

## ***Interactive comment on “Estimation of pre-industrial nitrous oxide emissions from the land biosphere” by Rongting Xu et al.***

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Dear reviewer #1,

Thanks very much for the precious comments and suggestions. All the comments and suggestions are addressed in the revised manuscript.

1. With respect to constraints on the overall PI global emissions of N<sub>2</sub>O, I have more confidence in the top-down approach using atmospheric concentrations and lifetimes of N<sub>2</sub>O, than the bottom up simulations of a highly parameterized process model. The most recent top-down estimate (Prather et al., 2015) is cited in passing by the authors, but the estimates are not included in the present manuscript. The estimates from the IPCC AR4 and from Davidson & Kanter (2014), mentioned in lines 53-54, were based largely on the 2012 paper by Prather et al., but their 2015 paper provides an

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important update on lifetime estimates and resulting PI emission estimates. They now recommend using lifetimes of 123 years for PI and 116 years for the present (+/- 9 years), and from those lifetime estimates, they derive a new PI emission estimate of 10.5 Tg/yr. Fortunately, this is very close to other estimates, including the one from this study. Nevertheless, it should be specifically cited.

Response: We have cited the recent study by Prather et al. (2015) in the introduction and discussion sections of the revised manuscript.

Line 53-55: Prather et al. (2015) provided an estimate of the pre-industrial emissions (total natural emission: 10.5 Tg N yr<sup>-1</sup>) based on the most recent study with a corrected lifetime of 116 years.

Line 260-267: “Top-down” methodology used to estimate N<sub>2</sub>O emissions is based on atmospheric measurements and an inversion model (Thompson et al. 2014). Prather et al. (2012) provided an estimate of 9.1±1.0 Tg N yr<sup>-1</sup> of natural emission in the pre-industrial era using observed pre-industrial abundances of 270 ppb and model estimates of lifetime decreased from 142 years in the pre-industrial era to 131±10 years in the present-day. Later, Prather et al. (2015) re-evaluated N<sub>2</sub>O lifetime based on Microwave Limb Sounder satellite measurements of stratospheric, which was consistent with modeled values in the present-day. The lifetime in the pre-industrial era and present-day was estimated as 123 and 116±9 years, respectively. The current lifetime increases the pre-industrial natural emission from 9.1±1.0 to 10.5 Tg N yr<sup>-1</sup>.

2. The point that the lifetime has probably decreased since PI times should be discussed. As far as I can tell, a varying lifetime cannot be incorporated into the one-box model (line 171) used by the authors. Perhaps the resulting global estimate is not terribly sensitive to this change, but that should be evaluated and discussed.

Response: In the revised version, we have removed the one-box model validation. In addition, we added the discussion of the decreased lifetime since PI times, which was also mentioned in the response to question #1 (Line 260-267).

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3. I fail to see how the analysis presented in Figure 7 and Table 1 provides additional confidence in the summed global estimate from this study. I can see the value of a sensitivity analysis of initial PI atmospheric concentrations and lifetimes, which Prather's papers have already done and for which they could be cited. In contrast, the analysis in Fig. 7 and Table 1 is clouded by the unclear source of annual emissions over the simulated time period and the validity of those assumptions. The text (lines 182-185) suggests that model output was used for annual emission estimates: "The mean with 95% confidence intervals, the maximum, and minimum values of estimates from DLEM simulations were applied as initial emissions to calculate the atmospheric N<sub>2</sub>O concentration in 2006 as shown in Table 1 (Scenarios 1-4 and baseline), as well as concentration changes from 1860 to 2006, as shown in Figure 7." However, the Fig. 7 captions indicates that the "net additions of anthropogenic N<sub>2</sub>O emission amount in different years were listed in Syakila and Kroeze, 2011." I don't understand which was used to estimate annual increments of N<sub>2</sub>O concentration in Fig 7 – was it model output, as indicated on lines 182-185, or was it the net additions estimated by S&K as indicated in the figure caption? Both have problems. S&K estimated fairly substantial N<sub>2</sub>O emissions from agriculture during the late 19th and early 20th centuries, but they also estimated a rather large decrease in natural emissions compared to 1500 (which are very difficult to estimate, see my further comments below), so their estimate of the net change relative to 1500 was small for this time period. However, the starting point for the present study is 1860. Therefore, it is incorrect to subtract this decline in natural emissions that preceded 1860 from the growth in anthropogenic emissions since 1860. S&K did this to show changes since their starting point of 1500, but using their "net additions" column without accounting for a different starting point in the present study introduces a significant bias. It is the net change relative to 1860 that is important for the present study, so the "net additions" estimated by S&K should be recalculated relative to 1860 if they are to be used in the analysis for Table 1 and Fig. 7. I showed in my 2009 paper, and Smith et al. (2012) have affirmed, that atmospheric N<sub>2</sub>O began rising significantly many decades before fertilizer use became common in the 1950s,

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and so the "net additions" to the atmosphere must have been larger than those estimated by S&K relative to 1500, although they may be similar if they were corrected to be relative to 1860. We speculate that this increase in emissions between 1860 and 1950 was due to mineralization of soil N as agriculture expanded into regions of previously untilled soils, thus mobilizing N for rapid cycling, including a fraction lost at N<sub>2</sub>O. I also suspect that the current DLEM may not include effects of soil mining when virgin soil is first tilled, so if Table 1 is based on DLEM simulations, as indicated in the text on lines 182-185, then I suspect emissions from 1860 to 1950 were underestimated, which would affect the slope of the trend line later in the analysis as well. I realize that the point of Figure 7 is not the accuracy of the simulated trend line, but rather the end point, but if the trend line agrees so poorly with the observations, then one has to question the validity of the model and the input data, which calls into question the reliability of the end point analysis. I believe that Fig. 7 and Table 1 could be replaced with citations of the sensitivity analyses done by Prather et al. (2012, 2015), but if the authors persist in wanting to include their own analysis, I would suggest that they utilize another source of "net addition" emissions than those of S&K relative to 1500.

Response: According to your suggestions, we have removed the one-box model validation. Instead, we cited the work done by Prather et al. (2012, 2015) and compared our results with theirs in the section 3.2.

Line 268-277: Natural sources for N<sub>2</sub>O include soil under natural vegetation, oceans, and atmospheric chemistry (Ciais et al., 2014). The emission from atmospheric chemistry was estimated as 0.6 with an uncertainty range of 0.3-1.2 Tg N yr<sup>-1</sup>. Syakila and Kroeze (2011) estimated global natural emissions from oceans as 3.5 Tg N yr<sup>-1</sup>. Oceanic emission was estimated as 3.8 with an uncertainty range of 1.8-5.8 Tg N yr<sup>-1</sup> in the IPCC AR4. However, the uncertainty range became larger (1.8-9.4 Tg N yr<sup>-1</sup>) in the IPCC AR5. In our study, the simulated N<sub>2</sub>O emission was from agricultural and natural soils. The natural emission was estimated as 5.78 (4.4-7.72) Tg N yr<sup>-1</sup>. Combining the atmospheric chemistry and the ocean emissions in the IPCC

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AR5 with the natural emissions from our study, the global total natural N<sub>2</sub>O emissions were 10.18 (6.5-18.32) Tg N yr<sup>-1</sup>. The large uncertainty range was attributed to the uncertainty from oceanic emission, atmospheric chemistry emission, and our estimation. The estimated global total amount (10.18 Tg N yr<sup>-1</sup>) in this study was comparable to the estimate (10.5 Tg N yr<sup>-1</sup>) by Prather et al. (2015) using the top-down approach.

4. The change in “natural” emissions before and after 1860 should be discussed. As I noted above, S&K deduce a substantial decline in natural emissions from 1500 to 1850. Similarly, I included a significant change in non-agricultural soil emissions due to tropical deforestation, which began growing rapidly in the late 20th century (Davidson 2009). Whether pre-1850 or post-1950, these changes in natural soil emissions are difficult to estimate, but the uncertainties that they represent should be considered, and biases resulting from how they are or are not included should be considered.

Response: We agree that different factors caused different variation patterns in N<sub>2</sub>O fluxes before and after 1860. We did not consider the pre-1850 natural emission change because we assumed emission in 1860 can represent the pre-industrial level although it has declined from 1500 to 1850. Our estimation from the process-based model can capture the N<sub>2</sub>O emission due to land use change in the late 20th century, but it is beyond the scope of this paper. Since pre-industrial N<sub>2</sub>O emission is not always stable and remains a large uncertainty, our estimation can only go back to 1860 and represent N<sub>2</sub>O level before intensive human disturbance.

5. While the top-down approach of Prather et al. (2012, 2015) and the one box model used in the present study help constrain total PI emissions, the soil emission estimate must still be made by difference between total emissions and oceanic emissions. While the AR5 estimate of 3.8 Tg N<sub>2</sub>O-N/yr (range: 1.8 - 9.4; Ciais et al., 2013) is widely cited for emissions from the oceans, it is highly uncertain, so simply subtracting 3.8 (or 3.5 – 4.5 as in Table 1 of the present manuscript) from a total PI source estimate of about 11 Tg N<sub>2</sub>O-N/yr (+/-1) doesn't really narrow the confidence estimate of the PI terrestrial source a great deal. Indeed, I just discovered a curious inconsistency between the AR5

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best estimate of 3.8 with a review paper by Voss et al. (2013), which cites that same 3.8 value for N<sub>2</sub>O emissions from the open ocean, but then adds another 1.7 Tg N<sub>2</sub>O-N/yr for emissions from the continental shelf regions. I don't know if the AR5 review of the literature failed to adequately represent continental shelf regions or if Voss et al. might be double accounting. If Voss et al. are correct, the AR5 estimate of oceanic emissions may be biased toward the low end, which would mean that the terrestrial PI source may more likely be in the range of 5 Tg N<sub>2</sub>O-N/yr or less. In any case, this highlights how uncertain the oceanic estimate is, which means we have to have similar uncertainty in the estimate of the PI terrestrial source. The narrow range of uncertainty in the present study's PI terrestrial source (6.03-6.36 Tg N<sub>2</sub>O-N/yr) reported on line 331 is unrealistically small.

Response: Yes, the soil emission estimation must still be made by difference between total emissions and oceanic emissions regardless of methodology (top-down or bottom-up). In the IPCC AR5, the average oceanic emission is 3.8 Tg N yr<sup>-1</sup>, with a larger uncertainty range compared with the estimate in the AR4. The estimate from Voss et al. (2013) indicated that oceanic emission was 1.7 Tg N yr<sup>-1</sup> more than the average in the AR5. It is because they considered the emissions (1.7 Tg N yr<sup>-1</sup>) from “rivers, estuaries, and coastal zones” as the marine emissions, as written in Table 7.7 of the IPCC AR4 Chapter 7. Thus, the average estimation in AR5 is still trustable. In this study, to compare with the results (10.5 Tg N yr<sup>-1</sup>) in Prather et al. (2015), we need to sum our estimate and other natural emissions. The global total natural N<sub>2</sub>O emissions were 10.18 (6.5-18.32) Tg N yr<sup>-1</sup> in the preindustrial era.

The small uncertainty range shown in the upper panel of Fig. 5 was the 95% confidence interval of the mean estimate, as explained in the manuscript. The uncertainty range of pre-industrial N<sub>2</sub>O emissions was present using the minimum and maximum estimate (4.76-8.13 Tg N yr<sup>-1</sup>) in this study, which was consistent with other studies, such as the reported estimates in the IPCC AR5. Here, the Bootstrap resampling method was used to define the uncertainty bounds of global mean N<sub>2</sub>O emission (6.20 Tg N yr<sup>-1</sup>)

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(shown in line 216-219 of previous manuscript). It was used to verify the stability of the LHS approach. The 95% confidence intervals (6.03-6.36 Tg N yr<sup>-1</sup>) of the mean did not represent the uncertainty range for pre-industrial N<sub>2</sub>O emission in this study. Thus, we will not report this narrow range in the revised manuscript to avoid the confusion.

6. The authors have misunderstood the emission estimates from my 2009 paper, which they incorrectly describe on lines 299-301: "However, the indirect emissions from the riverine induced by the leaching and runoff of manure applications in agro-ecosystems, legume crop N fixation, and human sewage discharging have not been addressed in Davidson (2009)." On the contrary, I derived emissions factors from a statistical model that was constrained by the historical record of atmospheric concentrations and fertilizer and manure use, so the emission factors derived from that analysis necessarily included all of the emissions, direct and indirect, that could be statistically correlated with historical fertilizer and manure use ("The sources attributed to fertilizers and manures include indirect emissions from downwind and downstream ecosystems, including human sewage." Davidson, 2009). Therefore, it is incorrect for the authors to calculate an additional indirect source (line 305) using IPCC default factors to add onto the estimate that they took from my paper that they misunderstood to be only direct emissions. They could either use an unmodified estimate from my paper or they could derive a new one, based on IPCC default values for both direct and indirect emissions based on estimates of BNF, fertilizer-N, and manure-N for 1860. Furthermore, note that the 0.42 Tg N<sub>2</sub>O-N/yr that they extracted from my paper for 1860 was for anthropogenic biological emissions (i.e., soils) only, and that there were also some other anthropogenic emissions at that time, such as biomass burning (see SI for Davidson 2009).

Response: We are sorry for the misunderstanding of this paper. We deleted this sentence and recalculated the overall N<sub>2</sub>O emissions from this paper. In Davidson (2009), two approaches (top-down and bottom-up) had been applied to estimate the anthropogenic biogenic N<sub>2</sub>O emissions in 1860. The estimates from top-down and bottom-up were 0.42 and 0.54 Tg N yr<sup>-1</sup>, respectively. Thus, the final number we used in this

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study was 0.5 with an uncertainty range of 0.4-0.6 Tg N yr<sup>-1</sup>. In addition, N<sub>2</sub>O from the biomass burning was assumed to be 0.2 Tg N yr<sup>-1</sup> in 1860 in Davidson (2009). In sum, the total anthropogenic N<sub>2</sub>O emission in 1860 was estimated as 0.7 (0.6-0.8) Tg N yr<sup>-1</sup> in Davidson (2009). We added below content in the revised version:

Line 321-323: The pre-industrial anthropogenic N<sub>2</sub>O sources in his study included biomass burning, agriculture activities (e.g., manure and fertilizer application, and the cultivation of legume) and human sewage, the sum of which was 0.7(0.6-0.8) Tg N yr<sup>-1</sup> (Davidson, 2009).

7. The authors should also acknowledge that there were anthropogenic effects on the N<sub>2</sub>O budget before 1860, so the 1860 fluxes don't necessarily represent only "natural" emissions. This includes some N<sub>2</sub>O from agricultural expansion that mined soil N and also added BNF, some biomass burning, a tiny amount of industrial and transportation sector emissions, and possibly a loss of emissions from degraded natural soils that had been plowed for centuries or millennia, some of which were highly eroded.

Response: Yes, the 1860 fluxes don't necessarily represent only "natural" emissions. In our study, when we mentioned "natural" emissions, we excluded the emissions from cropland soils. Our study has only addressed the anthropogenic emissions from cropland expansion and manure application, but we are unable to simulate the anthropogenic emissions from biomass burning and other sectors. As described in the response 6, we have added the discussion on the pre-industrial anthropogenic N<sub>2</sub>O emission in this manuscript (Section 3.4 Line 321-323).

See the section 3.4, Line 311-334:

3.4 The N<sub>2</sub>O budget in the pre-industrial era The observed N<sub>2</sub>O concentration is the result of dynamic production and consumption processes in soils as soils act as sources or sinks of N<sub>2</sub>O emissions through denitrification and nitrification (Chapuis-Lardy et al., 2007). There was a slight increase of atmospheric N<sub>2</sub>O concentration during 1750-1860 according to the ice core records, but showed a rapid increase from 1860 to

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present (Ciais et al., 2014). Nature sources of N<sub>2</sub>O emissions have been discussed in section 3.2 & 3.3. Previous studies found that there were some anthropogenic N<sub>2</sub>O emissions along with the natural sources in the pre-industrial era (Davidson, 2009; Syakila and Kroeze, 2011). Syakila and Kroeze (2011) found anthropogenic N<sub>2</sub>O emission began since 1500 because of the biomass burning and agriculture. The total anthropogenic N<sub>2</sub>O emission in their study was estimated as 1.1 Tg N in 1850. In addition, Davidson (2009) derived a time-course analysis of sources and sinks of atmospheric N<sub>2</sub>O since 1860. The pre-industrial anthropogenic N<sub>2</sub>O sources in his study included biomass burning, agriculture (e.g. manure and fertilizer application, and the cultivation of legume) and human sewage, the sum of which was 0.7 (0.6-0.8) Tg N yr<sup>-1</sup> (Davidson, 2009). Thus, anthropogenic N<sub>2</sub>O emission has already existed in 1860, but in a small magnitude as compared with the contemporary amount.

Davidson (2009) mentioned that there was possibly a certain amount of N<sub>2</sub>O loss in the pre-industrial period through atmospheric sink and the reduced emission from tropical deforestation. He estimated the anthropogenic sink as 0.26 Tg N in 1860. In addition, the deforestation of tropical forest might have caused a loss of N<sub>2</sub>O emissions in 1860, which was estimated as 0.03 Tg N (Davidson, 2009). However, studies have shown that the conversion of forest to pasture and cropland could increase or have no effect on N<sub>2</sub>O emissions because the effects depended on disturbance intensity of human activities on soil conditions (van Lent et al., 2015). For instance, N<sub>2</sub>O emissions tended to increase during the first 5-10 years after conversion and thereafter might decrease to average upland forest or low canopy forest levels in the non-fertilized croplands and pastures. In contrast, emissions were at a high level during and after fertilization in fertilized croplands (van Lent et al., 2015). Thus, more work is needed to study how forest degradation affects N<sub>2</sub>O fluxes (Mertz et al., 2012).

8. Although my comments above all focus on the PI global total estimate, perhaps the more important contribution of this manuscript is the simulated spatial distribution of those PI soil emissions. It is not surprising that the model simulates the majority of

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the soil emissions coming from tropical forest soils. That is also true today for nonagricultural soils. There are a few curious details that jump out at me from the map (Fig. 4). Why are emissions from the Amazon Basin and SE Asia so much lower than from the Congo Basin? Other models that I am aware of don't show that difference (e.g., Zhuang et al., 2012; Stehfest & Bouwman, 2006; Potter et al., 1996). Which of the datapoints in Fig. 3 are from tropical forests and which continents are they from? Is there validation support for the Congo having much higher emissions than the Amazon or SE Asia? More discussion would be helpful to interpret the variation shown in this map, such as where agriculture was or had been, where wetlands are, and where there are hot spots other than tropical forests. For example, I see a bunch of small red spots that appear to be near the Andes range, which puzzles me, but perhaps there is a good explanation. Ditto for why Northeastern Brazil, which is generally rather xeric, shows up as a hot spot. Also curious are the hot spots in southwestern China and the southeast coast of Australia.

(1) Why are emissions from the Amazon Basin and SE Asia so much lower than from the Congo Basin? Other models that I am aware of don't show that difference (e.g., Zhuang et al., 2012; Stehfest & Bouwman, 2006; Potter et al., 1996).

Response: There are three major explanations for the spatial pattern differences among various studies. Firstly, the vegetation map in our study includes at most five biome types (at most four natural vegetation types and one crop type) within each grid cell. For example, in the Congo and Amazon Basin, the major natural vegetation type is Tropical Broadleaf Evergreen Forest (TrBEF). Many other models only include one vegetation type within each grid cell. This difference can cause large difference in spatial distribution of N<sub>2</sub>O emissions between our results and other model simulations.

Secondly, DLEM simulates both soil nitrogen transformation and nitrogen export or leaching into riverine ecosystems (see section 2.1). Many other models don't simulate nitrogen leaching or export. In our simulation, we found that high rainfall (especially heavy rainfall events) can cause a large amount of available nitrogen exports to riverine

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ecosystems and thus reduce soil available N and N<sub>2</sub>O emissions in these grid cells.

The third cause may be the difference in model driving data. Stehfest & Bouwman (2006) used the mean annual precipitation and annual temperature developed by New et al. (1999) during 1961-1990. In Zhuang et al., (2012), they used the monthly data from the original literature and a historical climate database from the Climate Research Unit during 1961-2002 (Mitchell and Jones, 2005). While, DLEM used the long-term mean climate datasets (daily CRUNCEP climate data) from 1901-1930 to represent the initial climate state in 1860.

In the two studies mentioned above, they stated that soil and climate characteristics are major factors that affect N<sub>2</sub>O emissions. Unfortunately, neither of them showed the spatial distribution of precipitation or temperature, and the correlation between the climate and N<sub>2</sub>O emissions. Moreover, through comparing the spatial N<sub>2</sub>O emission map from those studies, we found that the distributions and magnitudes of emissions in the Congo, Amazon Basin, and Southeast Asia also differed significantly. The spatial patterns of annual precipitation and temperature in this study are shown in Fig. S2. The Congo, Amazon Basin, and Southeast Asia are located in the tropics. The three regions have similar annual temperature (Fig. S2a), but have significantly different annual precipitation (Fig. S2b). In some areas in Amazon Basin and Southeast Asia, the annual precipitation was even higher than 3000 mm. In contrast, the annual precipitation in the Congo varied from 1300 to 2000 mm. In the DLEM, we explicitly considered the daily N leaching and runoff. Because of the heavy rainfall in the Amazon Basin and Southeast Asia, more N might be leached from the soil during the wet season, which could cause the lower annual N<sub>2</sub>O emissions. In addition, both denitrification and nitrification are highly affected by the soil water content. As field experiments revealed N<sub>2</sub>O or NO could be reduced into N<sub>2</sub> when soils are in saturation (Davidson et al., 2000), DLEM also represent the formation and proportion of N<sub>2</sub>O in total nitrogen oxides, considering the effect of soil moisture change. Thus, excessive soil water content during the wet season in Amazon Basin and Southeast Asia might reduce the activities

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of microbes, thus causing smaller amount of N<sub>2</sub>O emission.

(2) Which of the datapoints in Fig. 3 are from tropical forests and which continents are they from?

Response: Firstly, we are sorry for the mistake in this Figure. The x-axis should be “observed N<sub>2</sub>O emission” rather than “simulated N<sub>2</sub>O emission”. In the new version, we redraw this figure. In addition, we used different symbols to mark all sites in Fig. 3 to make it clearly show the locations of all 20 sites. The information of each site can be found in the supplementary material (Table S1).

(3) Is there validation support for the Congo having much higher emissions than the Amazon or SE Asia?

Response: There were only two sites in the validation from southeast Asia (site 14) and the east coast of Austria (site 10). Unfortunately, there was no available validation to support the arguments that the Congo has much higher emissions than the Amazon Basin or Southeast Asia. However, we did find some measurements in Kim et al. (2016), which could support our estimates in the central Africa. In their study, they calculated the average N<sub>2</sub>O emission from ten observations in the Congo Basin, which was  $4.2 \pm 1.5$  kg N ha<sup>-1</sup> yr<sup>-1</sup> and close to our estimates in this region.

(4) More discussion would be helpful to interpret the variation shown in this map, such as where agriculture was or had been, where wetlands are, and where there are hot spots other than tropical forests.

Response: The spatial distributions of cropland and wetlands have been provided in the manuscript. Meanwhile, the emission from pre-industrial cropland was discussed in line 227-236. It is hardly to make sure the certain crop types 150 years ago. Thus, N<sub>2</sub>O emission from cropland remained quite uncertain. For the N<sub>2</sub>O emission from wetlands and peatlands, we have discussed in line 248-258. We did not include the estimate of pre-industrial wetlands or peatlands because of the uncertainty of wetland

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area and distribution, but it will be included in the future study. The results in the DLEM simulation indicated that where natural vegetation was, specifically the tropical forest, were hot spots for N<sub>2</sub>O emissions in the pre-industrial period. Some scattered hot spots the reviewer mentioned were also from the tropics as described below.

(5) I see a bunch of small red spots that appear to be near the Andes range, which puzzles me, but perhaps there is a good explanation. Ditto for why Northeastern Brazil, which is generally rather xeric, shows up as a hot spot. Also curious are the hot spots in southwestern China and the southeast coast of Australia.

Response: We have noticed those “hot spots”. Near the Andes range and in southwestern China, those mountains have higher altitudes and smaller amount of annual precipitation compared with the adjacent basins. Less N leaching happened in those regions. Meanwhile, both regions in the tropics that are dominant with TrBEF. Thus, it is possible that N<sub>2</sub>O emission was higher in this circumstance. In the Northeastern Brazil and the Southeast coast of Australia, both regions are along the coast. Both regions were not xeric according to the annual precipitation data used in this study. In the Northeastern Brazil, the dominant vegetation type is still TrBEF. Similarly, less N leaching and proper soil water content might cause higher amount of N<sub>2</sub>O emissions. In the east coast of Australia, anthropogenic activities contributed a large amount of N deposition in 1860 compared to other regions of Australia. Several grids with higher emissions were dominant with Temperate Broadleaf Evergreen Forest (TBEF). Meanwhile, the precipitation was generally higher along the Australian coast. Thus, higher N deposition with proper precipitation might cause this high N<sub>2</sub>O emission.

Technical Points 1. Line 41: This statement ignores that some anthropogenic emissions were already present prior to or at the beginning of the industrial revolution.

Response: It is true that there existed anthropogenic N<sub>2</sub>O emission before 1860; however, the total amount is substantially lower than the contemporary human-induced N<sub>2</sub>O emissions. The description of anthropogenic emissions in the pre-industrial era

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was added, shown as “Human-induced biogenic N<sub>2</sub>O emissions are calculated by subtracting the pre-industrial emissions (Tian et al., 2016), even though a small amount of anthropogenic N<sub>2</sub>O emissions was present before 1860, which was estimated as 1.1 Tg N yr<sup>-1</sup> in 1850 by Syakila and Kroeze (2011) and 0.7 (0.6-0.8) Tg N yr<sup>-1</sup> in 1860 by Davidson (2009).”

2. Line 55: Add recent results from Prather et al. 2015.

Response: The latest study done by Prather et al. (2015) has been added into line 56, shown as “Prather et al. (2015) provided an estimate of the pre-industrial emissions (total natural emission: 10.5 Tg N yr<sup>-1</sup>) based on the then-most-recent model study with a corrected lifetime of 116±9 years.”

3. Line 70: Change “is” to “are” because the word “data” is plural: “the data are”. Response: It has been revised.

4. Line 178: Use estimate from Prather et al. 2015.

Response: We have removed the section 2.4.2 and section 3.2, which described the one-box model validation of simulation results. Thus, there is no need to replace the N<sub>2</sub>O lifetime in this section. In addition, we added the comparison of the estimate in this study with the estimation by Prather et al. (2012, 2015) in the section 3.2.

5. Line 312: Consider other estimates, such as those of Voss et al. 2013.

Response: We carefully read the paper from Voss et al. (2013) and found that their estimates were directly from the IPCC AR4 (Table 7.7 in Chapter 7). In their paper, the N<sub>2</sub>O emission from ocean was 5.5 Tg N yr<sup>-1</sup> because they considered the emissions (1.7 Tg N yr<sup>-1</sup>) from “rivers, estuaries, and coastal zones” as the marine emissions. Thus, the average marine emissions are 3.8 Tg N yr<sup>-1</sup>, as shown in Table 6.9 of Chapter 6 in the IPCC AR5.

6. Figure 2. I don't understand the units. How can these units of crop area apply to each individual pixel?

C14

Response: To avoid the confusion of the unit, it has been changed from “km<sup>2</sup>” to “km<sup>2</sup>/grid”. The size of individual pixel is 0.5 degree, equivalent to around 2500 km<sup>2</sup> at the equator. Meanwhile, we have crop area fraction in each pixel (mentioned in the section 2.1 & 2.2). Then, in each grid, crop area fraction multiplying the pixel size represents the crop area. The numbers in the legend mean the cropland area in each 0.5-degree pixel.

7. Figure 3. The data used for this graph should be referenced.

Response: All papers that used for the graph cited in the new version.

8. Figure 5. The bottom panel is all that is needed. The top panel is redundant. However, you could also add a panel of mean flux per hectare, which would be useful, because it is difficult to compare fluxes across continents when the contents have such different total areas.

Response: We agree with the reviewer. We have removed the top panel. Instead, we added a panel of N<sub>2</sub>O emission rates per unit area (g N m<sup>-2</sup> yr<sup>-1</sup>) with uncertainty ranges at continental-level in 1860, as shown in Fig. 5 (a).

9. Figure 6. The two panels are largely redundant. The pie chart could include both the percentage of the total and the estimate of Tg/yr, which would obviate the need for the upper panel. However, again, the mean flux per hectare by biome would be an interesting panel to add.

Response: We agree with the reviewer. We have removed the top panel. Instead, we added a panel of N<sub>2</sub>O emission rates per unit area (g N m<sup>-2</sup> yr<sup>-1</sup>) with uncertainty ranges at biome-scale in 1860, as shown in Fig. 6 (a). In addition, we added the biome-scale emission amounts and their uncertainty ranges into the pie chart, as shown in Fig. 6 (b).

10. Table 2. The number of significant figures shown is excessive. I suggest rounding to the nearest Gg. The uncertainties are such that any fraction of a Gg is meaningless.

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Response: Since the one-box model section has been removed, Table 1 was deleted, and “Table 2” was changed to “Table 1”. The uncertainties have been removed. We added the biome- and continental-scale N<sub>2</sub>O emissions in the supplementary material (Table S2). For the mean annual N<sub>2</sub>O emissions (Tg N yr<sup>-1</sup>) and emission rate per unit area (kg N ha<sup>-1</sup> yr<sup>-1</sup>), we have listed all numbers in the Table S3. We included the revised figures as below:

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Please also note the supplement to this comment:

<http://www.clim-past-discuss.net/cp-2016-103/cp-2016-103-AC1-supplement.pdf>

Interactive comment on *Clim. Past Discuss.*, doi:10.5194/cp-2016-103, 2016.

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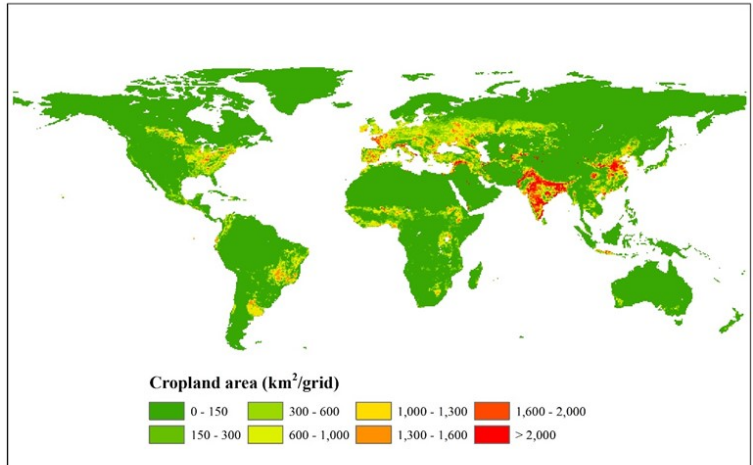


Fig. 2 The spatial distribution of cropland area in 1860.

Fig. 1.

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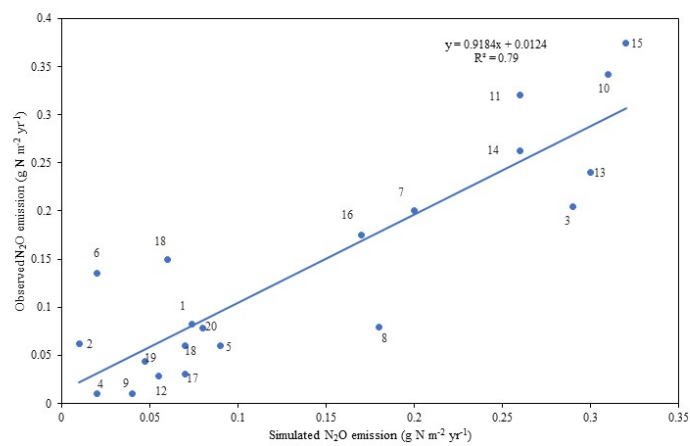


Fig. 3 The comparison of the DLEM-simulated N<sub>2</sub>O emissions with field observations. All sites were described in the supplementary material (Table S1).

Fig. 2.

C20

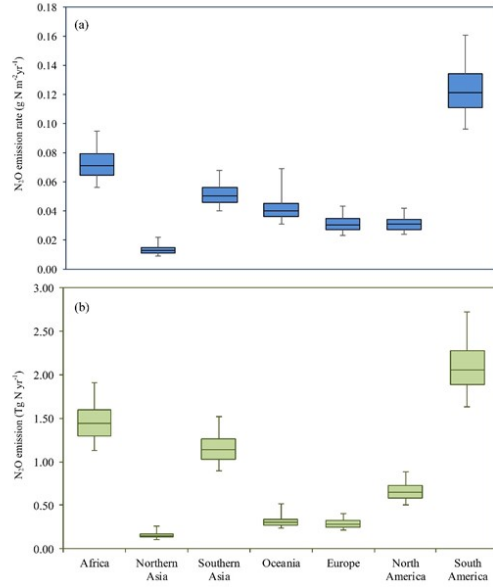


Fig. 5 Estimated  $N_2O$  emission rates (a) and emissions (b) with uncertainty ranges at continental-level in 1860. Solid line within each box refers to the median value of  $N_2O$  emission rate or amount.

Fig. 3.

C21

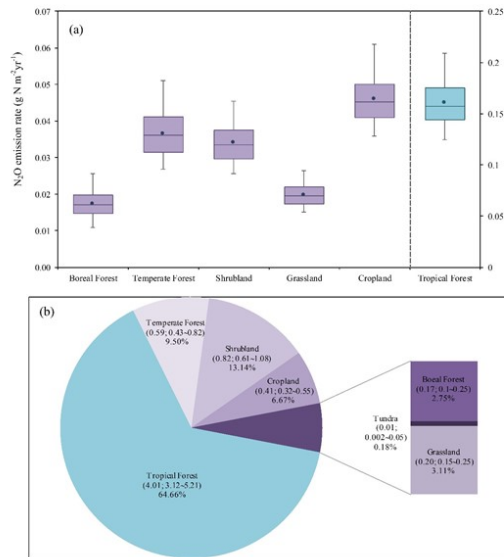


Fig. 6 (a) Estimated  $N_2O$  emission rate at biome-level in 1860 with the median value (solid line), the mean (solid dot), and the uncertainty range of emission rates from different biomes. The emission rate in the tundra was removed because of the extremely small value (less than  $0.003 g N m^{-2} yr^{-1}$ ); (b) Estimated  $N_2O$  emission (Tg  $N yr^{-1}$ ) with uncertainty ranges and its percentage (%) at biome-level in 1860.

Fig. 4.

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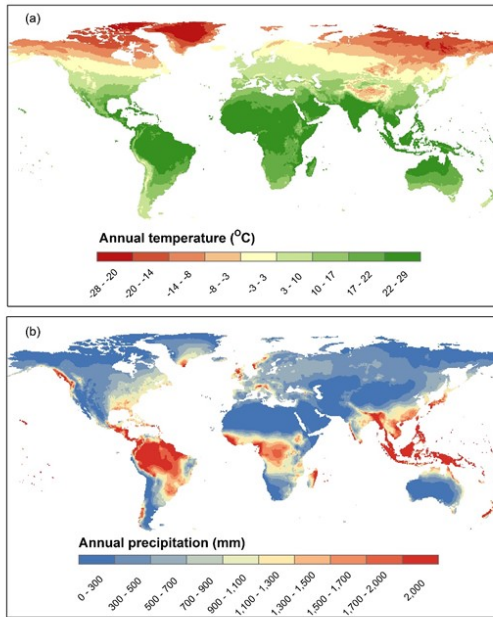


Fig. S2 (a) The average annual temperature during 1901–1930; (b) The average annual precipitation during 1901–1930.

Fig. 5.