

Interactive comment on “Simulation of atmospheric N₂O with GEOS-Chem and its adjoint: evaluation of observational constraints” by K. C. Wells et al.

Anonymous Referee #1

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General comments

This study presents the development of a new inversion framework for N₂O fluxes based on the atmospheric chemistry transport model, GEOS-Chem and its adjoint. Using this new framework, the authors examine the constraint provided on the surface fluxes as well as the stratospheric losses of N₂O by different observational datasets, including ground-based and aircraft datasets. The authors also examine how the well different regions of the globe can be constrained with the current observation network and provide indications of where additional observations could greatly improve the constraint. Overall, the paper is well written and scientifically sound, therefore, I recommend it for publication after a few minor comments and corrections have been

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addressed.

Although the paper is generally well structured, it does appear to jump around a bit when describing the various tests that were carried-out. For example, at the beginning of section 5 (P5378), it would be helpful to include a list of the types of tests (in addition to Table 3), or at least just mention that there are also tests considering temporally varying errors in the prior parameters. Another point that I found confusing was whether or not transport (and measurement) errors were added to the pseudo-observations, as the impact of these is mentioned later in section 5.2 (P5383).

Specific comments

P5370, L20: please change this to: “Agricultural activities such as fertilizer application and animal waste management increase the substrate available for nitrification and denitrification pathways leading to enhanced...” as by writing that the reactions are enhanced suggests increased rates of reaction.

P5371, L5-8: suggest that the authors also include reactive nitrogen substrate in this list, as it is one of the most important determinants of N₂O emissions

P5374, L8: by “loss frequencies” do the authors mean photolysis cross-sections or other, please clarify.

P5375, L23: by enforcing a minimum value of the posterior scaling parameters of zero implies that the fluxes cannot change sign in the inversion, i.e., a negative flux cannot become positive and vice-versa. Is this what the authors mean? In which case it is not quite correct that regions with a prior negative flux (e.g. over the ocean) cannot become more negative, but rather that they cannot become sources?

P5375, L19: does the size of the state vector apply to the two-year inversion period? Given that the number of elements for the stratospheric loss parameters is 192, I would be presume so, but it would be helpful to state this here.

P5376, L6: could you please give the order of magnitude of the transport errors cal-

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culated? This would be interesting to know, especially in connection to the influence of the transport errors on the ability for the inversion to simultaneously optimize the emission and loss parameters.

P5378, L23: do you add any random noise to the pseudo-observations, and if so, was this consistent with the error characteristics of the observation error covariance matrix?

P5378, section 5: by adding a spatially uniform error to the prior values of the scaling parameters, you are testing the ability of the inversion to correct a uniform bias, which effectively means one degree of freedom. However, it would be also a useful test to see how well random spatially distributed errors can be corrected.

P5380, L1-5, did the authors examine how the correction to the biased a priori scaling factor varied from season to season? I would expect that there would be some dependency on season as well due to the seasonal variation stratosphere-to-troposphere (STT) mixing on the tropospheric mixing ratios of N₂O. This would be useful information to include.

P5382, L5-10, I think here it is important that the authors make a distinction between the lower and upper stratosphere. The vast majority of the loss of N₂O occurs in the upper stratosphere, therefore, the influence of a bias in the loss will only be seen in the troposphere (where the nearly all the observations are made) after the time delay for transport from the upper stratosphere to the troposphere, which is long, i.e. 1-2 years. However, mixing from the lower stratosphere to the troposphere can occur on shorter timescales of weeks to months.

P5394, L1: although in the future, satellite retrievals of N₂O may reach the precision and accuracy needed to help constrain N₂O emissions, current retrievals and instruments are not at this level: the error on N₂O retrieved from AIRS is about 7 ppb in the troposphere (for comparison, this is more than 3 times the inter-hemispheric gradient in N₂O). At present, the AIRS satellite retrieved may be helpful in addition to ground-based observations for, e.g., establishing the vertical profile of N₂O for the ini-

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tial conditions but not in solving for surface emissions of N₂O.

Technical comments

P5369, L8: replace “aboard” with “on-board”, i.e., the adjective

P5370, L13: 100-year

P5370, L15: “those of any other ozone depleting substance”

P5370, L16: replace “reactions” with “pathways” as nitrification and denitrification each involve a series of reactions

P5375, L23: “. . .ocean regions with a net N₂O uptake are not stronger sinks than in the prior. . .”

P5377, L25: please put the phrase in brackets “(in general. . .)” after “lowermost stratosphere” to make the sentence easier to understand

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