Atmos. Chem. Phys. Discuss., 13, C6832–C6835, 2013
www.atmos-chem-phys-discuss.net/13/C6832/2013/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

Received and published: 12 September 2013

General comments:

The manuscript presents an estimate of global and regional N2O emissions from atmospheric inversions. Measurements from different networks are combined, which is a particularly demanding task in case of N2O because significant offsets exist between the different laboratories and networks. Spatial gradients and temporal trends of N2O 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 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.

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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 N2O mixing ratios and hence on the emission estimates. Even 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 N2O 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 N2O 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 N2O mixing ratios at the stations should be given.

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

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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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)

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

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

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?

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

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 are estimated to be 0.78 TgN2O-N/yr.

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.

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).

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

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

Table 1b: Ochsenkopf, Germany

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

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.

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

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

Corazza, M. et al. (2011), Inverse modelling of European N2O emissions: assimilating observations from different networks, Atmos. Chem. Phys, 11(5), 2381–2398, doi:10.5194/acp-11-2381-2011.

Kaminski, T., P. Rayner, M. Heimann, and I. Enting (2001), On aggregation errors in atmospheric transport inversions, J. Geophys. Res, 105, 4703–4715.

Saikawa, E. et al. (2012), Global and regional emission estimates for HCFC-22, Atmos. Chem. Phys, 12(21), 10033–10050, doi:10.5194/acp-12-10033-2012.

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