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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Relating atmospheric N_2O concentration to N_2O emission strength in the U. S. Corn Belt" *by* Congsheng Fu et al.

Anonymous Referee #1

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General comments:

This study attempts to optimize emissions of N2O in the Corn Belt region of the US using observations of atmospheric N2O mixing ratios from a single tall tower site, KCMP. However, the methods used are not appropriate and involve gross assumptions that influence the results. More specifically, the method employed to optimize the emissions is over simplified: only one scalar is estimated and used to scale all the emissions in the inner domain (corresponding to the whole Corn Belt region) with no accounting for errors in the spatial distribution of the prior emissions. The low correlation between the modelled and observed mixing ratios, even after optimization, indicates that the spatial distribution of the prior emissions is likely wrong. Strong emissions close to the tower would have a very strong influence on the mixing ratio, if these are missing in the prior, then scaling all emissions equally will not correct for the error in the

prior. There are also concerns about the representation of the background mixing ratio, which was based on observations at Niwot Ridge, a high altitude site (>3000 m above sea level). Furthermore, there are concerns about the accuracy of the modelled atmospheric transport (see specific comments below). Additionally, in places, the references do not support the statements made (some examples are given under the specific comments below).

Considering these major shortcomings, I do not consider this paper acceptable for publication. In order to become acceptable for publication the authors would need to make significant improvements to their method to address the following major concerns:

1) Account for errors in the spatial distribution of the prior emissions. This would mean solving scalars for the emissions (or for the emissions themselves) for each grid cell or subregions of the inner domain.

2) Improve the estimate of the background concentration to account for the origin of the air masses when entering the inner domain

3) Account for uncertainties in the observed and modeled concentrations

4) Address concerns about the model representation of concentrations at the KCMP site, e.g., the vertical layer selected to represent the observations

5) Include observations also from the NOAA sites to help constrain the emissions

Specific comments:

P1, L18-19: EDGAR agricultural emissions (direct and indirect) have no seasonality, therefore, it is expectable that during the peak emission season, the inventory emissions will be much lower. However, what is important is how the inventory compares to the annual mean emissions.

P1, L17: How were the emissions optimized? This should be stated in the abstract.

P1, L19: The authors state the "total emissions" which would imply the integrated

emission (i.e., in units of mass) but give the emission in units of flux. Rather they should say that this is the “mean” emission.

P1, L21: Do the authors refer here to their optimized fluxes? It is not clear. If they are referring to the optimized fluxes (which are higher in June than EDGAR) this does not indicate that the IPCC emissions are underestimated. Firstly, EDGAR is an independent inventory and is not the same as what is reported to the IPCC (also note the IPCC does not estimate emissions). Secondly, only annual mean emissions are reported to the IPCC, so it is not possible to compare the optimized emissions (based on about 80 days) with the annual mean. Lastly, the fact that concentrations simulated using the optimized emissions (i.e., based on the comparison with observations at KCMP tower) compare well with NOAA observations does not necessarily mean that the fluxes are an improvement. This depends on the footprint of the NOAA observations and if there is a notable improvement compared to using the prior, i.e., EDGAR.

P1, L27: ODP needs to be specified the first time

P1, L28: May refer to Prather et al 2015, lifetime of 116 years

P1, L36: Both references given are only about European emissions (not about North American or the Corn Belt). Please either change this sentence use references about the Corn Belt.

P2, L2: There is nothing empirical about the equations used in inverse methods (i.e., top-down methods). The optimization can be done in various ways but always based on statistical theorems, e.g., Baye’s Theorem.

P2, L22: This may be true of STILT but it is not generally true of Lagrangian models. Lagrangian models can also be used to simulate fields of mixing ratios in space and time. Please correct this statement.

P2, L24-25: This is not true. The reference Corrazza et al. presents a Eulerian model based inversion study for Europe and there are multiple other studies, e.g., Bergam-

aschi et al., 2015.

P2, L31: “Empirical” is not correct in this context. Empirical must be observation/ experiment based.

P4, L17: Changes in tropospheric N2O mixing ratio depend on the surface fluxes but also the atmospheric transport. Please add “atmospheric transport”.

P4, L19: Vertical diffusion does not contribute much to the vertical mixing of N2O, but rather turbulence in the BL and convection. Please correct this.

Fig 1: By “default emission” do the authors mean “prior emission”. If the prior is meant this should be stated (and not “default”).

Eq. 1: Since N2O is inert in the troposphere, the transport is linear (and can be defined as a linear operator) and since the authors are considering the change in mixing ratio with respect to a background, why is “a” not equal to one? In other words, why should Mc not be equal to Mf?

P4. L35-37: The background defined for the observations at KCMP tower should be equivalent to the background in your model. The modeled background mixing ratio is that modeled using natural soil and non-agricultural emission over the whole domain plus some initial boundary mixing ratio (which is not discussed). However, the mixing ratio observed at NWR could be very different from the model background. NWR is a high altitude station (>3000 m above sea level). The authors should re-do the calculations using a background comparable to their model background. Also, they should state what was used for the initial mixing ratio at the model outer boundary. Was this based on global model simulations or other?

P5. L1-2: The authors mean that the observed enhancement should match the difference between the default and background simulations?

P5. L2-3: The disagreement could also be due to errors in the assumed background based on NWR (see comment above).

P5. L3-5: The model errors are critical to the results obtained and need to be accounted for in the method. It is not acceptable to leave the model errors to the discussion.

P5. L5-12: The method presented assumes that the adjustment needed to the prior fluxes is constant throughout the inner domain. This requires a further assumption that the change in mixing ratio at KCMP is equally sensitive to all fluxes in the inner domain. This assumption needs to be stated as it is a very important assumption and has large implications on the results obtained. (This is known as "aggregation error" in inverse problems). A more rigorous approach would be to estimate an emission-scaling factor for sub regions within the inner domain, e.g. based on the grid cells of WRF-Chem, which would reduce the aggregation error.

Fig. 2a-c: I am surprised that the correlations are so low for the comparison of scaled and default simulated mixing ratios. Can the authors please explain why the correlations are not much higher? Also, it should be stated in the caption that figures a-c are using an emission-scaling factor of 25.

Fig. 3. and P6, L25-35: The correlations even with the optimized fluxes are quite low. Did the authors examine different model layers to see which one best represents KCMP observations at 185 m above ground level? Are other species measured at KCMP, e.g., CH4 or CO? If so, how well does WRF-Chem model capture the variability in these? This could tell you about the general performance of your transport model.

P7, L14-18: Turbulent mixing is the main process how tracers are mixed in the boundary layer, diffusion plays only a minor role, if any.

P7, L38: The authors should also state the local time for the data selected since this is what is relevant in the selection.

P8, L9-11: This is rather an indication that the background (based on NWR) is far too simplified. The background calculation does not take into account where the air

masses are actually coming from into the domain, neither the altitude nor the horizontal direction.

P8, L11-15: The variability in the observations should not be considered as a problem, but rather as information. The fact that the model cannot capture the variability shows that there is a problem in the model, in the fluxes and/or the transport.

P8, L18-20: Low variability of the observations (or small enhancements above background) do not limit their usefulness in inversions. Rather, it is also important information, which indicates that the emissions are low during these periods. It does require, however, an accurate background estimate (which is just as important in periods of high emissions). Furthermore, the variability of the observations gives important information about the sources, as the atmospheric transport is not constant in time, i.e., changes in atmospheric transport mean that the sensitivity of the observations to different areas changes in time. Lastly, why were the NOAA observations not used in the optimization as well?

Section 4.2: An important analysis that is missing here is which model layers best represents the observations at the three heights on the tower, especially 185 m height from which observations were used for the optimization. Since the WRF-Chem model overestimates the vertical gradient between 32 and 185 m both during daytime (factor of $\sim$8) and nighttime (factor of $\sim$4) there appears to be an issue with vertical mixing between the layers used to represent the 32 and 185 m heights. It seems that the vertical mixing is too weak and that fluxes at the surface are not adequately mixed in the BL up to, e.g., 185 m, such that the model underestimates the concentrations at this height.

P9, L21-25: The statement about the Miller et al. (2012) study is not correct. Miller et al. use both a geostatistical and a Bayesian inversion for the optimization with a spatial resolution of 1x1 degrees. They do not fit a single scaling factor for all of the central US.

Technical comments:

Fig. 2 caption: delete “of” after “relationships.

P6, L25: “noise” is always singular.

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