

## Interactive comment on "Top-down constraints on global N<sub>2</sub>O emissions at optimal resolution: application of a new dimension reduction technique" by Kelley C. Wells et al.

## Anonymous Referee #2

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This paper uses a multi-inversion hierarchy to derive top-down constraints on N2O emissions for 2011. The goal is to make a detailed evaluation of the 3 different methods and their impacts on inversion results. All methods are based on the adjoint of the GEOS-Chem chemical transport model, where 4D Var is considered the "standard" approach, as well as two alternative ways for aggregating the results, given that the existing observational network is insufficient to fully constrain N2O emissions at the gridscale level. The first approach uses the 4DVar method, but aggregated to the traditional 6 continents and 3 oceans. The more novel approach tested is a new SVD-based technique based on the "prior- preconditioned Hessian of the 4D-Var cost function."

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An additional goal is to address the impact of initial condition uncertainties using 6 different approaches. This analysis is performed first and an optimal approach is selected for use in the evaluation of the 3 different inversion methods.

The paper is well written and logically organized. While some of the mathematics, particularly the SVD approach, are beyond my ability to evaluate, I found the results and discussion interesting and insightful. My main criticisms are, first, there seems to be a predisposition to claim the SVD results as the "best estimate of the true global flux." This conclusion is not clearly based on objective criteria. Other interpretations that might be more critical of SVD are not discussed, including the odd, spiky SVD results (e.g., in South America, Africa and the Tropical Oceans in Figure 7). Second, there is an unwarranted emphasis on the results of Chen et al. 2016, which are often presented as though they were primary results of what is overall an impressive and interesting body of work. I recommend publication with some relatively minor revisions detailed below.

## Specific comments

Abstract L31-32 "the inversions reveal a major emission underestimate in the US Corn Belt (which may extend to other intensive agricultural regions), likely from underrepresentation of indirect N2O emissions from leaching and runoff." Please clarify an underestimate relative to what? Also, the last part of this sentence is supported only on p12L30 with a reference to Chen et al. 2016. It is not supported by the current study and does not really belong in the abstract as a new primary finding.

As an aside, I will make a few comments about Chen et al. 2016, which is referenced multiple times (e.g, again on P17L27) as the source of the conclusion that the underestimate of indirect emissions is responsible for the underestimate of agricultural emissions in prior inventories. Realistically, I don't think the Chen et al. methodology is able to separate indirect and direct emissions. Their prior direct agricultural source

is based on EDGAR, which is at least somewhat reliable since it is computed using gridded N inputs from fertilizer, etc. multiplied by emission coefficients. In contrast, the indirect source is based on the CLM45-BGC nitrate leaching and runoff flux, which is unreliable and almost certainly wrong (see, e.g., Houlton et al., Nature Climate Change, 5, 398, 2015). The Chen methodology then assumes those 2 prior sources accurately represent the spatial and temporal distribution of direct and indirect N2O emissions, respectively. That methodology is fraught with uncertainty. Moreover, the fact that (as stated on p16L28) indirect emissions peak earlier than direct emissions is a red flag that something is wrong. This result doesn't make sense, given that indirect emissions, by IPCC definition, occur later and downstream/downwind of direct emissions.

P2L24 Crutzen et al., 2008; Davidson, 2009 are not really bottom-up emissions. They are based more on a top-down approach (in a global box model sense) of comparing the observed atmospheric N2O increase to the rate of external N inputs and anthropogenic N fixation.

P3L10-12 It seems somewhat over-critical to say previous aggregation has been informal and ad hoc. It's been based largely on geographical and political boundaries, i.e, North vs. South America, Pacific vs. Atlantic Ocean, etc., which are logical regions of interest.

P3L10-18 Exact totals are given for the ocean, GFED and EDGAR non-agricultural sources, but the Saikawa non-agricultural land and the EDGAR agricultural source are not specifically stated, yet these are the largest component sources. Please report them too. Two additional points are that the Saikawa source was based on a global model without cropland, such that it included a "non-agricultural" soil source from land such as the US Midwest where crops are grown. Also, the EDGAR v4.2 total is about 1.7 TgN/yr from industry, wastewater and energy. To bring up to the reported 2.3 TgN/yr, I wonder if the authors have included the EDGAR savanna, forest, grass and agricultural fire fluxes (of 0.84 Tg Nyr), which might be redundant with the GFED source? (Note: my numbers are from 2005 and thus may be slightly different from

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## 2008.)

P4L29 Please state the time resolution of the inversion somewhere around here.

P5L19 Negative emission scaling factors may be appropriate for some oceanic regions, especially during seasonal cooling in regions where the biological source is small and thermal solubility-driven uptake may dominate the air-sea flux.

P5L27-29 Can we infer from this that the total observational uncertainty (which is also referred to as model-data mismatch uncertainty) is typically about 0.45 ppb? It would be useful to state this. It is interesting and unexpected that the observational uncertainty dominates the model representation uncertainty. At only 0.2 ppb, the model representation error seems substantially underestimated. Also, considering that the grid resolution is 4x5 degrees, how many grid boxes actually "surround" any given observation and what kind of heterogeneity is missed inside the actual box that contains the measurement?

P7 The SVD-method is complex to the point of being unfathomable for many readers (including me!), so we must take it on faith that the calculation is accurate. Given the lengthy form of equation 5, I am concerned that it would be easy for human errors to slip into the calculation. What assurances do we have that such errors will be detected?

P9-10 I found this section difficult to follow and did not emerge with a clear understanding of why certain initialization methods are better than others. I'm not sure what to suggest to help clarify, but one step might be to include some columns for the NH-SH gradient in Table 2 in addition to (or perhaps instead of) the separate NH and SH bias columns. Those are not really referred to in the text, while the "overly strong interhemispheric gradient" is mentioned on P9L26 but is not obvious in Table 2.

P11L8 The statement that SVD "appears to provide the best estimate of the true global flux" seems based on fairly limited and/or subjective criteria. Furthermore, it is not obviously true that SVD agrees best with HIPPO. In fact, it seems to agree worst from 30S-

30N. (This is attributed on P11 to the fact that "the spatial distribution (in the tropics) is particularly difficult to resolve," but this is not necessarily a satisfactory explanation.) Is the comparison to HIPPO based on subjective visual inspection or some more quantitative measure? Also, are we sure the HIPPO calibration scale is not systematically biased from the data used in the inversion, especially given the adjustments described in section 2.3?

P13 section 4.3.3. The results for Europe indicate a fairly dramatic reduction from the prior. Please state the total non-agricultural prior source in EDGARv4.2. How much of the total 1.70 Tg N prior does it comprise?

P14L31 The results of 3.35-3.48 are above the range found by Buitenhuis (2.4 +/- 0.8).

P17L20 It's perhaps notable here that the EDGAR industrial source has dropped by about a factor of 2 in version subsequent to v4.2 used here.

P17L23-24. I think the main issue is that the seasonality in the existing inventories used here is governed by natural soil emissions from a model without crops. The EDGAR agricultural source with no seasonality is then added. However, the hotspot of emission is in agricultural areas where the seasonality is influenced by spring fertilizer input. Thus the seasonality of the existing inventories was predictably wrong from the outset.

P31 Figure 3 caption. Do the bars show the median (as currently stated) or the mean of the 3 inversions? How meaningful is the median of just 3 values? Would it be better to just show all 3 results + prior, i.e., 4 bars per region?

P34. I'm not sure Figure 6 adds much value to the paper. Furthermore, why is the KCMP measurement of primary interest to the current study?

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-637, 2017.

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