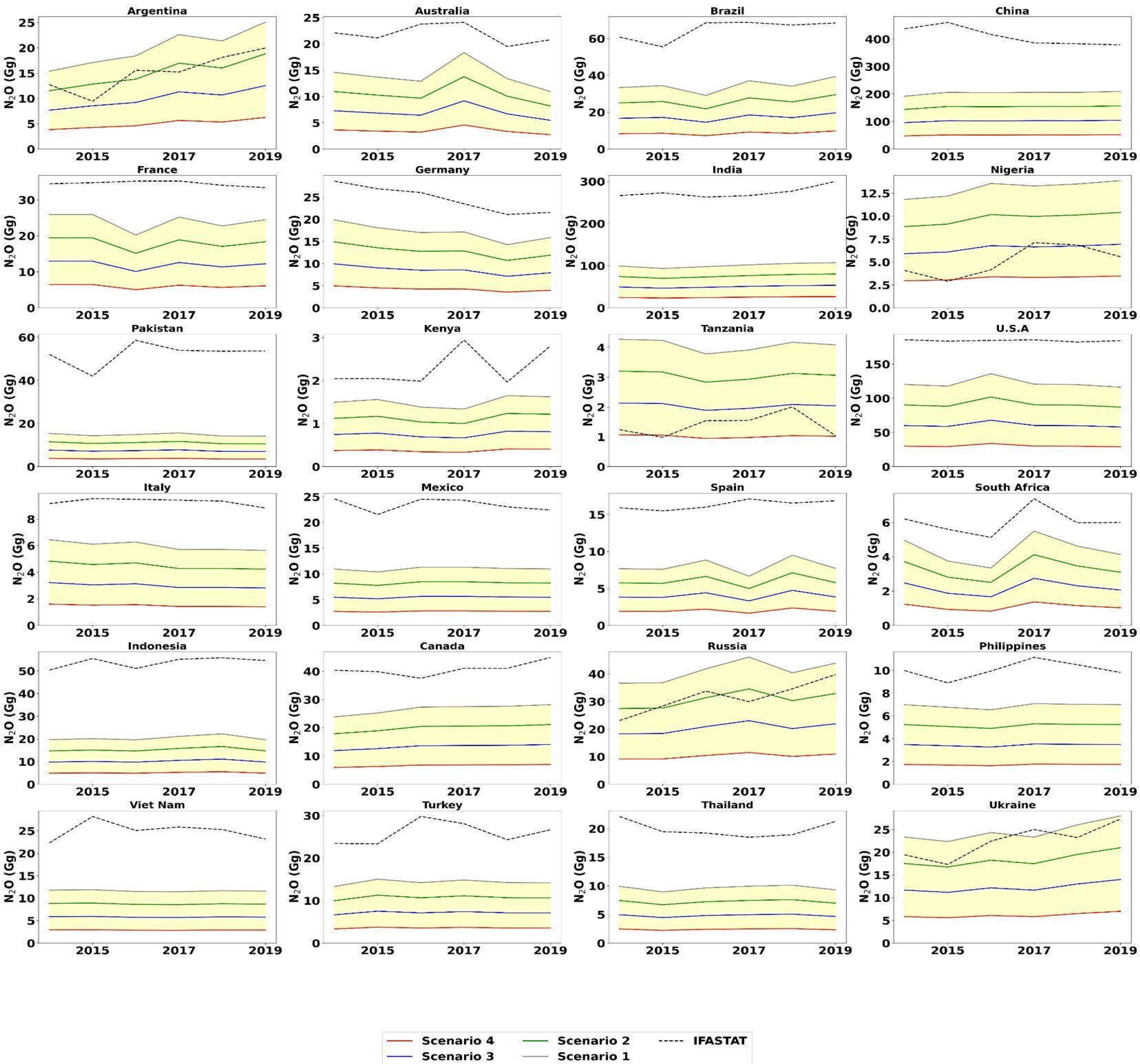


Figure 4 cont.

b)

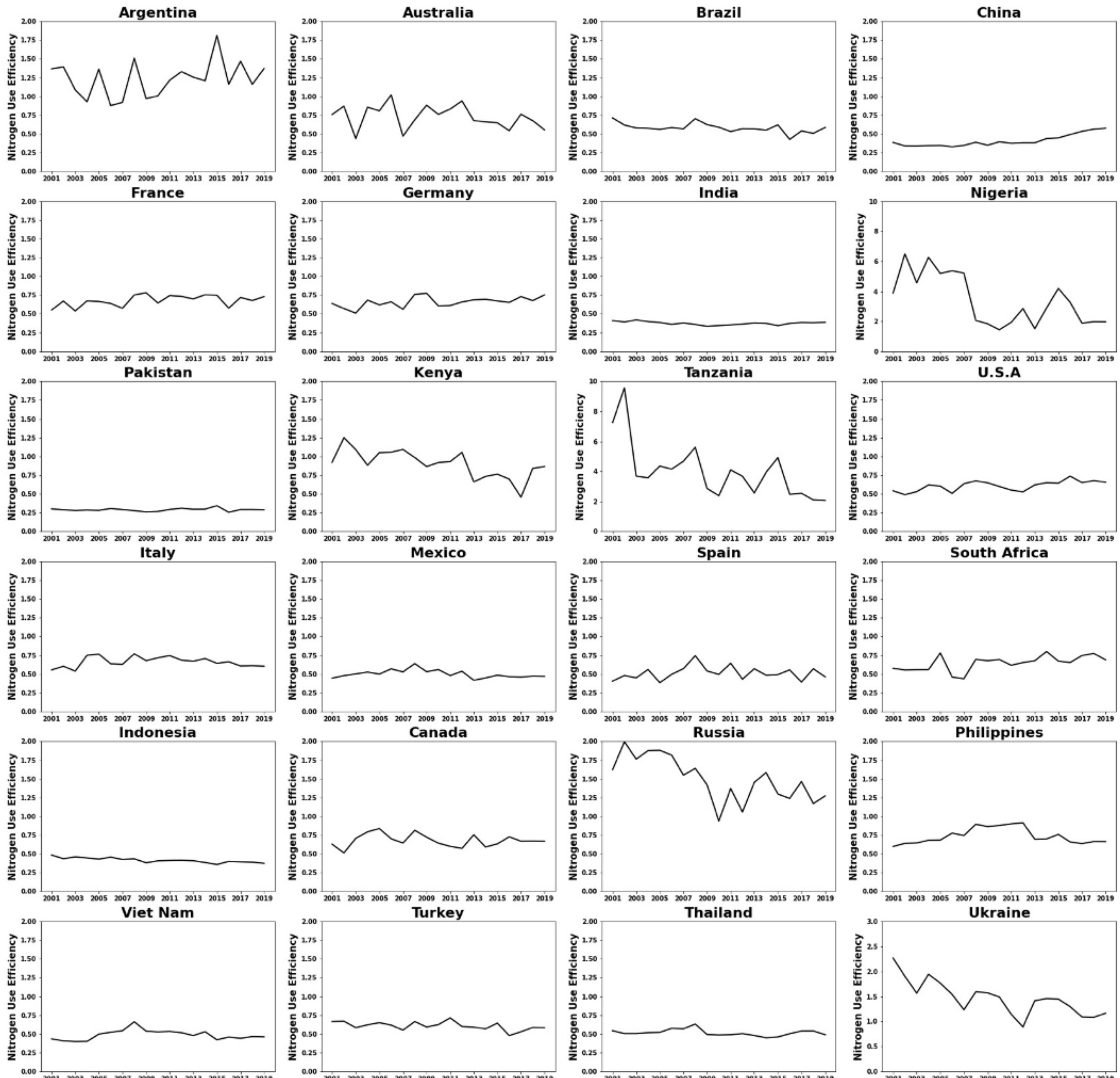


Figure 4 a) Time-series (2014-2019) of N_2O from IFASTAT and N_2O estimated from scenarios 1, 2, 3 and 4. Note, y-scales, N_2O (Gg) are different for each country; **b)** Nitrogen use efficiency (NUE) for each country based on scenario 1 NFERT and IFASTAT. Note, y-scales are different for each country.

In this study, we include 94 countries, which represent the majority of total synthetic nitrogen fertilizer use at global scale (~98%). According to our estimation, total direct N_2O emissions from synthetic nitrogen fertilizer use combined for 94 countries are 1623, 1610, 1636, 1585, 1600, and 1652 gigagrams (Gg) for years 2015, 2016, 2017, 2018, 2019, and 2020 respectively. China, India, and the U.S.A. are the top three emitter countries for the period 2015 to 2020 contributing more than 50% of total emissions every year (Table 3). Including the next set of highest emitters - Brazil, Pakistan, Indonesia, and Canada - the total percentage contribution reaches more than 60% of total emissions (>900 Gg) every year (Table 3).

Table 3 List of the 7 countries which contributed at least 69% to the total global direct N_2O emissions due to synthetic nitrogen fertilizers during the period of 2015-2020. Values are measured as percent (country total/global total *100).

Country	Percent contribution to total emission by year					
	2015 (%)	**2016 (%)**	**2017 (%)**	**2018 (%)**	**2019 (%)**	**2020 (%)**
China	29.1	28.2	25.3	24.0	23.6	22.8
India	16.3	16.7	16.3	16.6	17.3	19.0
U.S.A	11.2	11.9	10.6	11.6	11.3	11.2
Brazil	3.7	3.3	4.7	4.1	4.5	4.3
Pakistan	3.2	2.6	3.5	3.3	3.3	3.3
Indonesia	3.1	3.3	3.4	3.6	3.1	3.2
Canada	2.5	2.4	2.4	2.7	2.5	2.7

In addition to country-level estimation, the accumulated emissions per year at a continent level for Asia (AS), Europe (EU), America (AM), Africa (AF), and Oceania (OC) are provided in Table S1. As expected, AS is the highest emitting continent with a total emission of 949 Gg (58.7%) to the total emissions at a global scale (averaged over the study period), AM, EU, AF, and OC emit 360 Gg (22.3%), 233 Gg (14.5%), 51 Gg (3.2%), and 21 Gg (1.3%), respectively (Figure 5). Even though AF is quite significant in the land area, it only represents 3.2 % of total global emissions due to the limited access to synthetic use N fertilizers in African countries. AS and AM continents collectively represent more than 80% of the total global emissions.

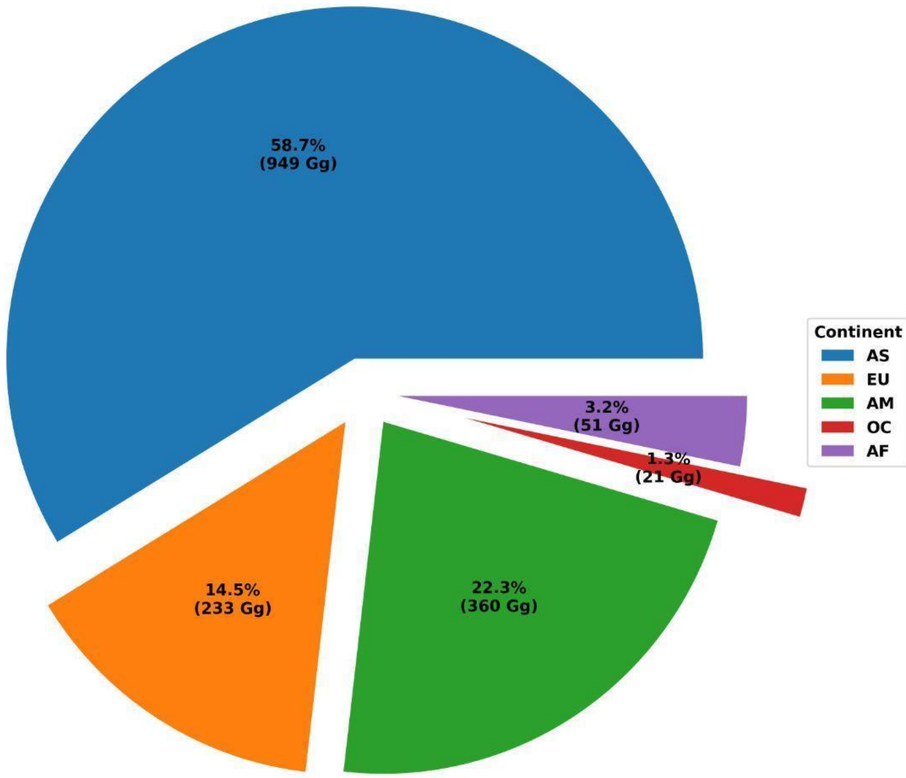
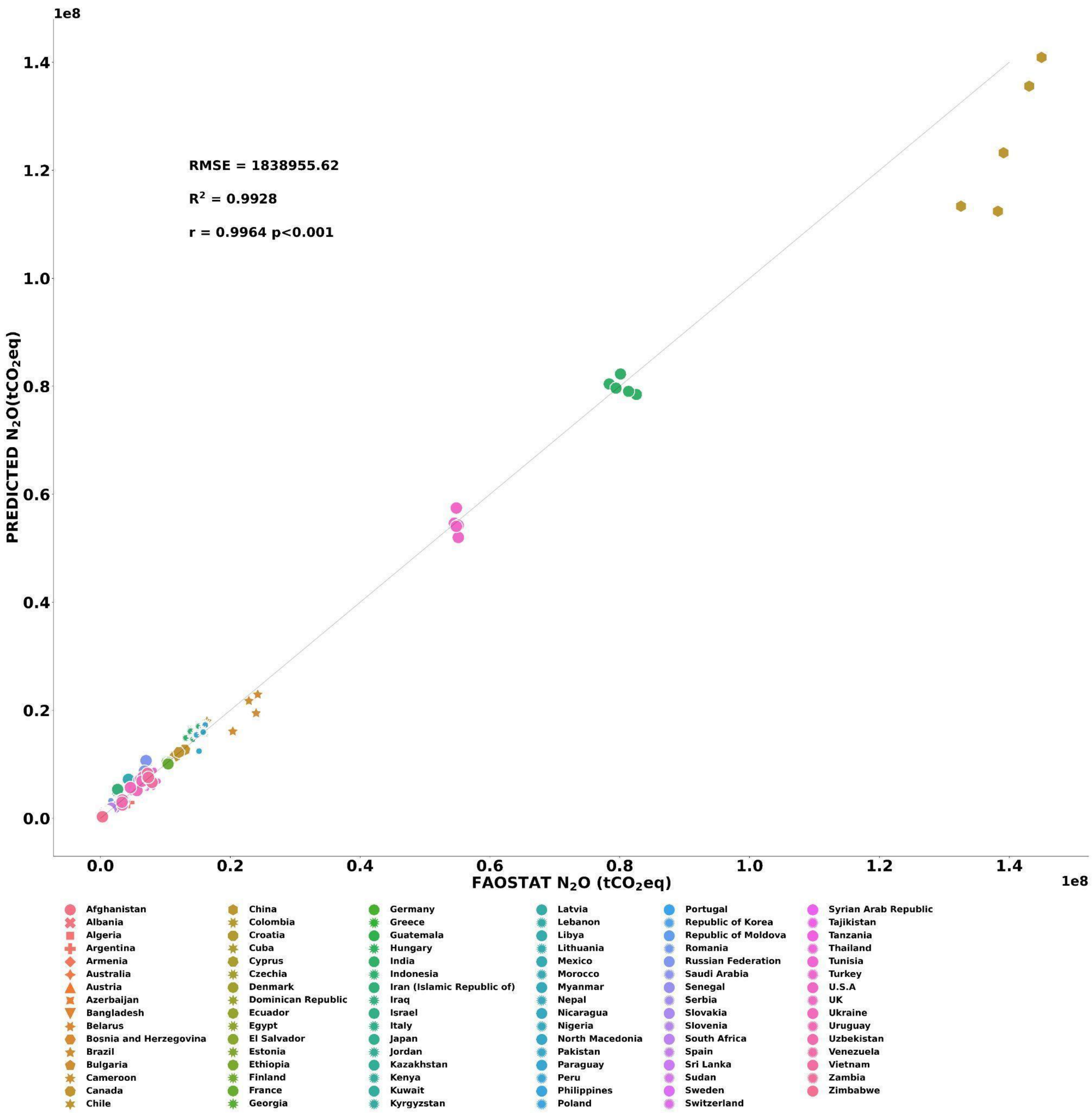


Figure 5 Average percentage contribution of each continent to global total direct N_2O emissions averaged over the period of 2015-2020. In parentheses are the average amount of emissions (Gg) for the same period. Legend key as follows: Asia (AS), Europe (EU), America (AM), Africa (AF), and Oceania (OC).

Our predicted N_2O emissions are independently compared against country-level estimates provided by FAO and UNFCCC inventories (Figure 6). FAO and UNFCCC provide direct N_2O emissions estimation from synthetic nitrogen fertilizer use at country scale. FAO estimates are computed at Tier 1 following the 2006 Guidelines for National GHG Inventories of the Intergovernmental Panel on Climate Change (IPCC, 2006). The primary data source for FAO is national data collected via the FAO Fertilizers questionnaire sent to FAO focal points in the national government. The UNFCCC provides direct N_2O emissions from managed agricultural soils reported by the countries. In this inventory, Annex I group countries' direct N_2O emissions from agricultural soil are calculated by combining IPCC's higher tier approaches (Tier 2 and 3). UNFCCC has limited countries which separately report direct emissions for the category of inorganic N fertilizers applied to managed soil.

Overall, the model estimated emissions correlate with FAO and UNFCCC data inventory with an R^2 of 0.99 and 0.98, respectively ($p<0.001$). As discussed earlier, according to our estimations, China, India, U.S.A, and Brazil are the top four emitters globally, which is also reflected in FAO emission data (Figure 6). In comparison to FAO, we are slightly underestimating the emissions for China and Brazil. However, our estimates are close to FAO for the other two countries, India and U.S.A. For countries like the U.S.A and Australia, our estimates are more comparable to FAO than UNFCCC, which might be because we are following the IPCC Tier 1 approach to calculate the final direct emissions from N fertilizer amounts like FAO. On the other hand, UNFCCC reporting countries use IPCC's higher tier approaches (Tier 2 and 3) for emission estimates.



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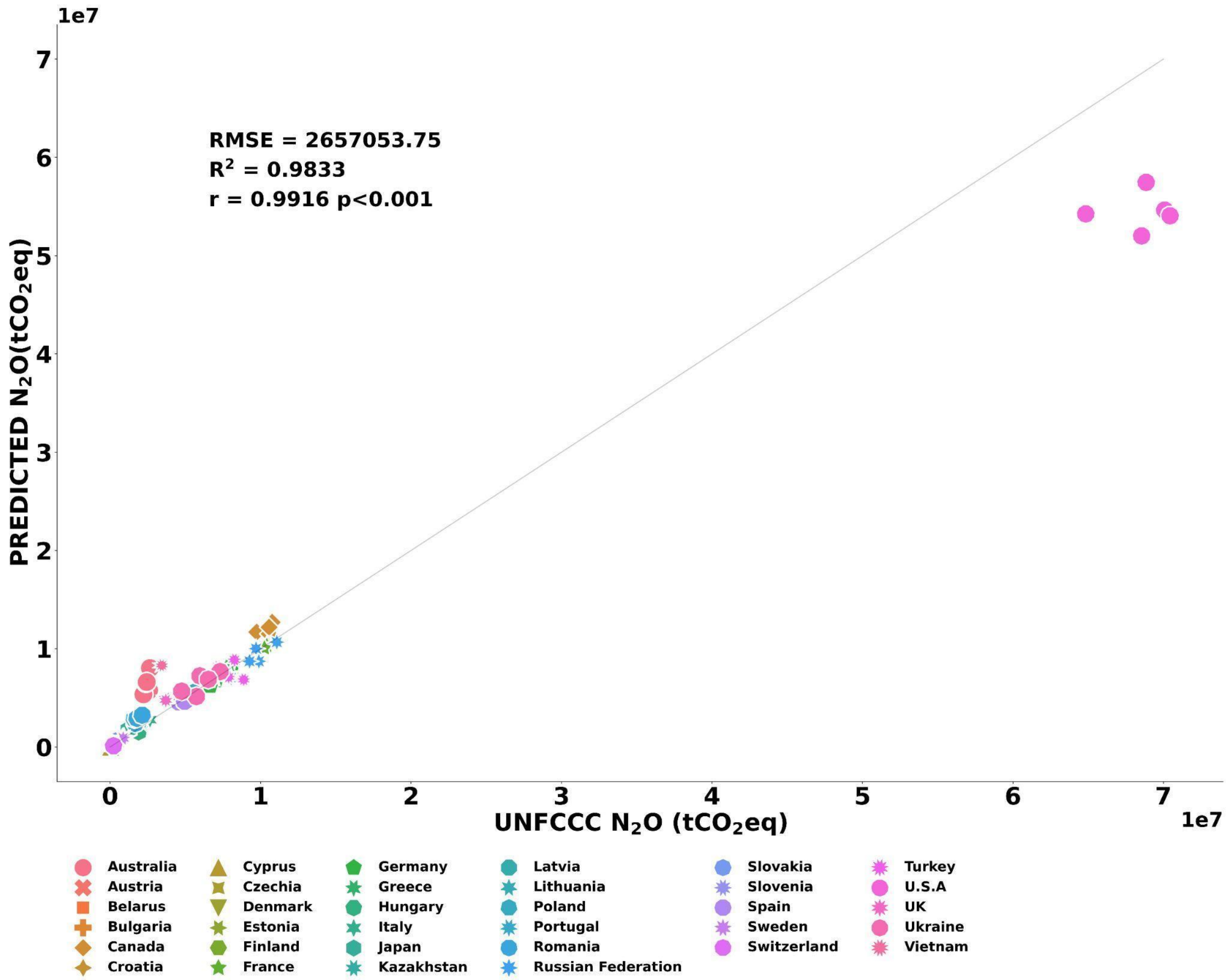

Figure 6 The model predicted N_2O validation with FAOSTAT (top image) and UNFCCC (bottom image) at the country-level for the time period 2016-2019. Note, each figure has different scales.

To further analyze the results, we compared emission in the form of time series for 24 countries from 2015 to 2019 with FAO and UNFCCC inventory (Figure 7). These 24 countries are selected from each of the five continents (Asia, America, Europe, Africa, and Oceania). According to our calculation, in total, these 24 countries represented 86.4%, 85.8%, 85.2%, 85.0%, 84.0%, and 85.4% of total global emissions in 2015, 2016, 2017, 2018, 2019, and 2020, respectively.

In countries with both FAO and UNFCCC estimations available, for example, in the USA, our predicted estimates of 182 Gg, 193 Gg, 174 Gg, 183 Gg, and 181 Gg are closer to the FAO 185 Gg, 183 Gg, 185 Gg, 183Gg, 184 Gg, for period 2015-2019. Similarly, for Australia and Canada, our estimations follow FAO estimation. However, our estimates are underestimated compared to UNFCCC (217 Gg, 231 Gg, 230 Gg, 235 Gg, 236 Gg) for the USA and overestimated for Australia and Canada. On the other hand, for a country like Russia, this pattern is reversed; our predicted emissions of 24 Gg, 29 Gg, 33 Gg, 29 Gg, and 36 Gg are closer to UNFCCC estimates of 27 Gg, 31 Gg, 32 Gg, 33 Gg, 37 Gg for the period 2015-2019. In our analysis, Germany has a continuous reduction in the emissions from 2015 (28.4 Gg) to 2019 (20.9 Gg), which matches the UNFCCC inventory, 27.3 Gg in 2015 to 22.3 Gg in 2019 (Figure 7). The discrepancies between our estimates and UNFCCC might be due to the difference in method. For example, countries like the USA use different approaches for national scale N_2O emissions assessments, which involve process-based models, such as the DAYCENT (Del Grosso et al., 2012), along with the IPCC emission factor. For some countries UNFCCC emission values are not available between 2015 to 2019, and we compared our emission with FAOSTAT only. In China, which is the highest emitting country (as discussed earlier), our estimates are underestimating emissions as compared to FAOSTAT. However, both are showing decreases in emission levels from 2015 to 2019. Other high emitting countries like India and Pakistan show a close match to FAOSTAT estimates.

In addition to time-series comparison, to further analyze the changes in emissions in the last five years, we have compared the percentage change in emissions between five years from 2015 to 2019 to FAO and UNFCCC (if available; Table S2). Positive or negative percent change indicates increase or decrease in emissions level between 2015 and 2019, respectively. According to our estimates 14 countries out of 24 are showing the same sign (positive or negative) of percent change in emissions compared with FAO or UNFCCC. Another contrasting difference between our method and FAO estimates is the temporal variability in the emissions. Our model suggests high temporal variability in emissions for countries like Argentina, Australia, and Brazil compared to FAO estimates. The reason could be that our predicted NUE for crops depends on the change in crop yield from one year to another. Therefore, if yield changes from one year to another within a country, as it normally does, changes in the emissions are reflected in our estimates. To further validate our estimations, we compared crop-specific country-level N fertilizer amounts of 7 crops (Maize, Wheat, Rice, Sugar cane, Rapeseed, Tea, Sugar beet) with IFASTAT data for 59 countries (Figure S1). Out of these 7 crops, Maize receives the maximum amount of N fertilizers, representing 20 % of global N fertilizer used on all the crops, followed by wheat with 18% and rice 16 % (IFA report, 2022). These three crops alone represent more than 50% of the total N fertilizer used globally. Our estimates show statistically significant correlation with IFASTAT data inventory with an R^2 of 0.94.

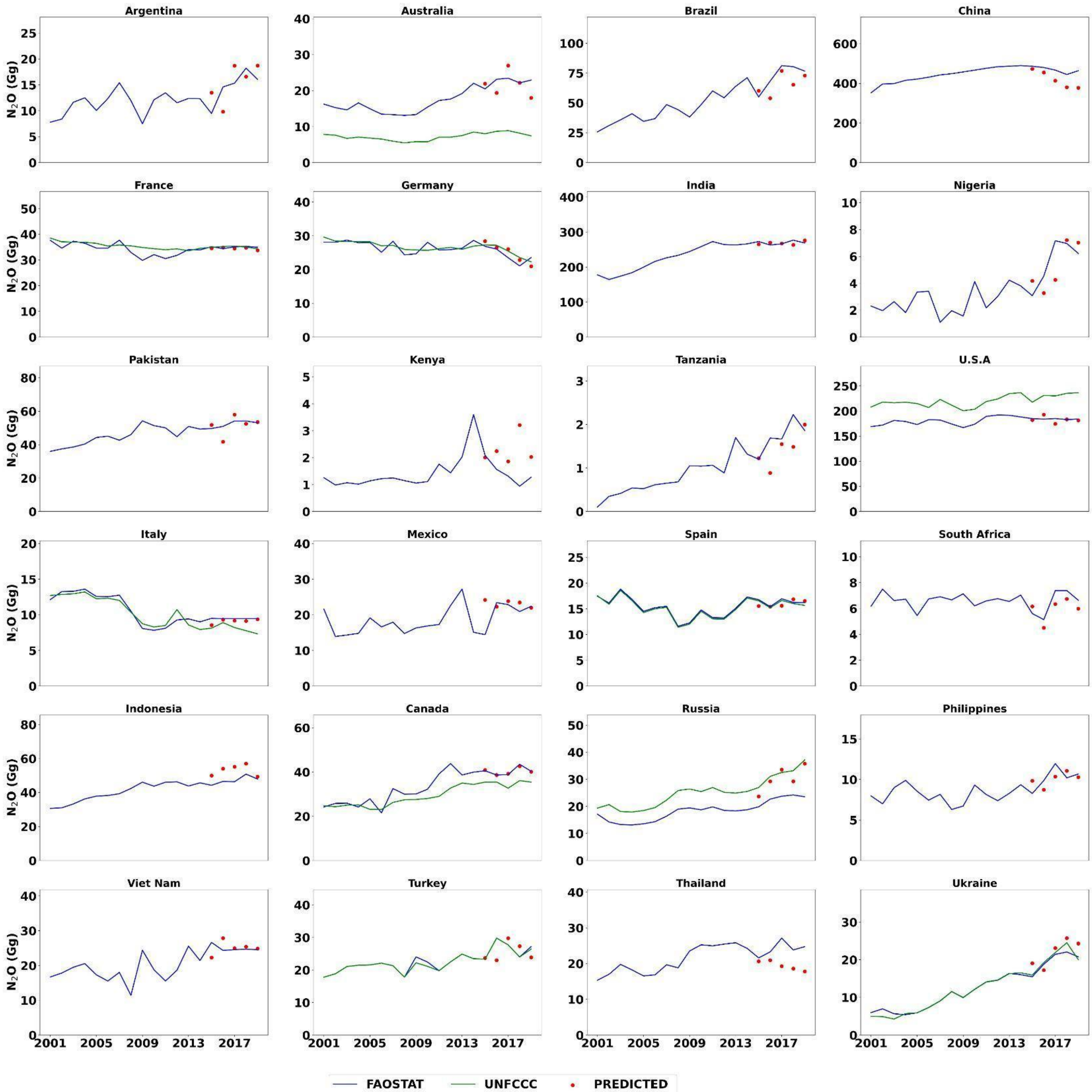

Figure 7 Time-series comparison of model-predicted N_2O emissions (red-dots) with FAO (blue-line) and UNFCCC inventory (green-line). Units- y-axis: Gigagram; x-axis: Year. Note: y-scales are different for each country. For the x-axis, FAO and UNFCCC data is plotted for the time period 2001-2019 and the model calculated emissions are for the time period 2016-2019.

4. Conclusion

In this study, we estimated direct N_2O emissions from synthetic N fertilizer use at a country scale using a novel data-based model which assumes crop N uptake is proportional to N fertilizer application. Our estimates include 94 countries, representing the majority of total synthetic nitrogen fertilizer use globally (~98%). Total direct N_2O emissions were 1623, 1610, 1636, 1585, 1600, and 1652 gigagrams (Gg) for years 2015, 2016, 2017, 2018, 2019, and 2020, respectively. The top 7 emitters included China, India, the U.S.A., Brazil, Pakistan, Indonesia, and Canada, contributing more than 60% of total emissions (>900 Gg) annually. Emissions accumulated at a continental level suggest AS, AM, and EU as the significant emitting continents with a total emission of 1542 Gg (95.5 %) to the total emissions at a global scale (averaged over the study period). Our study can help to figure out the countries with higher emissions, which can guide the government in those countries to tighten their policies for emission reduction.

Even though our method to calculate the emissions was very different from FAO or UNFCCC, and it embeds assumptions (defensible for the most part given the scale of the estimates), overall, the predictions show a good correlation with other global emission inventories. Therefore, independently validating the results of other direct N_2O emissions inventories. This validation will build more trust in the present emissions inventories while also finding the inconsistencies if they exist.

As a next step, we aim to reduce uncertainties in our method further. For example, we use crop yield and area based on national census data of FAO, which is self-reported by countries and can be problematic for countries that might have under or over-reported their agricultural areas. For example, Seto et al. (2000), discuss the high differences in the satellite-derived and reported agricultural land area in China. Satellite-derived estimates of total agricultural area in the ten counties were 115% greater than the reported in government yearbooks. Similarly, national standards of agricultural data collection remain poor in many regions, such as sub-Saharan Africa (Carletto et al., 2013).

In the next phase, we will include more refined approaches based on remote sensing data for crop area and crop vigor estimates, machine learning algorithms to train predictions on existing datasets, and process-based models (*i.e.,* SALUS) to provide ancillary data on soil water dynamics, soil temperature, infiltration, soil N availability and N_2O estimates. In addition, we will improve our assessment of NUE for the crops using proxies such as climate (*i.e.,* Growing Degree Days, precipitation), satellite-derived vegetation indices (*i.e.,* Normalized Difference Vegetation Index, NDVI), and drought indices, including actual evapotranspiration (AET) and potential evapotranspiration (PET).

5. Acknowledgements

This study was funded by Climate TRACE.

6. Supplementary materials

Table S1 Total direct N_2O emissions due to synthetic nitrogen fertilizers for 5 continents for years 2015-2020 (all values rounded to one decimal place). Note, only countries modeled are included in the continent estimates.

	Total emission (Gg)					
Continent	**2015**	**2016**	**2017**	**2018**	**2019**	**2020**
Asia	985.7	969.9	954.2	910.0	918.9	960.5
America	349.5	348.4	363.3	362.1	362.5	378.0
Europe	219.9	226.9	239.3	238.0	240.1	239.1
Africa	46.4	45.8	52.4	52.6	60.0	54.4
Oceania	21.9	19.4	27.0	22.2	18.0	19.7

Table S2 Comparison for the percent change in emissions at country scale between the 5 years period from 2015 to 2019 for this study and other inventories.

Country	% Change in emissions from 2015 to 2019		
	Our study	**FAOSTAT**	**UNFCCC**
Argentina	38.61	69.35	Not available
Australia	-18.03	12.05	-7.22
Brazil	21.21	39.39	Not available
China	-20.21	-4.63	Not available
France	-2.25	-0.68	-1.55
Germany	-26.23	-12.36	-18.24
India	4.08	-1.53	Not available
Nigeria	68.15	101.98	Not available
Pakistan	3.30	6.90	Not available
Kenya	0.91	-38.94	Not available
Tanzania	63.24	54.7	Not available
U.S.A	-0.35	-0.51	8.64
Italy	9.54	-0.48	-9.85
Mexico	-9.08	55.15	Not available
Spain	6.23	-3.41	-5.38
South Africa	-2.87	18.30	Not available
Indonesia	-1.24	8.25	Not available
Canada	-1.83	-0.54	-0.03
Russian Federation	51.12	18.73	37.71
Philippines	4.55	29	Not available
Vietnam	11.65	-7.91	Not available
Turkey	0.91	16.35	13.18
Thailand	-13.75	14.55	Not available
Ukraine	27.51	34.27	25.54

Table S3 GAEZ crop classification

1. Wheat	10. Yams and other roots	19. Olives
2. Rice	11. Sugarbeet	20. Cotton
3. Maize	12. Sugarcane	21. Tobacco
4. Sorghum	13. Pulses	22. Banana
5. Millet	14. Soybean	23. Stimulants
6. Barley	15. Rapeseed	24. Vegetables
7. Other cereals	16. Sunflower	25 CropsNES
8. Potato and Sweet potato	17. Groundnut	26 Fodder crops
9. Cassava	18. Oil Palm fruit	

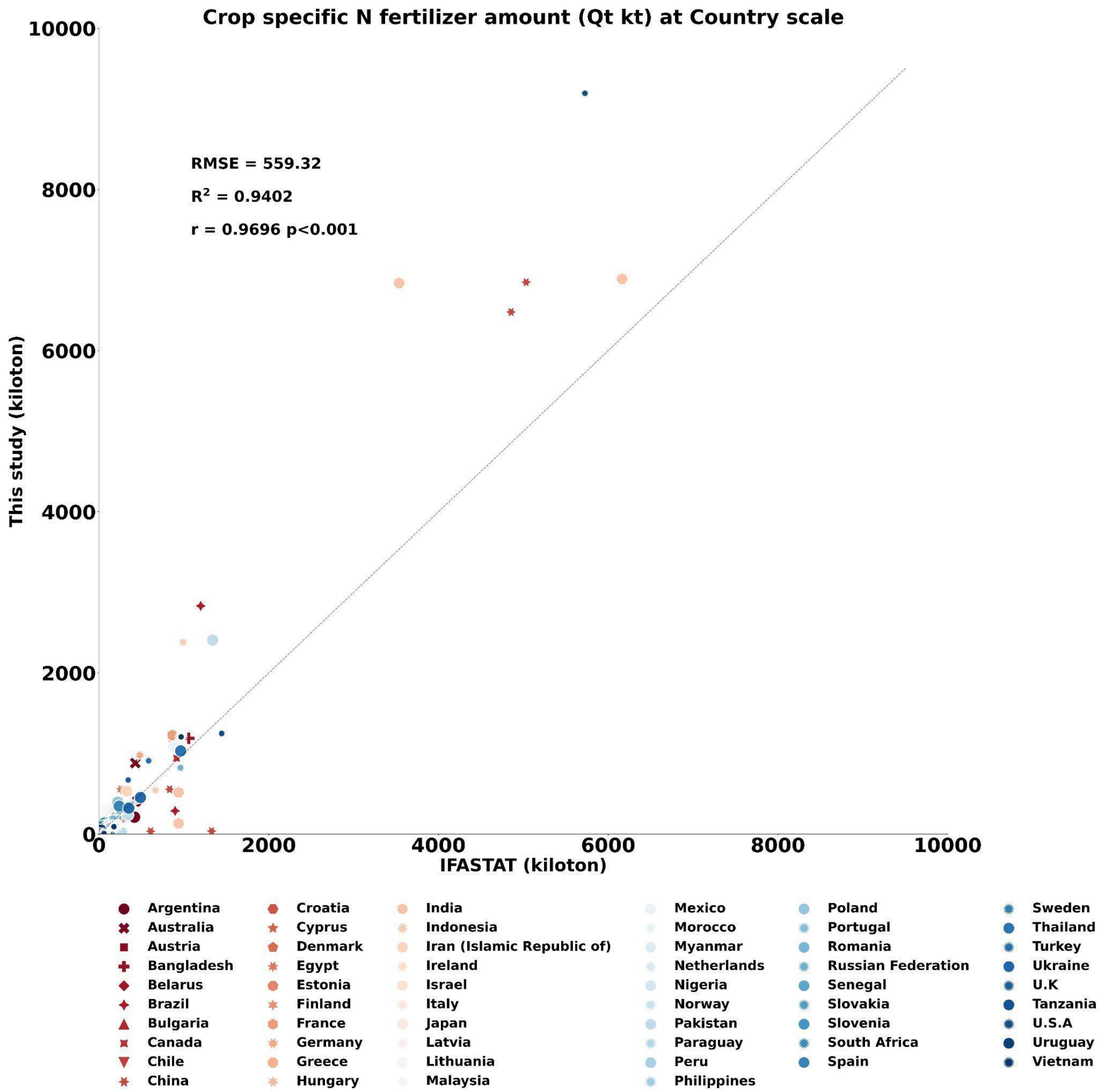

Figure S1 Comparison of crop-specific Nitrogen fertilizer at country scale between our study and IFASTAT for 2018. Units- y-axis and x-axis: kiloton (1000 metric ton).

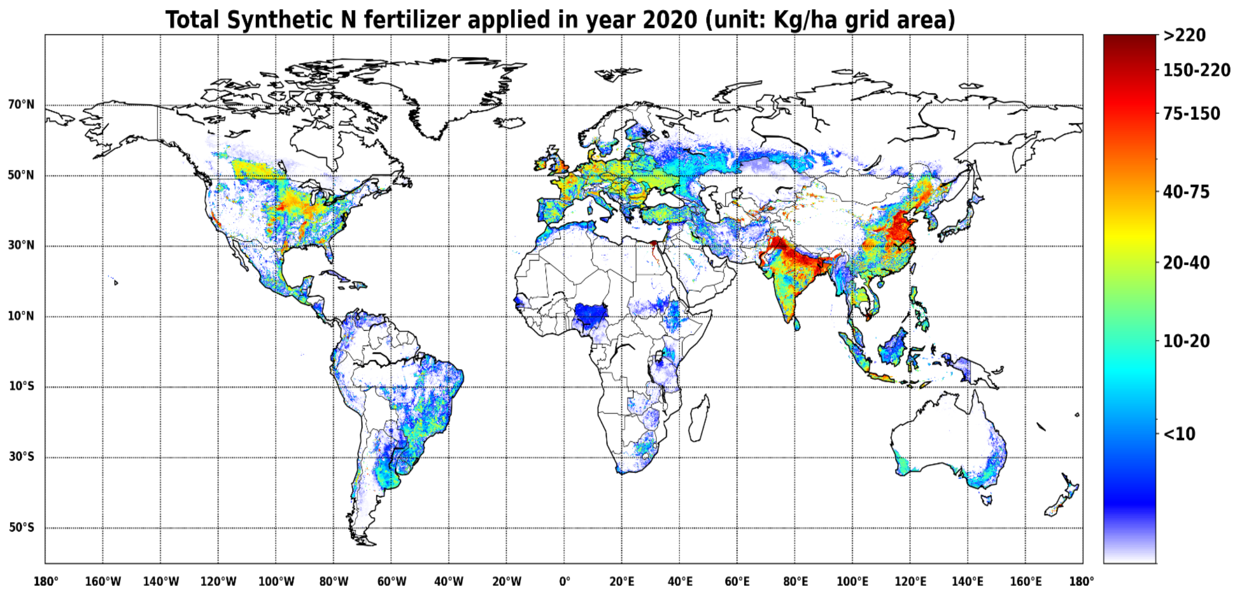

Figure S2 Total nitrogen fertilizer applied at grid scale 0.083° resolution (~10 km) for 2020. Units: Kg/ha grid area.

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