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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:
> The manuscript presents an estimate of global and regional N_2O emissions from atmospheric inversions. Measurements from different networks are combined, which is a particularly demanding task in case of N_2O because significant offsets exist between the different laboratories and networks. Spatial gradients and temporal trends of N_2O are in the same order of magnitude as these offsets and could therefore directly lead to significant biases in the inferred emissions. Therefore this harmonization is a prerequisite for the use of measurements from multiple networks in inversions. The

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authors carefully adjust the measurements from the different networks to the same calibration scale based on measurements at sites shared by two or more networks. Potential shifts in the calibration offsets are, however, not taken into account. This possibility should be discussed in the paper as well, at least briefly.

The standard calibration scales are not changing and for example NOAA's measurements all use the 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, and this divergence is small compared to other uncertainties."

> A major shortcoming of the paper is the absence of a careful assessment of the potential influence of the stratospheric sink and stratosphere-troposphere exchange on the simulated N_2O mixing ratios and hence on the emission estimates. Even

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though the authors restrict their study to the estimation of annual mean fluxes (to avoid the influence of potential errors in the simulated seasonal cycle of N_2O mixing ratios caused by known problems of the model to correctly simulate transport between stratosphere and troposphere) there can still be a potential influence of these processes on the internal variability of estimated emissions. The authors should describe in more detail the initialization (e.g. spin-up time) that was used to bring the model into equilibrium before the actual inversion. Otherwise the redistribution of N_2O between stratosphere and troposphere during the inversion can lead to large systematic uncertainties in the optimized fluxes. In this context also information on the statistics of the prior and posterior N_2O mixing ratios at the stations should be given.

We conducted a spin-up for 10 years starting with uniform latitudinal mixing ratio varying by vertical levels based on observations and considered the model as having reached equilibrium by comparing the modeled mixing ratios to surface observations at measurement sites that we used for the inversion and that had observations in 1995. We created the statistics of the prior and posterior N_2O mixing ratios at the stations and present them in the supplementary material. We also added the following in the text: "The spin-up was done for 10 years starting with a uniform latitudinal mixing ratio and with vertical distributions based on observations. We then compared surface mixing ratios to available observations in 1995 to start the simulation." We also included the following statement in the text, where we discuss the ways to make our inversion estimates more accurate: "Fourth, including the whole stratosphere (e.g., using the Whole Atmosphere Community Climate Model) and better implementation of the stratospheric sink may improve the simulation of stratosphere-troposphere interactions, which could result in a better representation of monthly variability that we observe in the measurements."

> Furthermore, a critical assessment of how well the sectoral emissions can be

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disentangled should be included, e.g. emissions from agricultural soil and industry are often co-located in Europe. Providing very detailed tables suggests that these sectoral estimates are as reliable as the estimate of the total emissions. Regional inversions are known to suffer from a potentially large aggregation error, which can cause significant uncertainties in the estimated emissions (e.g. Kaminski et al., 2001). The authors should comment on this and try to quantify the influence.

We do not mean to state that these are as reliable as the estimate of the total emissions. Indeed, we find the highest anti-correlation among the optimized error covariance between those two sources in Europe. This paper was our first attempt to understand sectoral emissions, while recognizing the large uncertainties associated with them. We have included a paragraph on aggregation error as well as an assessment on sectoral/regional emissions as the following: "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."

> The paper is well written and well structured. However, the paper seems to be part of a series of papers on inversions of different trace gases and large parts of the text describing inversion set-up and techniques are identical (word-by-word) with Saikawa et al. (2012). This must be avoided. If the model and inversion technique are already published elsewhere in a paper, then only a summary with reference should be given.

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We have taken out the overlap from the Saikawa et al. (2012) paper and only included a summary with a reference.

> Specific comments:
> P 19476, l 10: It should be mentioned in the introduction that there has already been at least one attempt to combine measurements from different networks and also to include a bias correction scheme in the inversions (Corraza et al., 2011)

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 all the measurements that we have included in this paper (including the new ones) from this many 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 to include all available measurements (i.e., in situ, flasks, aircraft, and ships), for the first time, from all available networks where we have reliable and consistent inter-comparison of measurements."

> P 19481, l 20: Which previous studies? Please give a reference.

We have included the references as follows: "Four forcing datasets used are: Global Meteorological Forcing Dataset (GMFD) by Sheffield et al. (2006); NCEP Corrected by CRU (NCC) by Ngo-Duc et al. (2005); Climate Analysis Section (CAS) by Qian et al. (2006); and Global Offline Land-Surface Dataset (GOLD) by Dirmeyer and Tan (2001) and Betts et al. (2006).

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> P 19485, l 8: : : :errors in this : : :

We have changed this sentence to “errors in this study”

> P 19485, l 21: On the other hand flasks are usually taken at specific times to represent background concentrations while in-situ measurements can include strong pollution events and therefore the standard error of in-situ data is not necessarily 3 to 10 times smaller. Please comment. How are in-situ and flask measurements combined at stations where both types are available?

It is true that in-situ measurements can include strong pollution events, but because of the number of measurements in-situ measurements can collect compared to the flask measurements, standard error, taking into the number of measurements, does get 3 to 10 times smaller than flask measurements. When we combine both in-situ and flask measurements, we created monthly averages, weighted by the number of measurements.

> P 19489, l 6: Unfortunately the paper by Saikawa et al. (2013) was not available at the time of the review.

It is now published in Global Biogeochemical Cycles and the citation has been changed to reflect this.

> P 19490, l 11: The study by Corazza et al. was not lagrangian-based and used not only NOAA-ESRL data but also quasi-continuous measurements from different station networks in Europe. In the paper by Corraza et al. emissions for north–central Europe

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are estimated to be 0.78 TgN_2O-N/yr.

We have rewritten the paragraph as follows: “Our estimates however agree with other studies done in Europe. For example, Corazza et al. (2011) used NOAA/ESRL and other quasi-continuous measurements and a four-dimensional variational (4DVAR) technique with an atmospheric transport zoom model to estimate European emissions in 2006 and quantified annual European N_2O emissions to be 0.78 TgN_2O-N yr^{-1}, and this value is in alignment with ours ($0.85 \pm 0.10 TgN_2O$-N yr^{-1})."

> P 19491, l 3, Figure 5: How can emissions for 2011 be part of the IPCC AR4 report that came out 2007? At least I could not find these numbers in Chapter 7, as cited. Are the emission estimates from UNFCCC? For Europe the differences between the IPCC estimates and the prior and optimized emissions are huge. What could be the reason? Please comment. Check with emissions reported to UNFCCC.

These are the emissions reported to UNFCCC and they are not part of the IPCC AR4 report, and we have corrected the citation. The large differences between the reported emissions and the prior are due to the differences between those emissions in UNFCCC and the global emissions inventory EDGAR v4.1 that we use for industrial and agricultural soil emissions. We write the following in the manuscript: “The large discrepancy we find between the UNFCCC estimates and the prior emissions in Europe is due to the difference from the EDGAR v4.1 emissions. Because optimized emissions are sensitive to prior emissions in Europe due to little sensitivity in the region, we find discrepancies in posterior emissions as well.”

> P 19491, l 17: This comparison is for global total emissions, not land emissions, cf. Tab. 3 in this study and Tab. 4a,b in Hirsch et al. (2006).

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We have corrected the sentence to be as follows: “Hirsch et al. (2006) estimated the global total land emissions to be 9.7-13.6 TgN_2O-N yr^{-1} for the period between 1998 and 2001. Our estimate of 12.4$\pm$0.58 TgN_2O-N yr^{-1} is in their range.”

> P 19492, l 2: Please briefly explain the concept of ODP-weighted emissions.

We write the following: “Furthermore, quantifying the ODP-weighted emissions, which provide an estimate of the impact N_2O has on global stratospheric ozone depletion in relative terms, results in 0.48Mt CFC-11-equivalent, and this is larger than the sum of the ODS emissions of those controlled by the Montreal Protocol (approximately 0.45 Mt CFC-11-equivalent) (Daniel et al., 2011).”

> P 19492, l15: ‘: : : as has been discussed in the past.’ Please give a reference here.

We have inserted the citation of Werner et al., 2007.

> Table 1b: Ochsenkopf, Germany

We have corrected this in Table 1b.

> Tables 5-8: How were the uncertainties defined? How do the prior uncertainties correspond to the 40

We have included 40% uncertainty for each sector and for each region for prior

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emissions. Table 5 gives aggregated regional emissions, and this is lower than 40% due to the correlations among sectors and regions.

> Tables 5-8 provide information that is also displayed in Figures 3a-d. The figures are much easier to conceive and provide even more information. Therefore the tables should be move to a supplement. Moreover, also Table 3-4 could be visualized as figures.

We included information on Tables 3 and 4 as Figures 3a and 3b. We have decided to keep the table in the text as it would be easier for the readers to compare these values.

> Figures 3a-b: The region map could be a separate figure and instead a panel with global emissions should be included.

We believe that how we have is easier to see everything on one page rather than putting the region map separately in one figure. We have created a panel with global emissions as well as global land and global ocean in a different figure as explained above.

> Figure 6: Please use the same scale for all uncertainty reduction plots.

We have recreated the figures using the same scale (except for Oceania, which has much smaller uncertainty reduction compared to others) for all uncertainty reduction plots.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 19471, 2013.