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Title Spatially explicit estimates of N_2O emissions from croplands suggest climate mitigation opportunities from improved fertilizer management

Running head Spatially explicit N_2O emission estimates

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Abstract

With increasing nitrogen (N) application to croplands required to support growing food demand, mitigating N_2O emissions from agricultural soils is a global challenge. National greenhouse gas emissions accounting typically estimates N_2O emissions at the country scale by aggregating all crops, under the assumption that N_2O emissions are linearly related to N application. However, field studies and meta-analyses indicate a nonlinear relationship, in which N_2O emissions are relatively greater at higher N application rates. Here we apply a super-linear emissions response model to crop-specific, spatially-explicit synthetic N fertilizer and manure N inputs to provide subnational accounting of global N_2O emissions from croplands. We estimate 0.66 Tg of N_2O-N direct global emissions circa 2000, with 50% of emissions concentrated in 13% of harvested area. Compared to estimates from the IPCC Tier 1 linear model, our updated N_2O emissions range from 20-40% lower throughout Sub-Saharan Africa and Eastern Europe, to >120% greater in some Western European countries. At low N application rates, the weak non-linear response of N_2O emissions suggests that relatively large increases in N fertilizer application would generate relatively small increases in N_2O emissions. Since aggregated fertilizer data generate

underestimation bias in nonlinear models, high-resolution N application data are critical to support accurate N_2O emissions estimates.

Introduction

Agriculture accounts for ~20-30% of global greenhouse gas emissions (Vermeulen *et al.*, 2012) and produces the majority (~59%) of anthropogenic N_2O emissions (Ciais *et al.*, 2013). Nitrous oxide is a potent greenhouse gas, and is the most important contributor to stratospheric ozone depletion, with associated negative health impacts (Wolfe & Patz, 2002), and decreased plant productivity (Sitch *et al.*, 2007). The largest source of N_2O emissions from agriculture is synthetic N fertilizer and manure application to croplands (Syakila & Kroeze, 2011), which is projected to increase by ~50% from 2000 to 2050 (FAO, 2012). Between 2001-2011, annual N_2O emissions from synthetic and manure fertilizers increased by 37% and 12%, respectively (FAO 2014b). Consequently, reducing N_2O emissions from croplands is critical for addressing climate change and ozone depletion concerns.

N_2O is produced from microbially-mediated nitrification and de-nitrification processes in soils, leading to emission rates that are modified by diverse climate, soil, and vegetative conditions, and are highly variable over time and space (Stehfest & Bouwman, 2006; Philibert *et al.*, 2012). These 'direct' emissions are distinct from 'indirect' emissions in which N_2O is formed from N volatilized or leached from managed soils (De Klein *et al.*, 2006), and N_2O emissions associated with land use change (Flynn *et al.*, 2012).

Emission factors (EF) are often used to relate applied N to N_2O emissions across broad spatial scales (De Klein *et al.*, 2006). For instance, the IPCC Tier 1 default method for estimating direct N_2O emissions from managed soils, hereafter referred to as the "linear model," predicts that 1% of applied N fertilizer is emitted as direct N_2O emissions (i.e., EF = 0.01) (De Klein *et al.*, 2006). For flooded or paddy rice, N_2O emission rates are lower because N_2O is unstable in the anaerobic conditions of wetland soils (Lal, 2006). Consequently, the IPCC suggests a lower emissions factor of 0.31% for calculating emissions from paddy rice (De Klein *et al.*, 2006). Using such linear methods, recent bottom-up estimates of direct N_2O emissions from synthetic N fertilizer application to crops combined with FAOSTAT estimates of direct N_2O emissions due to manure applied to soils are well-constrained, ranging from 1.0-1.2 Tg N_2O-N yr^{-1} (Supporting Information).

Despite the relative ease of applying linear emissions models to estimate N_2O emissions from crops, recent syntheses of field observations suggest a highly non-linear response. Specifically, N_2O emissions accelerate with increased N application (Philibert *et al.*, 2012; Kim *et al.*, 2013; Shcherbak *et al.*, 2014). This "superlinear" response is likely due to the relatively greater excess N unused by the crops at higher fertilization levels; this extra N is available to be emitted as N_2O (Van Groenigen *et al.*, 2010; Kim *et al.*, 2013; Shcherbak *et al.*, 2014). Reduced uncertainty associated with non-linear emissions models is well supported in the literature (Hoben *et al.*, 2011; Philibert *et al.*, 2012; Shcherbak *et al.*, 2014).

Until recently, sub-national crop-specific fertilizer application data with global coverage have been unavailable. Such spatially-explicit and crop-specific estimates of fertilizer-derived N_2O

emissions pinpoint particularly low- and high-emission locations and crop types, and are therefore vital for addressing these negative social and environmental impacts of fertilizer use (Montzka *et al.*, 2011; Reay *et al.*, 2012). Combining non-linear emissions models with improved accuracy with spatially resolved fertilizer application rates is a significant step towards global and accurate mitigation assessments (Reay *et al.*, 2012; Shcherbak *et al.*, 2014).

Here, we generate relatively accurate and crop-specific N_2O emissions estimates from global croplands. First, we update a recently developed non-linear N_2O emissions model (Philibert *et al.*, 2012) by incorporating additional emissions data sets (Shcherbak *et al.,* 2014, Stehfest & Bouwman 2006), extending the range of N application rates to 700 kg-N/ha, and differentiating paddy rice. We develop crop-specific estimates of manure application to croplands, and combine these rates with previously published estimates of synthetic N application (Mueller *et al*., 2012). With the updated model and N fertilizer application rates, we calculate spatially-explicit, crop-specific global N_2O emissions, and contrast these results with the IPCC Tier 1 linear model. Finally, we identify crops and regions where small changes in N application would generate large changes in N_2O emissions.

Materials and methods

Non-linear N_2O emissions model and uncertainty calculations

In the non-linear “NL-N-RR” model (NLNRR indicates a non-linear (NL) nitrogen effect (N) random intercept (R) random effect (R) model, henceforth “Philibert model”) of Philibert *et al.* (2012), N_2O emission rates are estimated from N fertilizer application rates using an exponential model with random parameters. Philibert *et al.* (2012) determined that this type of model performs better than linear models and exponential models with fixed parameters. The Philibert model was developed from a dataset of global N_2O emissions and N fertilizer application rates compiled by Stehfest & Bouwman (2006). Yet, the Stehfest & Bouwman (2006) dataset contains sparse data on high N application rates (>500 kg-N/ha) and limited experiments from major global ecosystems (e.g., Mediterranean) and regions (e.g., China). Moreover, N_2O emissions are reduced under continually flooded conditions such as those typical within rice paddies (De Klein *et al.*, 2006), yet the Philibert model does not account for such effects. Therefore, we updated the Philibert model by re-fitting this model to a dataset including experiments compiled by Shcherbak *et al.* (2014). We thus extended the experimental dataset from 985 to 1644 datapoints, including 30 experiments with N application rates >500 and ≤700 kg N ha^{-1}, and 125 experiments conducted in flooded rice.

To include the flooded rice effect, we developed an updated version of the original model that includes a specific parameter differentiating flooded rice from other crops. The model is based on the following equation:

$$Y_{ijk} = \exp\left(\alpha_{0i} + \alpha_{1i} X_{ij} + \beta Z_{ij}\right) + \varepsilon_{ijk} \qquad (1)$$

Here, Y_{ijk} is the N_2O emission rate (kg N ha^{-1} yr^{-1}) measured at the i^{th} experiment in the dataset (i=1 ... 259), for the j^{th} applied N dose X_{ij} (j=1 ... N_i), and the k^{th} replicate (k=1 ... K_{ij}). Z_{ij} is a binary variable equal to 1 if the crop is “flooded rice” and equal to zero otherwise, and β is a parameter corresponding to a “discount factor” for N_2O emission in flooded rice fields. The random terms α_{0i}, α_{1i}, and ε_{ijk} are assumed to be independent and normally distributed (as in Philibert *et al.,* 2012):

$$\varepsilon_{ijk} \sim N\left(0, \tau^2\right),\ \alpha_{0i} \sim N(\mu_0, \sigma_0^2)\ ,\ \alpha_{1i} \sim N\left(\mu_1, \sigma_1^2\right) \tag{2}$$

where α_{0i} is the log location-specific background emission, α_{1i} is the log location-specific applied N effect, ε_{ijk} is the residual error term, μ_0 is the log mean background emission, μ_1 is the log mean applied N effect, and $N(\mu, \sigma^2)$ represents a normal distribution with mean μ and standard deviation σ. The standard deviations σ_0 and σ_1 describe the variability of α_{0i} and α_{1i} across site-years. The values of μ_0, μ_1, β, σ_0, σ_1, and τ were estimated by an approximate maximum likelihood method, with the `nlme` statistical package in R (Pinheiro & Bates, 2000), as described by Philibert *et al*. (2012). The estimated parameter values are presented in Table S1. The resulting model, which we refer to as $NLNRR_{700}$, and a simpler version without a discount factor for flooded rice, are shown in Figure S4.

Emissions were averaged over site-years using the estimated values of the model parameters reported in Table S1. To analyze uncertainty and generate a confidence interval (CI) for the N_2O emissions model, we first sampled values for the parameters μ_0, μ_1, β in the probability distribution of their estimators (Table S1). For each sample of parameter values, we then generated mean values of N_2O emissions by averaging over the distribution describing site-year variability (i.e., $N(\mu_0, \sigma_0^2)$ and $N\left(\mu_1, \sigma_1^2\right)$). We repeated this process 20 thousand times to determine the 5th and 95th percentiles of the resulting distribution of N_2O emissions estimates.

Flooded rice distribution data

Rice cultivation consists of three major water management strategies: irrigated, rainfed, and upland. Irrigated and rainfed, or "flooded," rice fields typically emit less N_2O in response to additional N application compared to other crops and upland or "dry" rice (Akiyama *et al.*, 2005). Thus, differentiating between flooded and dry rice is essential to generate accurate crop-specific N_2O emissions estimates. To estimate the irrigated fraction of total rice harvested area, we used the MIRCA2000 dataset (Portmann *et al*., 2010), which includes monthly irrigated and rainfed rice growing areas, and maximizes consistency with the cropland data of Monfreda *et al*. (2008). For each of the 402 spatial units in the dataset, we calculated the fraction of irrigated area compared to total area (irrigated + rainfed) and then applied these fractions to the Monfreda *et al*. (Monfreda *et al.*, 2008; Portmann *et al.*, 2010) dataset. Doing so, we find that in 2000, 59% of rice harvested area was irrigated.

Within the remaining non-irrigated fraction (41%), we further divided rice into upland and rainfed systems. Huke and Huke (1997) present a comprehensive assessment of rice cultivation types across monsoon Asia, excluding Japan, circa 1990. We ingested these data into a vector-based GIS database, and converted them to 5 arc-minute raster data for analysis. We used the ratio of upland rice to the deep water plus rainfed area to assess the relative proportion of upland rice in each non-irrigated grid cell fraction. In regions not covered by Huke and Huke (1997), we applied the mean upland proportion from regions for which data are available. Overall, we find that 93% of total 2000 era rice harvested area is flooded.

Crop-specific synthetic N fertilizer data

The crop-specific synthetic N fertilizer dataset utilized for this study was compiled by Mueller *et al.* (2012), and provides estimates of synthetic N fertilizer application rates by crop circa 2000 (1997-2003). Data include national fertilizer consumption (across all crops), subnational consumption (across all crops), national crop-specific application rates, and subnational crop-specific application rates. These data were sourced from the UN Food and Agricultural Organization, fertilizer industry associations, fertilizer research institutes, and national agricultural or statistical agencies.

Crop-specific manure application data

For manure N inputs, we used gridded livestock manure maps (Herrero *et al.*, 2013), which represent 5 arc minute resolution estimates of pig, bovine meat, bovine milk, poultry, and sheep/goat manure production circa 2000. To calculate the fraction of total manure production applied to croplands, we used manure management data. These data consist of livestock-specific, regional estimates of manure management across livestock systems for bovines and sheep/goats (Robinson *et al.*, 2011), and across smallholder and industrial systems for poultry and pigs (Herrero *et al.*, 2013). We computed the mass of manure N applied to croplands (NA, kg yr^{-1}):

$$NA = N \times F_{MS} \times (1 - F_{MSO}) \times (1 - F_{LossMS}) \tag{3}$$

where N is total nitrogen produced (kg yr^{-1}), F_{MS} is the fraction of total manure managed, F_{MSO} is the fraction of managed manure destined to other uses (e.g., production of biogas), and F_{LossMS} is the fraction of managed manure N lost (e.g., through volatilization and leaching; Table S6).

We assumed that manure is applied only within the 5 arc minute grid cell in which it was produced, and computed manure application rate (kg ha^{-1} yr^{-1}) by dividing *NA* by crop harvested area (Monfreda *et al.,* 2008).

In some regions, estimated manure application rates are extremely high due to the large number of animals relative to cropland area. As an estimate of the upper-bound of manure applied to croplands in such situations, we capped total manure application at 700 kg N ha^{-1}, which exceeds the 99th percentile of the global manure application rate. For leguminous crops, we allowed manure application until total N applied (synthetic + manure) reached the 99th percentile of the global synthetic N application rate to the crop in question. We assumed a maximum combined synthetic + manure N application rate of 700 kg N ha^{-1}. To estimate manure applied to individual crops, we multiplied these capped manure application rates by crop harvested area.

We estimate 7.8 Tg of manure N applied to crops, which represents ~9% of the 86.3 Tg total N applied in the form of synthetic and manure fertilizer. This estimate is substantially smaller than other year 2000 estimates of manure-N applied to crops (e.g. 17.3 Tg, Liu *et al*., 2010) due to our use of more refined animal- and region-specific management factors describing the proportion of manure applied to crops (full discussion in the Supporting Information).

Response to marginal change in application rates

To identify crops and locations where altering N application rates would have a disproportionate effect on N_2O emissions, we calculated the incremental N_2O emissions change in response to a small change in N application. Specifically, we computed the change in total N_2O emissions due to a uniform additive incremental change in applied N. This calculation was carried out with high-resolution numerical differentiation. For conceptual clarity, we express results in terms of a marginal but finite N application rate change of +1 kg N ha^{-1}.

Sensitivity Analysis

We performed several sensitivity analyses. First, we quantified the impact of changes to the upper limit of N application in experimental data by examining total global emissions when the model was fit to datasets where the upper limit ranged from 500-700 kg N ha^{-1}. Second, we compared global direct emissions estimates estimated with our newly-developed model to those derived from the Philibert model. Finally, since assumptions of homogeneous fertilizer application rates can lead to underestimation bias for emissions estimates based on a superlinear model such as ours, we explored sensitivity to sub-regional-scale fertilizer application rate heterogeneity. Specifically, we constructed randomized fertilizer application datasets such that each pixel within the application rate dataset was a Gaussian random variable with a mean value equal to the sum of synthetic and manure N application, and a standard deviation equal to a constant multiple of the mean value. Resulting negative N application rates were set to 0, N application values >700 kg N ha^{-1} were retained but emissions estimates were calculated using the emissions factor corresponding to applied N=700 kg ha^{-1}.

Model Intercomparison

We compared emissions outcomes from our differentiated non-linear to alternative models – the linear IPCC Tier 1 model, and the non-linear model of Shcherbak *et al.* (2014) – by applying these models to our fertilizer application dataset.

Results

In 2000, we estimate 0.66 Tg N_2O-N (CI 0.56 to 0.78 Tg N_2O-N) total global direct N_2O emissions associated with 86.3 Tg of N applied to crops (78.5 Tg synthetic N and 7.8 Tg manure N, Figure 1), a global mean fertilizer application rate of ~68 kg N ha^{-1}. These N_2O emissions are highly concentrated, with 50% of emissions sourced from only 13% of the global cultivated area. (Figure 2).

Implied Non-Linear Emissions Factors

While the global mean non-linear emissions factor is 0.77%, implied emissions factors are influenced by the magnitude and variance of N application rates, and therefore differ greatly among crops and regions.

Wheat cropping generates 0.14 Tg N_2O-N, more N_2O than any other crop, and has a mean emissions factor of 0.82%. While maize receives 19% less total N fertilizer than wheat, higher N application rates generate an emissions factor of 0.91%, and maize's 0.12 Tg N_2O-N emissions are only 10% less than wheat. Potato is produced with mean N application rates of 98 kg ha^{-1}, 2.6% lower than those of maize, but has an emissions factor of 0.94% that is 3% higher due to more heterogeneous N application rates (Table 3). Soybean, a leguminous crop that fixes much of its own N and therefore requires relatively little N fertilizer input (mean of 29 kg ha^{-1} globally), has the lowest emissions factor of top crops (excluding flooded rice) at just 0.65%.

N_2O emissions vary widely across countries and regions (Table 2). China is the leading N_2O emitter (0.20 Tg N_2O-N, 31% of global emissions), followed by India and the United States. Emissions factors in these countries are 0.80%, 0.62%, and 0.84%, respectively. Although Western Europe's total emissions are lower than emissions in Asia and North America, the region has a mean emissions factor of 0.95% and hosts the two countries with the highest global emissions factors (Netherlands EF = 2.4%, Belgium EF = 2.3%). Some countries within low emission regions have very high intensities; for example, Egypt has an average fertilizer application rate of 199 kg N ha^{-1}, with crop-specific rates > 400 kg N ha^{-1} on 6.3% of cultivated area, resulting in a mean national emission factor of 1.34%. In contrast, Eastern Europe and Sub-Saharan Africa share the lowest implied emission factor of all global regions, just 0.69%.

As a result of highly heterogeneous N_2O emissions rates across crops and regions, some crop-country combinations produce particularly high or low total emissions (Table 1). Maize and wheat cultivation in the United States and China produces 21% of total global N_2O emissions. Since vegetable and fruit cultivation frequently requires high N fertilizer inputs, vegetable and melon production in China generates 4.5% of total global N_2O emissions. China's paddy rice, on the other hand, is the leading crop-country consumer of N fertilizer (receiving 6.3 Tg) but contributes only 3.0% of total global direct N_2O emissions from croplands.

Disproportionate N_2O emissions responses

A uniform addition of 1 kg N ha^{-1} across global croplands generates mean additional emissions of 0.0080 kg N_2O-N ha^{-1} (Fig 3.) While this global response is similar to the additional 0.0089 kg N_2O-N ha^{-1} derived using linear emissions factors, some regions show disproportionate responses including China (0.014 kg N_2O-N per kg N applied, 42% greater than the global average) and sub-Saharan Africa (0.0061 kg N_2O-N per kg N applied, 23% less than the global average). Regional responses within countries (Table S10) can vary greatly. For example, Shandong and Hunan provinces in China have differential response rates of 0.0161 and 0.0076 kg N_2O-N kg^{-1} N applied respectively.

Sensitivity Analysis

The 18% increase in emissions associated with the 95^{th} percentile model parameters (0.78 Tg N_2O-N) compared to mean emissions (0.66 Tg N_2O-N) provides a basis of comparison with other sources of uncertainty. Notably, this same increase in emissions can also be obtained by assuming fertilizer application rate heterogeneity at the sub-regional scale with a coefficient of variation (CV) of 54%. To further quantify the scale of impact of fertilizer application rate inhomogeneity: if N_2O emissions for the United States were calculated after aggregation of crop-specific fertilizer application rates to the national level, total emissions derived from our non-linear $NLNRR_{700}$ model would be 0.077 Tg N_2O-N instead of 0.090 Tg, an underestimation of 16%.

In contrast, increasing the maximum fertilizer application rate to 800 kg N ha^{-1} generates a 0.02% increase in total N applied, and with an assumption of constant EF beyond N application rates of 700 kg ha^{-1}, we find a 1.0% increase in total N_2O emissions. Therefore, our results are relatively insensitive to our choice of a maximum N application rate of 700 kg N/ha. Excluding manure N inputs, we find global direct N_2O emissions of 0.57 Tg. This implies a 15% increase in direct N_2O emissions in response to manure application, which adds 10% to total N application beyond synthetic N.

Model intercomparison

Emissions estimates calculated using the nonlinear model developed here are generally lower than results from the linear model. Our global emissions factor is 0.77, which is 14% lower than the mean global emissions factor of 0.89 calculated with the linear model. Even greater differences are apparent among regions (Table 1, Fig. 4). In China, where N application averages 158 kg ha^{-1}, our non-linear N_2O emissions estimate is 6% lower than the linear estimate (Table 2). In contrast, extremely low N application rates (11-42 kg ha^{-1}) throughout most of Sub-Saharan Africa, Eastern Europe, and Latin America lead to N_2O emissions ~26-31% lower than assessed with the linear approach (Table 3). However, in administrative units with very high N application rates, the nonlinear model occasionally estimates higher N_2O emissions. For example, China's provincial N_2O emissions estimates range from 6% greater (Hubei, Jiangsu, Napp ~210 kg ha^{-1}) to 15% lower (Heilongjiang, Napp = 114 kg ha^{-1}) than linear predictions (Table S3).

Discussion

By pinpointing crops and regions associated with disproportionately high or low N_2O emission levels, non-linear models such as the one developed and applied here offer the potential for identifying emission mitigation priorities, as well as locales where additional N application would be highly beneficial, increasing yields and reducing the emissions intensity of agriculture (Verge *et al.*, 2007; Tubiello *et al.*, 2013; West *et al.,* 2014). For example, Shandong province in China emits ~4% of global cropland N_2O, yet comprises just 1% of crop harvested area. Reducing N application rates by 5% in this province would cut provincial crop N_2O emissions by 9% and global crop N_2O emissions by 0.35%. In contrast, increasing N fertilizer application by 5% over Sub-Saharan Africa would increase N_2O emissions by just 2.7%. In sum, we bring greater accuracy to sub-global estimates of N_2O emissions associated with N fertilizer application to croplands.

Our results illustrate how refined empirical models of biogeochemical relationships require resolved data inputs to generate accurate predictions. Recently available sub-national synthetic fertilizer and manure distribution data, coupled with a sophisticated emissions model, demonstrate that N_2O emissions rates are unevenly distributed across the world's croplands. Future models must be constrained by a greater diversity and quantity of field studies, which are still lacking in certain regions such as the tropics (Stehfest & Bouwman, 2006; Montzka *et al.*, 2011; Reay *et al.*, 2012; Shcherbak *et al.*, 2014).

Comparison to previous estimates of N_2O emissions from global croplands

Total year 2000 global N_2O emissions of 0.66 Tg N_2O-N (CI 0.56 to 0.78 Tg N_2O-N) generated from cropland synthetic and manure N inputs to our nonlinear model are substantially lower than previous global assessments that applied linear emissions factors to synthetic N application rates and suggest direct N_2O emissions ranging from 0.8 to 1.0 Tg N_2O-N (Bouwman, 1996; De Klein *et al.*, 2006; Verge *et al.*, 2007; Flynn & Smith, 2010; Tubiello *et al.*, 2013). Since our aggregate global estimate includes emissions generated from manure N inputs, the reduced emissions produced from our model are particularly striking, and are lower for three main reasons. First, unlike these previous global studies, we account for reduced N_2O emission rates from paddy rice, which leads to substantially lower total emissions in both linear and non-linear approaches; for example, using a linear model with differentiated rice emissions lowers estimated N_2O-N from 0.86 Tg N_2O-N to 0.77 Tg N_2O-N (Table S2). Second, compared to linear emissions factors, the negative-concave model fit to an improved experimental dataset suggests reduced emissions at lower fertilizer application rates, and 78% of N fertilizer was applied at rates where linear modeled emissions exceed non-linear modeled emissions (below 135 kg ha^{-1} for flooded rice, 197 kg ha^{-1} for other crops). Third, the underestimation bias incurred by negative-concave models of N_2O emissions when used with spatially aggregated N fertilizer application data (Philibert *et al.*, 2012; Davidson & Kanter, 2014) leads these estimates to be conservatively low.

Limitations

The emissions estimates reported here exclude indirect emissions from leaching and volatilization, which comprise ~26% of total N_2O emissions associated with N application to croplands (FAO2014b). While our findings combine a non-linear model of direct N_2O emissions with crop-specific maps of N application, except for complex biogeochemical models, there is no analogous level of sophistication for estimating indirect N_2O emissions associated with N fertilizer application. Because there is greater excess N unused by the crops at higher fertilization levels (Van Groenigen *et al.*, 2010; Kim *et al.*, 2013; Shcherbak *et al.*, 2014), it is possible that indirect N_2O emissions increase in a superlinear manner as well. More sophisticated models of indirect emissions could also help to reconcile top-down and bottom-up N_2O emissions budgets (Griffis *et al.*, 2013).

Another limitation is our use of a single model for all crops (except rice), climates, and management practices. Because different crops have different N uptake characteristics, this could lead, *a priori,* to biases which would preclude comparisons of model-predicted direct N_2O emissions between crops. However, less than half of all global N applied to croplands is removed in harvested crop products (West *et al.*, 2014; Zhang et al. 2015). Thus, with approximately 50 times as much excess N as N_2O-N, differing N uptake rates do not by themselves preclude comparison among crop types**.** Moreover, climate and crop management are important controls on N_2O fluxes from soils (Stehfest & Bouwman, 2006; Berdanier & Conant, 2011; Aguilera *et al*., 2013), yet our models do not account for such variation.

Of course, our quantitative results depend on the methods applied to construct synthetic fertilizer and manure datasets, the particular form of the non-linear model, and associated parameter values (FAO, 2012; Philibert *et al.*, 2012; Shcherbak *et al.*, 2014). Despite these limitations, dataset and model uncertainties are expected to have largely local influence without altering regional differences in N application, which are well-established and may vary by several orders of magnitude (Vitousek *et al.*, 2009).

Finally, the bias associated with aggregated fertilizer data is inherent to superlinear models such as ours, and will lead this method to underestimate emissions. The coefficient of variation of the sub-regional heterogeneity in N application rate required to achieve the same increase in N_2O

emissions as using the 95th percentile model parameters is 54%. This provides one measure of how much accuracy is needed in fertilizer application rate data so that implied fertilizer rate homogeneity is not a dominant source of uncertainty. Improved monitoring and compilation of N application rates, and their variation, at a high spatial resolution will allow improved assessment of spatially-explicit N_2O emissions. We emphasize that while emissions estimates from linear models are insensitive to the degree of fertilizer data aggregation, non-linear models *require* spatially explicit, crop-specific fertilizer data (Fig S3, Table S5).

Policy implications

The non-linear, crop-specific emissions model developed and applied here indicates that increased fertilizer application is not strongly coupled to increased N_2O emissions at low N application rates, a major opportunity given increased crop production necessary to meet growing food demand (Tilman *et al.*, 2011; Foley *et al*, 2011). Other research indicates that in areas with low N application rates, small fertilizer additions generate the most substantial yield improvements; in other words, yield-response curves are also non-linear (Sanchez & Sanchez, 2010; Vermeulen *et al.*, 2012). Thus, our results suggest that Sub-Saharan Africa and parts of Eastern Europe – areas with fertilizer N application rates less than half of those in China– would realize the most favorable yield to N_2O emissions tradeoffs from additional N application. Conversely, small reductions in fertilizer application in high N input regions such as Eastern China and the Nile delta may yield substantially reduced N_2O emissions (West *et al.*, 2014b). These findings are consistent with N balance analyses indicating that more equitable allocation of N fertilizer across space generates large reductions in excess N (Mueller *et al.*, 2014). Balancing the positive benefits of N inputs for crop production with the negative impacts of excess N on ecosystem function and human health is critical for remaining within planetary boundaries with respect to N management (de Vries et al., 2013; Rockstrom et al., 2009).

Due to the underestimation bias associated with non-linear models as applied to aggregated data, we suggest the linear model remains a relevant method for estimating global N_2O emissions when it is possible to separate out fertilizer applied to irrigated rice. However, only the use of a non-linear model combined with spatially explicit and crop-specific N application rate data allows for the policy-relevant determination of how emissions factors vary spatially and between crops.

Policies encouraging increased N use in regions with low N application rates and cutbacks in N use in high application rate regions might be accompanied by promotion of field-scale efficiency practices – such as altering the rate, timing, and placement of fertilizer (Stehfest & Bouwman, 2006; Philibert *et al.*, 2012; Venterea *et al.*, 2012,) or introduction of nitrification inhibitors (Akiyama *et al., 2005.)* Such policies have well-documented environmental (Smith *et al.*, 2008; Ravishankara *et al.*, 2009; Reay *et al.*, 2012), health (Wolfe & Patz, 2002; De Klein *et al.*, 2006), and economic (Pellerin *et al.*, 2013) benefits, and researchers have explored reducing emissions via application protocols (Miller *et al*., 2010) and market mechanisms (Rosas *et al*., 2015.) Strategies aimed at mitigating N_2O emissions must consider the field-level relationships among management, emissions, and yields, and also rely on addressing socio-economic factors that are, at present, poorly understood (Zhang et al. 2015). Accurate N_2O emission models coupled with spatially-explicit, crop-specific N application data support development of GHG mitigation policies that influence farm-level outcomes.

Supporting Information is linked to the online version of the paper

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References

Aguilera, E. et al., 2013. The potential of organic fertilizers and water management to reduce N_2O emissions in Mediterranean climate cropping systems. A review. *Agriculture Ecosystems & Environment*, 164, pp. 32–52.

Akiyama H, Yagi K, Yan X (2005) Direct N_2O emissions from rice paddy fields: Summary of available data. *Global Biogeochem. Cycles*, **19**, GB1005. doi: 10.1029/2004GB002378

Akiyama, H., Yan, X. & Yagi, K., (2009). Evaluation of effectiveness of enhanced-efficiency fertilizers as mitigation options for N2O and NO emissions from agricultural soils: meta-analysis. *Global Change Biology*, 16(6), pp.1837–1846.

Alexandratos, N., Bruinsma, J., et al. *World agriculture towards 2030/2050: the 2012 revision.* (ESA Working paper Rome, FAO, 2012)

Berdanier, A.B. & Conant, R.T., 2011. Regionally differentiated estimates of cropland N2O emissions reduce uncertainty in global calculations. *Global Change Biology*, 18(3), pp.928–935.

Bouwman, A. F. Direct emission of nitrous oxide from agricultural soils. *Nutr Cycl Agroecosyst* **46,** 53–70 (1996).

Ciais P, Sabine C, Bala G et al. (2013) Carbon and other biogeochemical cycles. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, pp. 465–570.

Cambridge University Press.
Davidson, E.A. & Kanter, D., 2014. Inventories and scenarios of nitrous oxide emissions. *Environmental Research Letters*.
De Klein C, Novoa R, Ogle S et al. (2006) N2O emissions from managed soils, and CO2 emissions from lime and urea application. In: *IPCC Guidelines for National Greenhouse Gas Inventories*, Vol. Technical, pp. V4, Chapter 11–V4, Chapter 11. IPCC.
de Vries, W. et al., 2013. Assessing planetary and regional nitrogen boundaries related to food security and adverse environmental impacts. *Current Opinion in Environmental Sustainability*, 5(3-4), pp.392–402.
FAO (2014a) Food and Agriculture Organization of the United Nations http:// faostat.fao.org (Division: Statistics,Domain: Emissions-Agriculture) (accessed, November 2014)
FAO (2014b) Agriculture, Forestry and Other Land Use Emissions by Sources and Removals by Sinks: 1990–2011 Analysis. FAO Statistics Division Working Paper Series, 14/01. UN FAO, Rome, Italy. Available at: http://www.fao.org/docrep/019/i3671e/ i3671e.pdf (accessed 1 September 2014).
Flynn HC, Canals LMI, Keller E et al. (2012) Quantifying global greenhouse gas emissions from land-use change for crop production. *Global Change Biology*, **18**, 1622–1635.
Foley JA, Ramankutty N, Brauman KA et al. (2011) Solutions for a cultivated planet. *Nature*, **478**, 337–342.
Forster P (2007) IPCC 2007 Chapter 2 Radiative Forcing (ed Solomon S). 1–106.
Grassini P, Cassman KG (2012) High-yield maize with large net energy yield and small global warming intensity. PNAS, 1074-1079
Griffis TJ, Lee X, Baker JM, Russelle MP, Zhang X, Venterea R, Millet DB (2013) Reconciling the differences between top-down and bottom-up estimates of nitrous oxide emissions for the U.S. Corn Belt. *Global Biogeochem. Cycles*, **27**, 746–754.
Herrero M, Havlik P, Valin H et al. (2013) Biomass use, production, feed efficiencies, and greenhouse gas emissions from global livestock systems. *Proceedings of the National Academy of Sciences*, **110**, 20888–20893.
Hoben, J. P. *et al.* Nonlinear nitrous oxide (N_2O) response to nitrogen fertilizer in on-farm corn crops of the US Midwest. *Global Change Biol* **17,** 1140–1152 (2011)
Huke RE, Huke EH. (1997) *Rice area by type of culture: South, Southeast, and East Asia. A review and updated data base.* IRRI.
Kim D-G, Hernandez-Ramirez G, Giltrap D (2013) Linear and nonlinear dependency of direct nitrous oxide emissions on fertilizer nitrogen input: a meta-analysis. *Agriculture Ecosystems & Environment*, **168**, 53–65.
Lal, R. 2006 Encylopedia of Soil Science, CRC Press
Liu J, You L, Amini M et al. (2010) A high-resolution assessment on global nitrogen flows in cropland. *Proceedings of the National Academy of Sciences*. **107**, 8035-8040
Millar, N. et al., 2010. Nitrogen fertilizer management for nitrous oxide (N2O) mitigation in intensive corn (Maize) production: an emissions reduction protocol for US Midwest agriculture. Mitigation and adaptation strategies for global change, 15(2), pp.185–204.
Monfreda C, Monfreda C, Ramankutty N, Ramankutty N, Foley J, Foley J (2008) Farming the planet: 2. Geographic distribution of crop areas, yields, physiological types, and net primary production in the year 2000. Global Biogeochemical Cycles, 22, GB1022.
Montzka SA, Montzka SA, Dlugokencky EJ, Dlugokencky EJ, Butler JH, Butler JH (2011) Non-CO2 greenhouse gases and climate change. *Nature*, **476**, 43–50.

Mosier, A. R., Duxbury, J. M., Freney, J. R., Heinemeyer, O. & Minami, K. Assessing and Mitigating N_2O Emissions from Agricultural Soils. *Climatic change* **40,** 7–38 (1998)
Mueller ND, Gerber JS, Johnston M, Ray DK, Ramankutty N, Foley JA (2012) Closing yield gaps through nutrient and water management. *Nature*, **490**, 254–257.
Mueller ND, West PC, Gerber JS, MacDonald GK, Polasky S, Foley JA (2014) A tradeoff frontier for global nitrogen use and cereal production. **9**, 054002. doi: 10.1088/1748-9326/9/5/054002
Pellerin, S. *et al.,* How can French Agriculture contribute to reducing greenhouse gas emissions? Abatement potential and cost of ten technical measures. Summary of the study report, INRA (France) (2013)
Philibert A, Loyce C, Makowski D (2012) Quantifying uncertainties in N(2)O emission due to N fertilizer application in cultivated areas. *PLoS ONE*, **7**, e50950.
Pinheiro JC, Bates DM (2000) *Mixed-effects models in S and S-PLUS*. Springer.
Portmann FT, Siebert S, Doell P (2010) MIRCA2000-Global monthly irrigated and rainfed crop areas around the year 2000: A new high-resolution data set for agricultural and hydrological modeling. **24**, GB1011.
Ramankutty N, Evan AT, Monfreda C, Foley JA (2008) Farming the planet: 1. Geographic distribution of global agricultural lands in the year 2000. *Global Biogeochemical Cycles*, **22**.
Ravishankara AR, Daniel JS, Portmann RW (2009) Nitrous Oxide (N2O): The Dominant Ozone-Depleting Substance Emitted in the 21st Century. *Science*, **326**, 123–125.
Reay DS, Davidson EA, Smith KA, Smith P, Melillo JM, Dentener F, Crutzen PJ (2012) Global agriculture and nitrous oxide emissions. *Nature Climate Change*, **2**, 410–416.
Robinson T, Thornton P, Franceschini G et al. (2011) *Global livestock production systems.* Food and Agriculture Organization of the United Nations (FAO).
Rockström, J. et al., 2009. Planetary Boundaries: Exploring the Safe Operating Space for Humanity. *Ecology and Society*, 14(2), p.32.
Rosas, F., Babcock, B.A. & Hayes, D.J., 2015. Nitrous oxide emission reductions from cutting excessive nitrogen fertilizer applications. *Climatic change*, **132(2),** pp.353–367.
Sanchez PA, Sanchez PA (2010) Tripling crop yields in tropical Africa. *Nature Geoscience*, **3**, 299–300.
Shcherbak I, Millar N, Robertson GP (2014) Global metaanalysis of the nonlinear response of soil nitrous oxide (N2O) emissions to fertilizer nitrogen. *Proceedings of the National Academy of Sciences*. **111**, 9199–9204.
Sitch, S. et al., 2007. Indirect radiative forcing of climate change through ozone effects on the land-carbon sink. *Nature*. **448**, 791-4.
Smith P, Smith P, Martino D et al. (2008) Greenhouse gas mitigation in agriculture. *Philosophical Transactions of the Royal Society B: Biological Sciences*, **363**, 789–813.
Stehfest E, Bouwman L (2006) N2O and NO emission from agricultural fields and soils under natural vegetation: summarizing available measurement data and modeling of global annual emissions. *Nutrient Cycling in Agroecosystems*, **74**, 207–228.
Syakila A, Kroeze C (2011) The global nitrous oxide budget revisited. *Greenhouse Gas Measurement and Management*, **1**, 17–26.
Tilman D, Balzer C, Hill J, Befort BL (2011) Global food demand and the sustainable intensification of agriculture. *Proceedings of the National Academy of Sciences USA*, **108**, 20260–20264.
Tubiello FN, Salvatore M, Rossi S, Ferrara A, Fitton N, Smith P (2013) The FAOSTAT database

of greenhouse gas emissions from agriculture. *Environmental Research Letters*, **8**, 015009.
Van Groenigen JW, Velthof GL, Oenema O, van Groenigen KJ, van Kessel C (2010) Towards an agronomic assessment of N2O emissions: a case study for arable crops. *European Journal of Soil Science*, **61**, 903–913.
Venterea RT, Halvorson AD, Kitchen N et al. (2012) Challenges and opportunities for mitigating nitrous oxide emissions from fertilized cropping systems. *Frontiers in Ecology and the Environment*, **10**, 562–570.
Vergé XPC, De Kimpe C, Desjardins RL (2007) Agricultural production, greenhouse gas emissions and mitigation potential. *Agricultural and Forest Meteorology*, **142**, 255–269.
Vermeulen SJ, Campbell BM, Ingram JSI (2012) Climate Change and Food Systems. *Annual Review of Environment and Resources*, **37**, 195–222.
Vitousek PM, Naylor R, Crews T et al. (2009) Nutrient Imbalances in Agricultural Development. *Science*, **324**, 1519–1520.
West PC, Gerber JS, Engstrom PM et al. (2014) Leverage points for improving global food security and the environment. *Science*, **345**, 325–328.
Wolfe AH, Patz JA (2002) Reactive Nitrogen and Human Health: Acute and Long-term Implications. *AMBIO: A Journal of the Human Environment*, **31**, 120–125.
Zhang X, Davidson EA, Mauzerall DL, Searchinger TD, Dumas P, Shen Y (2015) Managing Nitrogen for Sustainable Development. Nature, 528, 51–59.

Figure Captions

Figure 1. Combined synthetic fertilizer and manure nitrogen (N) application rates to croplands circa year 2000. N application is depicted as harvested-area-weighted mean of N from synthetic fertilizer (Mueller *et al*., 2012) and manure (Herrero *et al*., 2013) and does not include manure application to pasture. Areas without 2000-era N fertilizer are shown in grey. Manure application to crops was calculated based on regional livestock management data from Herrero *et al.* (2013.) Arrow on right hand side of colorbar indicates saturation of values greater than 300 kg N ha^{-1}.

Figure 2. Nitrous oxide (N_2O) emissions response to application of nitrogen (N) fertilizer circa year 2000. Total direct N_2O emissions were calculated using a non-linear method that differentiates flooded rice from other crops, and are displayed as a harvested-area weighted average over 171 crops (Monfreda *et al*., 2008). Crop-specific N application rates account for both synthetic fertilizer (Mueller *et al*., 2012) and manure (Herrero *et al*., 2013). Units are kg N_2O-N per harvested hectare. Arrow on right hand side of colorbar indicates saturation of values greater than 2.5 kg N_2O-N ha^{-1}.

Figure 3. Nitrous oxide (N_2O) emissions response to application of nitrogen (N) fertilizer circa year 2000. Change in total direct N_2O emissions (kg N_2O-N emissions per harvested hectare) in response to an incremental change in N application rate (kg N per cultivated hectare, including synthetic fertilizer (Mueller *et al*., 2012) plus manure (Herrero *et al*., 2013) inputs) across harvested area for 171 global crops (Monfreda *et al*., 2008). N_2O emissions were calculated using a non-linear method that differentiates flooded rice from other crops, and

change is displayed as a harvested-area weighted average over 171 crops. Arrow on right hand side of colorbar indicates saturation of values greater than 2.5 kg N_2O-N ha^{-1}.

Figure 4. Crop-specific N application and associated direct nitrous oxide (N_2O-N) emissions estimated by a linear and non-linear model. (a) Total applied N in synthetic fertilizer and manure; **(b)** N_2O-N emissions calculated using the linear or IPCC Tier I model; **(c)** N_2O-N emissions calculated using the non-linear $NLNRR_{700}$ model developed here. Histograms are normalized such that the area of each bar is proportional to the fraction of total N applied (a) or N_2O-N emitted (b,c) The top 10 crops, ranked in each subfigure by applied N (a), and emitted N_2O-N (b,c) are shown in color, while all remaining crops are displayed in gray. "Vegetables" refers to "vegetables, not elsewhere specified" as defined by FAO.

Tables

Table 1. Emissions factor (EF) and N_2O response (kg N_2O-N emitted per kg N applied, $d(N_2O)/dN$) for the top ten crop/country combinations by total applied synthetic and manure N fertilizer (Gg, Table 1a), and the top ten crop/country combinations by applied N rate (kg ha^{-1}, Table 1b). We exclude crop/country combinations receiving <0.25% of total applied synthetic fertilizer and manure N. China flooded rice appears in both tables 1a and 1b. An extended version of this table is presented as Table S10 in the Supplementary on-line dataset.
Table 1a

Country	Crop	Total N Application	Mean N Application rate	Linear EF	Non-linear EF	$d(N_2O$-$N)/dN$
		Gg	kg ha^{-1}	%	%	0.01 kg/kg
China	**rice, flooded**	5407	183	0.31	0.36	0.56
United States	**maize**	4665	159	1.00	0.92	1.30
China	**wheat**	4517	171	1.00	0.93	1.35
China	**maize**	4321	176	1.00	0.94	1.39
India	**rice, flooded**	3283	84	0.31	0.28	0.35
India	**wheat**	3005	114	1.00	0.80	1.01
United States	**wheat**	1770	79	1.00	0.74	0.87
China	**rapeseed**	1139	160	1.00	0.90	1.29
Indonesia	**rice, flooded**	997	98	0.31	0.28	0.36
Pakistan	**wheat**	933	114	1.00	0.79	1.01

Table 1b.

Country	Crop	Total N Application	Mean N Application rate	Linear EF	Non-linear EF	d(N_2O-N)/dN
		Gg	kg ha^{-1}	%	%	0.01 kg/ kg
Egypt	**maize**	290	355	1.00	1.59	3.53
Egypt	**wheat**	259	258	1.00	1.17	2.09
Italy	**maize**	239	228	1.00	1.14	1.88
France	**maize**	388	220	1.00	1.14	1.84
Pakistan	**sugarcane**	216	210	1.00	1.03	1.64
Pakistan	**cotton**	576	194	1.00	0.98	1.51
Germany	**wheat**	456	184	1.00	1.04	1.53
China	**rice, flooded**	5407	183	0.31	0.36	0.56
China	**cotton**	792	180	1.00	0.94	1.41
China	**maize**	4321	176	1.00	0.94	1.39

Table 2. N_2O emissions by country for top 25 countries in terms of total N application. "Linear EF" is the emissions factor (EF) calculated using the IPCC Tier I linear method [0.31% for flooded rice, 1% for all other crops], "Non-linear EF" is total direct EF calculated using the nonlinear $NLNRR_{700}$ model developed in this article. $d(N_2O)/dN$ is the incremental change in N_2O emission associated with an incremental change in N application on all harvested area in units of kg N_2O-N/100 kg N. An extended version of this table is presented as Table S7 in the Supplementary on-line dataset.

Country	Total N Application	Mean N Application rate	Linear EF	Non-linear EF	$d(N_2O$-$N)/dN$
	Gg	kg ha^{-1}	%	%	0.01 kg/kg
World	86329	68	0.89	0.77	0.80
China	25627	158	0.85	0.80	1.14
India	12031	65	0.81	0.62	0.70
United States	10852	83	0.99	0.84	0.92
Pakistan	2414	122	0.96	0.89	1.07
Indonesia	2142	69	0.68	0.56	0.65
France	1937	109	1.00	0.95	1.10
Brazil	1872	38	0.96	0.70	0.69
Canada	1703	49	1.00	0.74	0.76
Germany	1579	128	1.00	1.02	1.24
Turkey	1405	69	1.00	0.75	0.83
Mexico	1239	73	0.99	0.72	0.83
Vietnam	1232	109	0.60	0.59	0.70
Spain	1189	81	0.99	0.79	0.89
Russian Federation	1128	14	1.00	0.62	0.63
Egypt	1104	199	0.93	1.34	1.94
Bangladesh	1088	75	0.48	0.39	0.47
Thailand	1041	58	0.62	0.53	0.54
Australia	975	42	1.00	0.70	0.72
Poland	949	78	1.00	0.77	0.87
Italy	872	94	0.97	0.85	0.97
Iran Islamic Republic	803	64	0.96	0.70	0.78
United Kingdom	716	124	1.00	0.92	1.15
Uzbekistan	603	126	0.98	0.82	1.07
Ukraine	580	21	1.00	0.62	0.65
Philippines	539	43	0.78	0.58	0.60

Table 3. N_2O emissions for 30 major crops. "Linear EF" is the emissions factor (EF) calculated using the IPCC linear method [0.31% for flooded rice, 1% for all other crops], "Non-linear EF" is total direct EF calculated using the nonlinear $NLNRR_{700}$ model developed in this article. $d(N_2O)/dN$ is the incremental change in N_2O emission associated with an incremental change in N application on all harvested area in units of kg N_2O-N/100 kg N. An extended version of this table is presented as Table S9 in the Supplementary on-line dataset.

Crop	Total N Application	Mean N App rate	Linear EF	Non-linear EF	$d(N_2O$-$N)/dN$
	Gg	kg ha^{-1}	%	%	0.01 kg/kg
wheat	16784	81	1.00	0.82	0.90
maize	13648	101	1.00	0.91	1.03
flooded rice	13585	97	0.31	0.32	0.38
cotton	3004	99	1.00	0.85	0.99
barley	2977	55	1.00	0.79	0.80
rapeseed	2644	108	1.00	0.87	1.04
soybean	2162	29	1.00	0.66	0.68
potato	1885	98	1.00	0.94	1.04
vegetable (other)	1834	126	1.00	0.93	1.16
sugarcane	1822	93	1.00	0.81	0.95
mixedgrass	1813	28	1.00	0.83	0.70
forage (other)	1192	67	1.00	0.77	0.83
sweetpotato	1039	116	1.00	0.90	1.09
sorghum	993	26	1.00	0.71	0.67
groundnut	814	37	1.00	0.72	0.71
non-flooded rice	789	73	1.00	0.76	0.85
bean	764	31	1.00	0.66	0.68
sunflower	729	36	1.00	0.74	0.71
coffee	718	72	1.00	0.80	0.86
maizefor	716	49	1.00	0.96	0.83
apple	674	126	1.00	0.97	1.18
sugarbeet	673	111	1.00	0.94	1.10
oats	617	47	1.00	0.74	0.75
alfalfa	580	29	1.00	0.68	0.68
banana	593	149	1.00	1.43	1.70
tomato	558	152	1.00	1.17	1.49
oilpalm	536	56	1.00	0.73	0.78
grape	487	70	1.00	0.80	0.85
mango	466	140	1.00	0.92	1.21
watermelon	458	156	1.00	1.04	1.39

Supporting Information Captions

Figure S1. Total direct N_2O-N emissions due to synthetic N fertilizer and manure application to crops calculated from the linear model

Figure S2. Difference in N2O-N emissions calculated with the IPCC Tier I linear model (De Klein et al., 2006) compared to emissions estimated from the non-linear model developed here.

Figure S3. Country-specific synthetic N and manure fertilizer application and associated direct N2O emissions estimated by linear and non-linear models.

Figure S4. Fitted median responses of N2O emissions for N application rates from 0 to 700 kg N/ha.

Figure S5. Model residuals as a function of applied N rate. . Residuals were calculated with the model described in Table S1 using the site-specific parameters.

Figure S6. Comparison of mean model and uncertainty for Shcherbak et al. (2014, blue), and the nonlinear model developed in this paper.

Figure S7. Uncertainty estimates of N2O emissions associated with empirical N application rates circa 2000.

Figure S8. Mean N2O emissions estimated using the NLNRR models derived from varying experimental datasets, and excluding flooded rice.

Table S1. Estimated model parameter values and variance-covariance matrix.

Table S2. Global direct N2O emissions under various modeling assumptions.

Table S3. N2O emissions by world region.

Table S4. N2O emissions by top 10 countries, administrative units, and crops.

Table S5. Summary of studies estimating direct N2O emissions in response to application of synthetic N fertilizer to crops.

Supporting Data Tables (in spreadsheet form)

Table S6. Percentage of manure N estimated as applied to croplands across regions, livestock systems, and livestock types.

Table S7. Summary direct N2O emissions statistics by country.

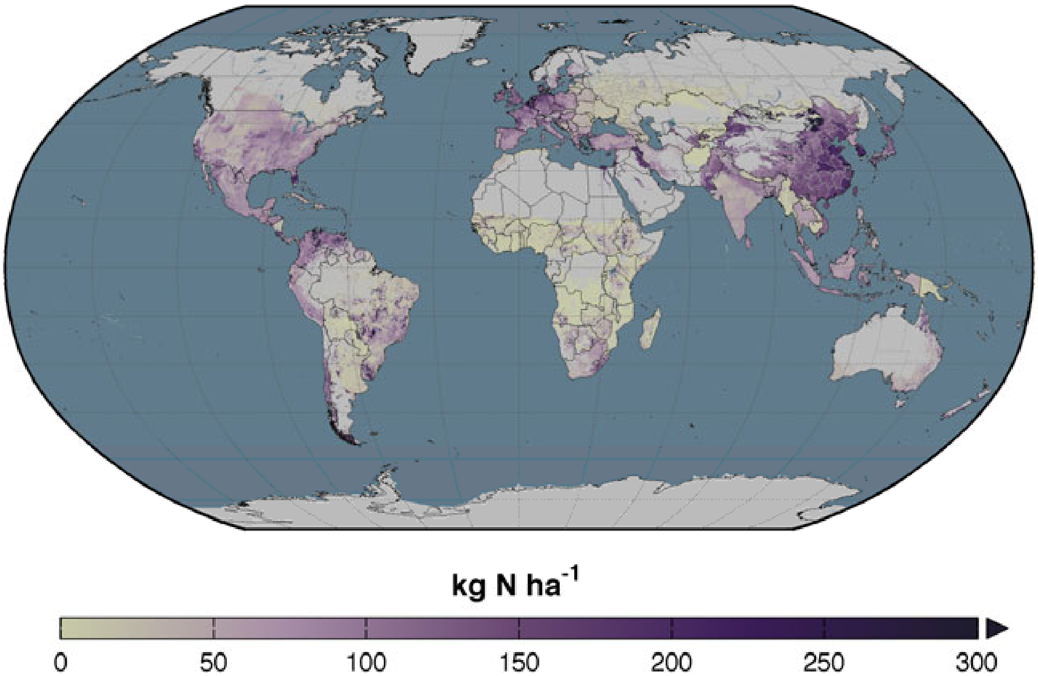
Table S8. Summary direct N2O emissions statistics by continent grouping

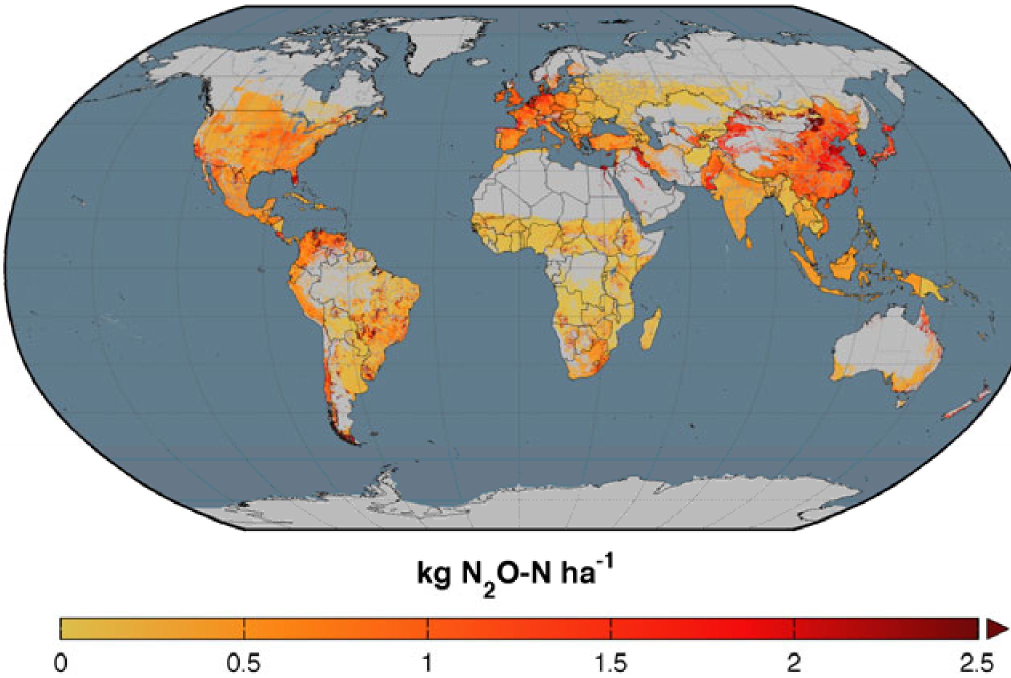
Table S9. Summary direct N2O emissions statistics by major crop

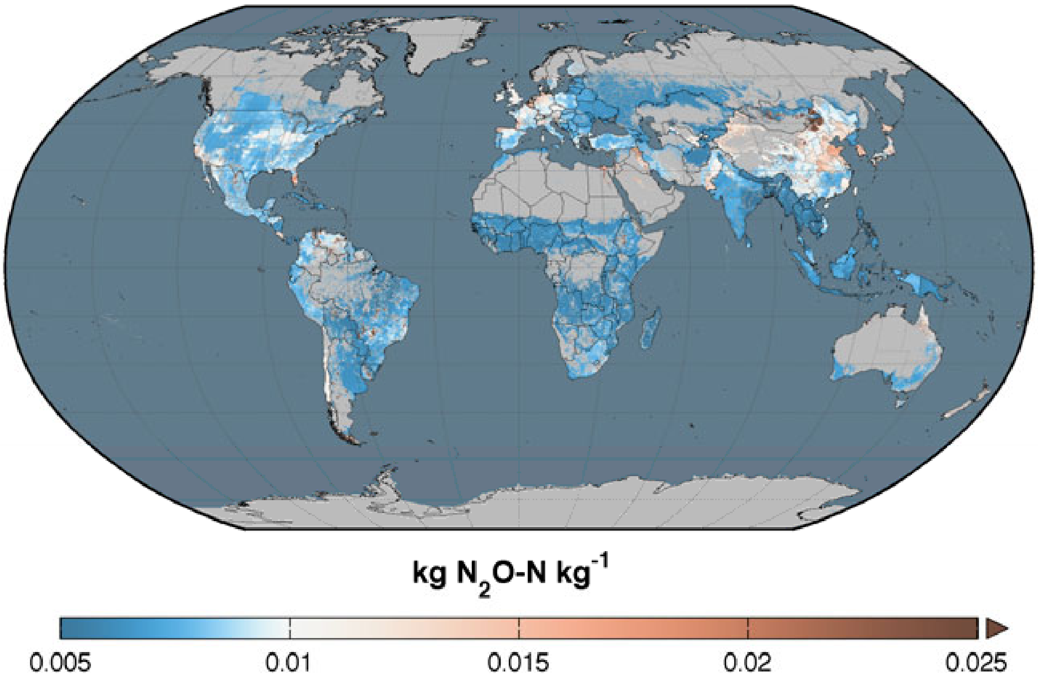
Table S10. Summary direct emissions statistics by state for the top three consumers of N fertilizer (China, US, India).

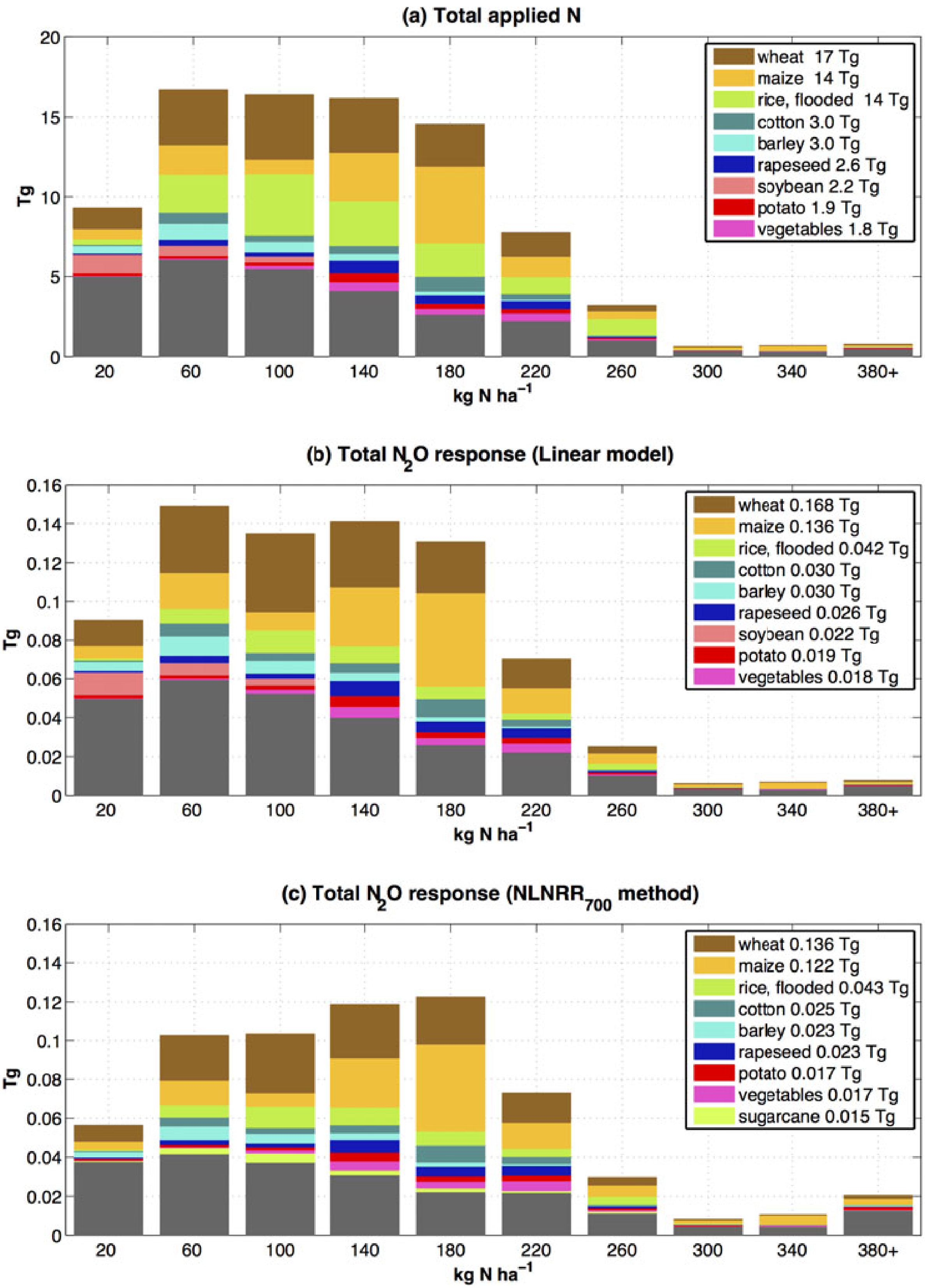
Table S11– Summary emissions statistics by crop/country combination for the combinations which account for more than 0.25% of all N fertilizer application to crops.

Table S12 - N2O Emissions calculated with NLNRR700 model


kg N ha^{-1}
0
50
100
150
200
250
300


kg N2O-N ha-1
0
0.5
1
1.5
2
2.5


kg N2O-N kg-1
0.005
0.01
0.015
0.02
0.025


(a) Total applied N
Tg
kg N ha^-1
20
60
100
140
180
220
260
300
340
380+
wheat 17 Tg
maize 14 Tg
rice, flooded 14 Tg
cotton 3.0 Tg
barley 3.0 Tg
rapeseed 2.6 Tg
soybean 2.2 Tg
potato 1.9 Tg
vegetables 1.8 Tg
(b) Total N2O response (Linear model)
wheat 0.168 Tg
maize 0.136 Tg
rice, flooded 0.042 Tg
cotton 0.030 Tg
barley 0.030 Tg
rapeseed 0.026 Tg
soybean 0.022 Tg
potato 0.019 Tg
vegetables 0.018 Tg
(c) Total N2O response (NLNRR700 method)
wheat 0.136 Tg
maize 0.122 Tg
rice, flooded 0.043 Tg
cotton 0.025 Tg
barley 0.023 Tg
rapeseed 0.023 Tg
potato 0.017 Tg
vegetables 0.017 Tg
sugarcane 0.015 Tg