Reply to Referee 1

We thank the reviewer for his/her very helpful and thoughtful review and respond to each point raised below.

1. General comments

The authors studied the estimates of N2O emissions from five inversion frameworks. They are doing a rigorous study imposing the same constraints on each inversion system (priori emissions and N2O observations). They first compared the atmospheric N2O as simulated by the systems using the posterior fluxes. Then they compared the results of the inversion systems in terms of posterior emissions. Using 5 systems is one of the strength of this paper. This allows to highlight the parameters that influence the results of the inversion process. That also allows to produce the uncertainty associated with the estimated emissions. The discussion of the results is very well conduced. The comparison of the result with results from independent study is welcome. The list of key results in the summary is appreciated. Overall, the paper is suitable for ACP.

2. Specific comments

page 5273, line 1: The abstract is focused on the N2O emission estimates. One line could be added on the validation with atmospheric observations.

We have now added the following sentence to the abstract: "The posterior modelled atmospheric N_2O mole fractions are compared to observations to assess the performance of the inversions and to help diagnose problems in the modelled transport."

page 5273, line 3: It could be useful to precise that the atmospheric inversion are performed using chemistry transport model (CTM). This would explain for example why they can use different meteorological data (next line).

We have now added that the inversions are based on CTMs.

page 5275, line 21: "posterior emissions" should be defined so that all readers could understand that these are the result of the inversion process.

We have now specified that the "posterior emissions" are those resulting from the inversion.

page 5277, lines 4 to 9: Iterative descent algorithms do not necessary require the gradient (even if this is usually the case for geophysical applications). Please rephrase.

We have now changed line 5 to the following: "Variational methods find the optimal **x** using an iterative descent algorithm, usually requiring calculation of the gradient of *J* at each iteration and do not require *H* to be a matrix operator"

Lines 12 and 13 should be changed accordingly. Analytical methods can also work

*with a non linear operator H as soon as the operator is differentiable and its linear **H**
is defined. Then Eq. 3 can be:*

$$x = x_b + BH^T(HBH^T + R)^{-1}(y - Hx_b)$$

*An outer loop can then be implemented in order to have a better approximation of **H**.
Please rephrase the paragraph.*

We have now added the following sentence immediately following Eq.3: "Analytical
methods can also be used in the case that *H* is non-linear if it is still differentiable and
that the linear **H** can be defined over a small range of **x**, in which case, an outer loop
is also required to better approximate **H**."

page 5277, line 10: Eq. 2 should be:

$$x = x_b + (H^T R^{-1} H + B^{-1})^{-1} H^T R(y - Hx_b)$$

Yes, thank you for pointing out this typo.

*page 5277, lines 10 and 11: Eqs. 2 and 3. The left hand side of the equation could
be x_a instead of x and you could specify that x_a is the posterior emission.*

We define x_b to be the prior emissions and x to be the posterior emissions.

page 5277, lines 15 to 19: The sentence is too long and not clear. Please clarify.

We have now changed this sentence to: "For inversions in the second category, the
chemistry-transport operator, **H**, represents the sensitivity of the observations to the
fluxes in each of a given number of predefined regions. Each column of **H** can be
found by running the CTM perturbing the fluxes in a given region and determining
the resulting change atmospheric mole fraction for all observations."

*page 5278, lines 2 to 7: For people that are not familiar with inversion, it is difficult
to understand why the methodology needs a spin-up period and why the last year is
not well constrained. Please add a reference that explain this or provide justification.*

We have now changed this paragraph to include a brief explanation of why we need a
spin-up period and why we do not include the last year in the emission totals:
"All inversions were run for the period 2005 – 2009 but only output from 2006
onwards was analysed as 2005 was used as a spin-up year. A spin-up period is
required to minimize the influence of the initial conditions on the posterior emissions.
Here, we chose 1-year for the spin-up as all models started with their best initial
conditions estimates established after previous longer integrations of the CTMs. Also,
when presenting mean emission results, the years 2006 – 2008 are used, as the end of
2009 is not as well constrained in the inversions (to constrain the end of 2009,
observations at the beginning of 2010 would need to included and these were not all
available at the time of preparing this study)."

*page 5281, lines 6 to 8: It's not clear to me how the "observed annual mean
meridional gradient" was derived from the observations. Is the explanation can be
found in the following sentence? Or does the following sentence explain how the
gradients are computed from the model? Please add more details on how the
gradients are computed (from both the observations and the models).*

The meridional mole fractions were calculated from the observations and the model simulations in the same way. In the models, the mole fractions were extracted at the sites corresponding to observations and, likewise, detrended and deseasonalized, and then finally averaged to give the annual mean at each site along a meridional transect.

page 5281, line 11: Please clarify if the agreement is for all the models. Does the correlation coefficient R2 > 0:9 mean that the correlation coefficients between each model and the observations are all greater than 0.9? Moreover, instead of "observed gradient at the surface", wouldn't it be better to use the term "gradient derived from observations at the surface"?

We mean that the R-squared value is at least 0.92 for each model, we have tried to make that clearer in the text. We have changed the sentence to: "For each model, a very good agreement was found with the gradient derived from surface observations (correlation coefficient $R^2 \geq 0.9$ for each model)".

page 5281, lines 11 to 14: Why do you discuss the case of MOZART4-I and you don't discuss the results of all the other models?

We compare all the models together with the observations and then mention that MOZART4-I has the anomaly of the offset. The other models tend to show a similar result, i.e. that they all underestimate the gradient in the total column in January and underestimate the absolute total column in November but better capture the gradient.

page 5283, lines 5 to 7: Do you mean that at MLO, all CTMs simulate a too early minimum, as it was also the case from the simulations using the a priori emissions? Please rephrase. Please also rephrase the second part of the sentence with for example:
"The amplitude of the seasonal cycle in the simulations using the a posteriori emissions is closer to the observed amplitude (compared to the amplitude from the simulations using the prior emissions)."

We have now rephrased lines 5 to 7: "At MLO, all CTMs simulate a too early minimum as was also the case using the a priori emissions. However, with the a posteriori emissions, the amplitude is closer to that observed."

page 5284, lines 23 and 24: The a priori is not plotted Fig. 5. This makes the last part of the sentence difficult to see ("close to that a priori"). Would you please add the a priori map of N2O fluxes on Fig. 5?

We have now added the prior emissions map to Fig. 5.

page 5286, line 10: Could you add a horizontal line on each panel of Fig. 9 with the median value for the region?

We have now added a dashed line to Fig. 9 indicating the median of the posterior emissions.

pages 5291 to 5293, summary and conclusions: One useful information you could

add is a summary of the parameters that influence the most the inversion. For example the inter-hemispheric exchange time is one of them as discussed Sec. 3.2.1. The way the observation are assimilated (monthly/weekly means versus sampled time) is another one as also discussed Sec. 3.2.1. Moreover, Sec. 3.2.2 explains the difference in the posterior estimates by a lack of observation in some regions. Would it be possible to add a paragraph on where we should have more data in order to reduce the uncertainty in the estimations?

We have included two additional "salient results" to the list in the Summary and Conclusions to include the dependence of the results on the inversion method and transport model:
- the largest uncertainties were found in the estimates for South and Tropical America and South Asia owing to uncertainties in the modelled atmospheric transport and to the poor observational constraint for these regions [already included]
- differences in the meridional distribution of emissions among the inversions were also found to depend on the inter-hemispheric mixing rate in the CTMs
- assimilating monthly mean observations from flask sampling networks most likely leads to an underestimate of the emissions

We have also added a phrase about where new observations should have more observations: "considerable uncertainties remain, especially in the less well-constrained regions of South Asia and South and Tropical America. These regions also appear to be very sensitive to uncertainties in the modelled atmospheric transport and are regions that should be targeted for new observation sites."

3. Minor revisions/comments

page 5275, line 22: "analyse the posterior emissions" instead of "analyse the emissions"

Done.

page 5275, line 23: "and investigate their cause" instead of "and their cause"

Done.

page 5277, line 2: "the minimum of the cost function (Eq. 1) is searched" instead of "is found".

Done.

page 5277, line 2: "Approaches for finding x that minimises" instead of "Approaches for finding x to minimise"?

Done.

page 5277, line 15: Pleas insert "(see Table 2)" at the end of this sentence.

We have included "(see Table 2)" at the end of line 15 instead.

page 5277, line 19: "changed" instead of "change"?

We consider "change" to be correct in this context.

page 5279, line 24: "nmol.mol   1" instead of "nmol mol   1"?

page 5283, line 12: "the latitude of MLO" instead of "this latitude".

Done.

page 5284, line 18: "MOZART4-I" instead of "MOZART-I".

Done.

page 5284, line 23: "global spatial distribution" instead of "global distribution"?

page 5284, line 28: " i.e. the annual mean posterior flux minus the annual mean prior flux" instead of "i.e. the mean posterior minus prior flux"?

Done.

page 5286, line 8: "there is" instead of "is there"?

We consider "is there" to be correct in this phrase.

page 5286, line 10: "plus or minus MAD" instead of "and MAD"?

Done.

page 5299, Tab. 1: Could you centre "Resolution" on top of "spatial" and "temporal" in the header of the table? Same for Tab. 2.

Done.

page 5304, Tab. 5: the NIES and MPI-BGC do not appear in Tab. 4.

NIES and MPI-BGC are included in Table 4.

page 5307, Fig. 1: "atmospheric N2O observations" instead of "atmospheric observations"?

Done.

Could you have two panels for this figure, one with the global map and one with a zoom over Europe as it seems that there's a lot of sites there?

While it is true that there are more sites in Europe compared to other regions, we prefer not to add a subplot for Europe only as this is a global study, and having a subplot for a single region would tend to suggest that special focus was given to that

region, which was not the case.

page 5308, Fig. 2: "with gradient derived from surface observations" instead of "with surface observations"?

Done.

page 5309, Fig. 3: Could you please add a title at each panel. For example: "Surface / Jan.", "Surface / Nov.", "HIPPO / Jan.", "HIPPO / Nov.".

Done.

page 5310, Fig. 4: The X-axes of the panels have no label.

Done.

page 5313, Fig. 7: Do you need to have your colour bar ranging until 0.4? Looking at the plot, the value of 0.3 should be enough.

Done.

page 5315, Fig. 9: You could add where to find the regions in the caption with for example: "for the 7 land regions (2 first rows) and 3 ocean regions (last row)". Same for the next figure.

Done.

page 5320, Fig. A3: You could change the limits of the Y-axis with respectively 0.5 and 1.1 ppb.yr-1.

Done.