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Interactive comment on “Inverse modelling of European N₂O emissions: assimilating observations from different networks” by M. Corazza et al.

Anonymous Referee #1

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This paper presents early results from a 4D variational data assimilation system for atmospheric N₂O, applied at coarse resolution globally and zoomed in at finer resolution (1x1) over Europe. This topic is appropriate for ACP and it is exciting to see this type of advanced top-down constraint applied to N₂O. On one hand, the early results are intriguing. On the other hand, as atmospheric inversions become more and more sophisticated, it becomes increasingly difficult to understand the details of the model and to assess whether various results are caused by real features in the data vs. biases and assumptions in the model. The reader finds himself in the position of having to decide whether to accept some conclusions more or less on faith.

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Below, I list several conclusions that might benefit from further scrutiny.

1) It seems hard to believe that the model calculates the same fluxes and biases among different networks regardless of whether the NOAA measurements, which are considered as the universal standard, are included. I am comfortable with the idea that the inversion can simultaneously solve for biases and optimal fluxes if the NOAA data are included, but I don't understand how this can be true without them, given that a bias is calculated for each individual station (rather than network) and that the fluxes have a heterogeneous spatial distribution and vary over a factor of 10 in magnitude (Figure 3). Could the model primarily be balancing out the atmospheric N₂O values at different sites to bring them all to a relatively uniform value? Judging from Figure 2, this seems to be the case, with all stations showing an a posteriori value around 320.5 ppb. What kind of spatial gradients then are left in the data to help guide the estimate of the spatial distribution of sources? To help answer these questions, it might be useful to show in the Supplementary Figures a contour map, for a selected month or two, of the observed atmospheric N₂O mixing ratio over Europe before and after the bias corrections.

Regarding the use of the NOAA flask data as the unbiased standard, I am concerned about using bi-weekly flask data, with an average flask pair agreement of 0.4 ppb, to identify biases in the in situ data. It is considerably easier to filter out anomalous readings and problems associated with data representativity using high frequency in situ data rather than flask data, especially for a gas with low signal to noise like N₂O. What kind of biases might be introduced in the inversion due to uncertainties in the NOAA data?

2) Re: Section 4.2.2: It seems somewhat misleading to say the model improves a priori emissions if the main improvement is to scale up emissions. Simple back of the envelope calculations, such as those described in Hirsch et al. [2006], make it clear that the GEIA inventory at 13.6 TgN/yr substantially underestimates total N₂O emissions. A more challenging question is whether the 4DVAR method can improve the spatial and temporal distribution of emissions. It would be interesting to tabulate whether the

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relative percentage of European emissions on a country-by-country is changed significantly for prior and posterior fluxes. Clearly Britain's relative share must decrease, based on Figure 3, but I see no obvious reason why Britain's emissions should be overestimated by either GEIA (Figure 3) or the UNFCC (Figure 5) while most of the rest of Europe has been underestimated. This seems more likely to be an artefact of the inversion rather than a real result.

On a related note, please give a reference and brief description of the UNFCC estimates. These are reported, I believe, on a country-by-country basis and are estimated using a different methodology than the gridded GEIA sources used as the prior.

3) The seasonal cycle in N₂O data over Europe has an amplitude around 0.7 ppb, with a relatively deep minimum in late summer, which is probably caused in large part by an influx of depleted air from the stratosphere. Given that the inversion is restricted to a ~1 year time span, how can we be sure that the inversion is properly partitioning seasonality in the data between surface sources and stratospheric influences? Could the stratospheric influence be affecting the seasonality of sources presented in Figure 5? Some additional, related comments: a) p.26329 states that the stratospheric destruction reactions have pronounced seasonality, but the more relevant issue is the seasonality of Strat-Trop Exchange. Do we have evidence that the TM5 captures the seasonality of STE accurately? b) Please describe in more detail how high the TM5 model extends into the stratosphere and how the ECHAM5/MESSy1 sinks are incorporated into the TM5. c) Is there a reference to support the claim that the May-June emissions peak in Figure 5 is "very likely" (p.26340, line 15) related to fertilizer use in Europe? (It seems perhaps a bit late for a spring fertilizer application.) Also, why would Benelux or Britain have a fall emissions peak, in contrast to many of their neighbors, given that these countries have N-intensive agricultural production? An acknowledgement that some if not most of the apparent seasonality in the fluxes may arise from uncertainties in the model, particularly in the handling of STE, might be needed.

Despite the skeptical remarks above, I like this paper overall and appreciate the large

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effort that went into setting up the 4DVAR system and making it operational for N2O. It will be interesting to see how well the model can constrain surface N2O emissions in Europe as the model and data evolve and are refined. I support publication after the paper is revised to include a more thorough discussion and acknowledgement of the model's limitations and uncertainties.

Some additional minor comments:

Abstract, line 23: The sentence about Southern Europe is ambiguous. Please state more clearly.

p.26324, lines 4-5, please give a better quantitative summary of the number of stations. "various" and "a number of" are unnecessarily vague terms.

p.26326, line 8. The Dlugokencky et al. [1994] reference is for methane and predates the start of the NOAA N2O program by several years. I don't think there is an updated reference specific to the analytical aspects of the N2O flask program, but perhaps a reference like Hirsch et al. [2006] should be added.

p.26325, last full paragraph. Please clarify which stations have in situ data, etc. It is ambiguous as written whether the stations other than the CHIOTTO towers are in situ.

p.26330, line 1, sentence beginning, "Due to the correspondence. . ." Please clarify this confusing sentence.

p.26331, While I understand the need for brevity, it would be useful to give a short explanation of what the variables actually are, beyond a purely mathematical description. Some of the less obvious terms are B and H. What is the "background error" and what goes into H? Is it, e.g, an adjoint of the TM5?

p.26334, lines 1-2 and throughout paper. "Associated to" should be "associated with"

p.26336, line 3, "Shortly" should be "briefly"

Figure 1., please clarify in the caption whether the biases to the right of the plots are

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those estimated in the inversion or calculated directly from the data comparison.

Figure 2., needs x-axis label, at least at the bottom of the graph.

Figure 4. Please explain in more detail why this ratio represents the reduction of uncertainty.

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