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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global and regional emissions estimates for N2O" *by* E. Saikawa et al.

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Thank you very much for your comments on our paper. We appreciate your time to read our manuscript and give us these comments. Below please find our reply.

> General comments

> This manuscript presents new estimates for N_2O emissions at the global and continental scale using observations of N_2O mole fractions and a global transport model in an inversion framework. The emissions are estimated annually from 1995 to 2008 and are analysed for regional trends and inter-annual variations. This study represents a large effort in terms of compiling and analysing the observational data,

and determining the calibration offsets between each of the networks included. The authors have also made considerable effort in compiling different prior estimates for the natural soil emissions.

> While one novel aspect of this study is the optimization of the emissions by source type (natural soils, agricultural soils, industry, biomass burning and oceans), this also presents one major concern: that there is no observational constraint for the allocation of the emissions on land to agricultural soil, natural soil, industrial or biomass burning sources. There is not even a geographic separation of the sources, as the emissions are solved at continental or sub-continental scale.

> The emissions from the inversion also seem to be strongly dependent of the prior estimate (in Fig. 3). In Fig 3a there is only error reduction in Europe, North America and South Asia and for the other regions the posterior emissions are almost undistinguishable from the prior. In Fig 3b, again the a priori and a posteriori industrial emissions are almost the same with very little to no error reduction. In Fig 3c, for natural soil emissions there is error reduction only for Northern Asia, Southern Asia, North America, and Central South America. It is surprising, however, that there is error reduction for e.g. Northern Asia and Central South America where there is only a very weak observational constraint but in Europe, where there are many more observations, there is very little error reduction.

> For these reasons, the authors should consider including one figure showing the total emissions (all sectors) for each region and the uncertainty and extend section 4.1 explaining why they think it is justified to try to solve the emissions by sector. Also, to help the author's case (i.e. that it is justified to try to solve the emissions by sector) a figure or table showing the correlation of the posterior errors for each sector and region should be included. Furthermore, although this paper focuses on the optimization of

emissions by sector (natural versus agricultural etc) there is no mention of delta-15N isotope measurements, which could help provide a constraint on this. Admittedly, these measurements are rare but may still provide a weak constraint on the different source types at continental scales.

We have included a figure 3b that shows the total emissions for each region and the uncertainty. We have also quantified the correlation of the posterior errors for each sector and region and rather than including a table or figure which is not too helpful, we have also inserted the following in section 4.1 to explain why we think it is justified to solve the emissions by sector: "A large aggregation error is possible by conducting inversion to solve for aggregated regions and sectors as we do here because we assume that the spatial distribution in the prior emissions is correct (e.g., Kaminski et al., (2001); Meirink et al., (2008)). We therefore calculated the average correlations (R) between optimized emissions to evaluate if we find high anti-correlations between them in the neighboring regions or specific sectors. There are 9 cases where R values lie between -0.7 and -0.8, between the natural soil in Northern and Southern Asia, and between agricultural soil and anthropogenic emissions in Europe. However, more than 99.7% of the absolute R values are less than 0.3. We therefore stress that total emissions are a robust result but emissions by source sector and regions have much larger uncertainties."

> Lastly, although this study is generally well presented English language editing is recommended.

We have edited the paper in the revised manuscript.

> Specific comments

> p19473, l21-24: Actually, this statement is not correct, there are examples of at least 2 previous studies using multiple network data, i.e. Corazza et al., 2011 and Thompson et al. 2013.

We did not intend to say that this was the first attempt to combine measurements from different networks but what we meant to state was that this was the first attempt to include this many measurements (including the new ones) from diverse different networks. We have made clarifications as follows: "Although there have been papers that combined measurements from different networks for N_2O inverse modeling (e.g., Corraza et al. (2011), Huang et al. (2008)), the goal of the paper was, for the first time, to include all available measurements (i.e., in situ, flasks, aircraft, and ships) from all available networks where we have reliable and consistent inter-comparison of measurements."

> p19476, l9-11: Again, this is actually not the first time that these data have been combined in an inversion (refs. given above).

As mentioned above, we rewrote this sentence to be clearer of our intention.

> p19477, l10-12: Was only one error estimate used throughout the whole time period? It is known for example that the NOAA CCGG data prior to circa 1998 is much less certain than the data collected after this. Therefore, it would be important to account for this change in measurement accuracy in the observation uncertainties.

Yes, only one error estimate was used throughout the whole time period. It is true

that NOAA CCGG data prior to 2000 is indeed much less certain than the data collected after that. However, the standard calibration scales are not changing and for example NOAA's measurements all use NOAA-2006A scale and AGAGE uses SIO-98 scale. Shifts in the differences between co-located measurements result from uncertainty in propagating the scale to in situ measurement sites and those shifts result from various factors. This is why we include generous uncertainties when we assign the measurements of discrete samples and those are taken into account in our analysis. We modified the paper as follows to incorporate this comment: "We compare atmospheric N_2O measurements collected from each group as they are based on different absolute calibration scales (Table 2) and differences exist among measurement networks. Because the global average mole fraction of N_2O increases at approximately 0.2-0.3% per year (see Fig. 2), the calibration ratio of 0.9975 corresponds to a one year's rise in mole fraction, and thus the calibration difference of as small as 0.6% can be significant. Hence, it is very important for us to adjust all of the measurements into a single scale, even though calibration appears to be fairly close to each other. Shifts in the differences between co-located measurements result from uncertainty in propagating the scale to in situ measurement sites and those shifts result from various factors but the standard calibration scales are not changing over time. We include generous uncertainties to account for these shifts in the differences over time."

> p19479, l4: The authors should say in the text that this ratio is between the calibration scale of a given network to that of AGAGE, i.e. calibration scales are referenced to that of AGAGE. Also, it should be made clear at the beginning that this is the ratio of the N_2O data (this only becomes apparent later in this same paragraph).

We have rewritten the paragraph as follows: "We compare atmospheric N_2O measurements collected from each group as they are based on different absolute calibration

scales (Table 2) and differences exist among measurement networks. Because the global average mole fraction of N_2O increases at approximately 0.2-0.3% per year (see Fig. 2), the N_2O calibration ratio of 0.9975 corresponds to a one year's rise in mole fraction, and thus the calibration difference of as small as 0.6% can be significant. Hence, it is very important for us to adjust all of the measurements into a single scale referenced to that of AGAGE, even though calibration appears to be fairly close to each other."

> p19479: Should mention the impact of chosing AGAGE as the reference scale as opposed to e.g. NOAA. Since the scales are diverging, chosing one rather than another will have a small impact on the global and regional trends.

As mentioned above, the standard calibration scales are not diverging and for example NOAA's measurements all use NOAA-2006A scale and AGAGE uses SIO-98 scale. Shifts in the differences between co-located measurements result from uncertainty in propagating the scale to in situ measurement sites and those shifts result from various factors. This is why we include generous uncertainties when we assign the measurements of discrete samples and those are taken into account in our analysis. We modified the paper as follows to incorporate this comment: "We compare atmospheric N_2O measurements collected from each group as they are based on different absolute calibration scales (Table 2) and differences exist among measurement networks. Because the global average mole fraction of N_2O increases at approximately 0.2-0.3% per year (see Fig. 2), the N_2O calibration ratio of 0.9975 corresponds to a one year's rise in mole fraction, and thus the calibration difference of as small as 0.6% can be significant. Hence, it is very important for us to adjust all of the measurements into a single scale referenced to that of AGAGE, even though calibration appears to be fairly close to each other. Shifts in the differences between co-located measurements result from uncertainty in propagating the scale to in situ measurement sites and those shifts

result from various factors but the standard calibration scales are not changing over time. We include generous uncertainties to account for these shifts in the differences over time."

> Section 2: The authors do not discuss the changing precision and accuracy of the measurements at all in this section, which is an important consideration when looking at trends and inter-annual variations.

We have included the largest precision and accuracy errors for each instrument for the given period, but this would not change the results too much because the change in precision and accuracy is not too large over this period of time we are analyzing.

> p19481, l24: The reference(s) to these previous studies should be given here.

Hirsch et al. (2006) and Huang et al. (2008) are included in the revised manuscript.

> p19482, l14: Again, the authors should state which studies they are referring to.

Hirsch et al. (2006), Huang et al. (2008), Thompson et al. (2011), and Corrazza et al. (2011) are in the revised manuscript.

> p19483, l1-2: The estimated lifetimes of N_2O should be stated here.

We rewrote this sentence as follows: "The lifetime of N_2O, calculated by the ratio of the annual total global burden to the loss rate calculated in the chemical transport model is 116$\pm$5 yr, which is slightly lower than the current estimates of its lifetime of

131$\pm$10 yr (Prather et al., 2012) but is in alignment of 114 yr with Montzka et al. (2011)."

> p19483, l28: The temporal resolution of the emission sensitivities (i.e. in the Jacobian matrix H) should be stated, is this 1 year?

We have rephrased it as "We calculate the sensitivity to a change in annual emissions by dividing. . ." in the revised manuscript.

> p19484, l16-19: This belongs in the results section.

We have moved this to the results section.

> p19487, l5: The winter soil emissions in northern Eurasia are expected to be very low as the ground is frozen in most places during this time. Perhaps the authors are referring to the uncertainty in N_2O emissions from thawing permafrost, which could potentially be an important source of N_2O in the high northern latitudes? (see Elbering et al., 2010)

In previous studies, researchers (e.g., Groffman et al. (2011)) found N_2O being emitted in winter under snow and thawing conditions. There are still large uncertainties but there are possible winter/spring emissions that have not been well accounted for so far. We have included literature citations in this paragraph to make it clearer of our intentions.

> p19487, l20-22: From Fig. 1, Northern Asia does not appear to be as well covered by observations as e.g. North America or Europe.

Northern Asia has good coverage in terms of continuous in-situ measurements and because of the wind direction and large emission sources, we are able to see clear sensitivities at some measurement sites that are important for inversion. We have rewritten as follows to make this clearer: "We achieve these results because natural soil has the largest emissions among all sources and because we have the largest number of long-standing stations close to our Northern Asia region (see Fig. 1) that have high sensitivities."

> p19488, l5: Please state the confidence level and which statistical test of significance was used.

We realized that this was misleading when we cannot conduct any statistical test. Therefore, we have rewritten this sentence as follows: "Inter-annual variability is visible in the result, but emissions with one-sigma standard deviation in recent years are still significantly larger than similar estimates for 1995 and 1996 (see Fig. 3c), and this aligns well with a recent study showing increased N_2O emissions due to agriculture (Park et al., 2012)."

> p19488, l1-3: Fig. 3b shows a decreasing trend in the prior and posterior industrial emissions for Europe and North America, which is based on EDGAR-4.1. How well determined is this decreasing trend? Could it be that the increase in agricultural emissions found in the inversion may be compensating for the decreasing industrial emissions?

Europe is showing a decreasing trend in industrial emissions but not necessarily so in North America. The reviewer is correct, however, that there are largest anti-correlations

between anthropogenic and agricultural soil emissions in Europe, and this indicates the possibility of some compensating impacts. As mentioned earlier, we have inserted the following to make this point clearer: "A large aggregation error is possible by conducting inversion to solve for aggregated regions and sectors as we do here because we assume that the spatial distribution in the prior emissions is correct (e.g., Kaminski et al., (2001); Meirink et al., (2008)). We therefore calculated the average correlations (R) between optimized emissions to evaluate if we find high anti-correlations between them in the neighboring regions or specific sectors. The largest we find are between the natural soil in Northern and Southern Asia, and between agricultural soil and anthropogenic emissions in Europe. We therefore stress that total emissions are a robust result but emissions by source sector and regions have much larger uncertainties."

> p19488, l27-28: There only appears to be an obvious convergence of natural soil emission estimates for the regions Northern Asia, North America and Southern Asia (in fact it is difficult to tell because the subpanels in Fig. 3 are very small).

If you see Fig. 3e, the means of the four forcing datasets for Africa/Middle East and Central/South America, are also getting closer as the years pass by. However, it is true that GOLD emissions estimates in Europe stay much higher than the other estimates even in later years, and so we do not argue that this is happening in all regions. We rewrote the sentence as follows: "Interestingly, however, compared to the differences in the prior emissions estimates from the four different forcing datasets, the optimized emissions converge in North America, Central/South America, Africa/Middle East, Northern Asia, and Southern Asia, giving us confidence in the inversion results."

> p19491, l5: The AR4 estimate referred to here is actually Bouwman et al. 2002 and the original reference should be given. Presumably the authors have calculated the totals for North America and Europe from Bouwman's data themselves as this is not

given in the IPCC AR4 report.

These are from UNFCCC and not from the AR4 estimates. We have corrected this in the new manuscript.

> p19492, l11-13: Are these percentages the percentage of the total N_2O emission from each region? This should be made clear.

We have rewritten this sentence as follows: “Indeed, the 5yr mean of the percentage of regional anthropogenic emissions shifts from 69.3% between 1995-1999 to 79.0% between 2004-2008 in Southern Asia, and similarly from 44.0% to 47.9% in Central/South America.”

> p19492, l14: Should be specified that this is the natural soil and ocean emissions.

We have changed the sentence to read as follows: “These results indicate, however, that the natural soil and ocean emissions still share approximately 2/3 of the global total N_2O emissions, as has been discussed in the past (Werner et al. 2006)”

> p19493, l11-13: The authors should mention the work of Corazza et al. (2011), who incorporated the optimization of the calibration scale offsets into their inversion. While it is indeed true that more work is needed to harmonize the calibration scales and measurement networks, considerable effort has been made to account for these offsets in inversions.

What we argued for was to have the standardized calibration scale for all measure-

ments, rather than optimizing the calibration scales.

> p19494, l1-5: Again, the authors should recognize the work of Corazza et al. (2011) who have performed a high-resolution inversion of N_2O globally with a zoom over Europe.

We have rewritten the sentence as follows: “Second, the use of finer-resolution chemical transport models and meteorology data would also allow us to disaggregate regions further and detect sensitivities to atmospheric mole fractions due to increases in emissions more accurately, as shown by the work of Corazza et al. (2011) focusing on Europe.”

> Technical corrections

> p19474, l11: grammatically correct: “Since N_2O is inert within the troposphere, it has a long atmospheric lifetime: : :”

The sentence is changed.

> p19474, l13: no comma after “emissions”

Comma is taken away.

> p19474, l17: not “has” but “have”

Changed to “have”.

> p19474, l18: need to add “sources” after “anthropogenic” to be grammatically correct

Changed the sentence to: “There are large uncertainties associated with estimated emissions from each of these sectors, but approximately 2/3 of the emissions have been attributed to natural soil and ocean, with the remaining to anthropogenic sources (Khalil et al., 2002; Denman et al., 2007; Nevison et al., 2007).”

> p19476, l4: “sectorial” (and elsewhere e.g. section 4 heading, Table 3)

We have decided to use “emissions by source sector” in the entire manuscript.

> p19477, l27: “as well as at the seven following sites:: : :”

Changed.

> p19478, l25: it would be more accurate to say that the Tohoku University data cover a wide latitude range from north of Japan to Australasia.

We have changed the sentence as follows: “The data between 2003 and 2009 are new and cover a wide latitude range from the north of Japan to the Oceania region.”

> p19479, l7: “: : :the calibration scales appear to be close to one another.”

Changed.

> p19481, l12: “reproduces” suggests that the seasonal and inter-annual emissions have been validated and are correct, which means that there would be no need to perform an atmospheric inversion (at least for this source type), therefore, “estimates” or “simulates” would be more appropriate here.

We have validated the model against measurements looking at the seasonal and inter-annual variations and we found that they reproduce very well at some sites whereas not so much in others. We have therefore changed the sentence as follows to be clearer about our point: “For natural soil emissions, we use another newly developed process model (CLMCN-N_2O) that is able to reproduce the seasonality and inter-annual variability of global emissions at some sites (e.g., Amazon) and not at others (e.g., North America).”

> Generally: attention should be paid to the use of articles and commas.

We have edited the manuscript especially focusing on this point.

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