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***Interactive comment on* “Sensitivity of inferred regional CO source estimates to the vertical structure in CO as observed by MOPITT” by Z. Jiang et al.**

Z. Jiang et al.

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We thank the reviewers for their thoughtful and detailed comments. Below we respond to the individual comments. In addition to the revisions discussed below, we made significant modifications to the manuscript. Four figures were removed to make the paper shorter and more concise. We have also moved the discussion about the optimization scheme and the validation of the MOPITT data to appendices to make the manuscript easier to read.

This manuscript presents sensitivity studies of top-down estimates of regional CO

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sources to: a) differences in the information content between profile retrievals and surface retrievals, and b) differences in model representation of OH spatiotemporal distribution. The premise of these sensitivity studies is that errors in modeled vertical structure of CO (and assumptions of OH) translate to errors in inferred CO sources, especially when investigating local-to-regional emissions. While I commend the authors for tackling this issue (which is certainly challenging), this issue is not something new. As noted by the authors, several studies have reported these errors (including previous studies by the authors themselves, as well as inversion studies in the CO₂ community). It begs the question whether this manuscript provides a unique contribution to inverse modeling studies. There are certainly interesting dimensions (or components) of the problem that requires attention which will help the community to improve accuracy in emission estimates. The manuscript however focuses (at least from the reviewer's point of view) on comparisons and sensitivity, which is already known to account for the major portion of the systematic uncertainties of the source estimates.

As the reviewer correctly indicated, the effects of biases in OH and convective transport on CO inversion analysis have been reported by several previous studies. For example, in our previous work, Jiang et al. (2013), we compared the CO source estimates in June-August 2006, inferred from different MOPITT datasets to study the influence of convective transport.

The key question is how to mitigate the effect of biases in OH in CO inversion analyses? One approach is to use observations of CO near the source regions, i.e. surface data, to minimize the influence of chemical ageing on the emitted CO. The surface level (or lower tropospheric) MOPITT multi-spectral retrievals represent one such dataset. Indeed we showed that the impact of OH distributions over contiguous United States was reduced from 64% to 40%, by using MOPITT surface layer data. As far as we are aware, this is the first inversion analysis using the surface MOPITT level data to produce regional source estimates over the annual cycle. Our results also showed the CO emission estimates for North America and Europe are more sensitive to OH

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biases, compared to those from Asia, which we attributed to the timescale for transport of air from these continental regions. Previous studies (e.g. Stohl et al., 2002) have looked at the timescale for transport of air masses from different continental regions, but our analysis is the first to interpret the inversion results in the context of the transport timescale. Our results suggest that developing age of air metrics might be helpful for understanding differences between inversion analyses from different models. We have added text to the conclusions to more clearly make this point.

The reviewer recommends a major revision for this manuscript. Overall, the reviewer finds this manuscript to be a bit confusing, unclear, and unfocused. Please see specific comments for details of major concerns.

(1) Title: It is unclear whether the author is referring to the sensitivity of inferred regional source estimates to the ‘modeled’ vertical structure. First of all, the reviewer suggests using ‘top-down’ rather than ‘inferred’ since there are other means of inference that doesn’t involve inverse modeling. Second, it is not that the vertical structure of CO as seen from MOPITT is wrong, the sensitivity is due to the fact that the modeled vertical structure is not represented accurately (and that this error in the model is not represented in the inversion accurately) leading to errors in the estimates. The reviewer suggests modifying the title.

Thanks for your suggestion! The title has been changed.

(2) p. 1 line 14. What do you mean by ‘signals’?

Here we mean concentration of the tracer gases. It has been changed.

(3) p. 1 line 15-16. sensitivity . . . to the ‘modeled?’ vertical CO distribution

Changed.

(4) p. 1 line 17-18. Suggests to use consistent terminology (to avoid unnecessary confusion) on ‘assimilation’ and ‘inverse analysis’.

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We note that “inversion” and “assimilation” refer to the same mathematical approach in the context of the manuscript. We are using a 4-D assimilation system, but because we are optimizing a model parameter, it can be referred to as an inversion analysis. Nevertheless, we have modified the manuscript to keep the terminology consistent.

(5) p. 1 line 19-20. a reduction . . . and an increase . . . relative to ???

It is relative to a priori emission estimates. Changed.

(6) p. 1 line 21. . . . suggesting an overestimate of the a priori isoprene source of CO. . . Is this due to errors in modeled vertical structure (that is unaccounted for) rather than sources (e.g., isoprene oxidation). It is unclear (even upon reading the text) that it is possible to tease out (or disentangle/attribute) this discrepancy.

A very good question! We believe that it is due to both. We are clearly not capturing the vertical structure, and as we noted in the manuscript, previous studies have documented the overestimate of isoprene in the version of MEGAN employed in study. Unfortunately, we cannot disentangle the effect of the bias in isoprene emission and vertical CO distribution within the framework of this work. This is why we suggested: “assimilating a broader range of composition measurements to provide better constraint on tropospheric OH and the biogenic sources of CO is essential for reliable quantification of the regional CO budget” at the end of abstract.

(7) p. 1 line 25. ... discrepancies in convective transport in the model ... How do you know this? Please cite or show.

This is the abstract, we would prefer not to include a citation in the abstract. We now cite in the text the work of Ott et al. (2009) who looked at the sensitivity of the distribution of trace gases to the parameters in the GEOS-5 convection parameterization.

(8) p. 2 line 26-27. . . . from the CO profiles were significantly higher than those estimate from the surface layer retrievals during . . . Does the CO profiles also include the surface layer retrieval? What is the reason behind using only the surface layer

retrieval? Shouldn't the default be using the profile or retrievals with at least 2 pieces of information (TIR – free troposphere and NIR –surface). The reviewer understands that some retrievals are derived from TIR radiances and a comparison of information content between retrievals is informative in itself but it appears it is not the focus of this manuscript. If it is, please state/describe it explicitly.

The surface layer retrieval is also included in the MOPITT CO profile.

As shown in Figure 1 of Jiang et al. (2013), the surface layer retrieval of MOPITT v5 has strong sensitivity in lower troposphere over continent: the sensitivity peaks at 900 hPa. In Jiang et al. (2013) we looked at the impact of convective transport errors in the context of assimilating the surface level and profile retrievals. Details of the two datasets are discussed in Jiang et al. (2013). We are interested in using the surface level data in this study because the CO distribution in the free troposphere is more susceptible to model bias, from transport and the OH distribution. We hope to improve the reliability of the CO source estimates by using measurements in lower the troposphere, closer to the CO sources.

(9) p. 2 line 29-33. ... vertical transport of air from the North American and European boundary layer is slower than from other continental regions... and North America and Europe is more chemically aged . . . Can this be just due to errors (bias) in model transport (i.e., issues of representing frontal systems or synoptic meteorology or even mesoscale convection)? If so, it is unclear if we can make some conclusions on relative age of air unless when compared to observed tracers.

As the GEOS-Chem model is driven with reanalysis data, the descriptions on the large-scale transport should be reliable. Indeed, reanalyses such as GEOS-5, MERRA, and ERA-Interim provide the best description of the large synoptic features. This has been demonstrated by the use of these analyses in aircraft campaigns. However, as the reviewer indicated, the descriptions on the small-scale processes could be problematic, because of the coarse-resolution simulation in this work. For example, the altitude of

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a plume exported from North America due to frontal lifting could be biased because of the coarse resolution.

We note that we have a companion paper that we are about to submit, in which we constrained the North America CO emission with higher resolution, $0.5^\circ \times 0.667^\circ$, the native resolution of GEOS-5, to mitigate some of the issues that might arise as a result of the coarse resolution of the work in this manuscript. In that analysis we obtained results that are consistent with those in this manuscript.

We also note that this issue of the difference in transport over the continental regions has been shown previously by other studies, such as Stohl, et al. (On the pathways and timescales of intercontinental air pollution transport, J. Geophys. Res., 2002). They looked at the transport of idealized tracers from the main continental source regions. They pointed out that because there is less cyclogenesis and less vigorous convection over Europe, “emissions from Europe tend to remain in the lower troposphere.” They also found that “in terms of vertical transport, the North America tracer... behaves intermediately between the Asia and Europe tracers.” Our analysis was motivated by this earlier work. We now quote the Stohl et al. (2002) study in the revised manuscript.

(10) p. 2 line 42-43. . . . should the implication be more towards the use of vertical profile datasets?

Ideally, vertical profiles would be better because they provide more information. However, if the model itself has biases in transport or in the chemical sink of CO, inversions rely on middle troposphere measurements may be problematic. In that case, surface (or lower tropospheric) data would be better, assuming the spatial coverage of the data is good and the representativeness errors in the model are small (which is not the case for the in situ point measurements from the surface network).

(11) p. 3 line 59. . . . included in the inverse analysis of CO₂ (sources and sinks?) . . .

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Changed.

(12) p. 3 line 70-71. Suggestion: in model parameterization of convective transport, chemical sink of CO, and long-range transport.

Changed.

(13) p. 4 line 74. What do you mean by CO signals?

It is actually the CO concentration. Changed

(14) p. 4 line 91-94. Please rephrase. Why would errors in CO accumulate in the free troposphere? Also, if the manuscript focuses on convective transport, shouldn't CO be more mixed across the layers? It would be informative to show vertical/horizontal distribution of OH since the two versions of OH distribution may not only be different in the vertical but also near/over source regions and downwind.

The manuscript does not focus on convective transport. Hopefully, that is clearer in the revised version.

The OH abundance generally peaks in the mid-troposphere. As shown in the new Figure 6, there are large differences between the OH concentrations throughout the free troposphere. Considering the lifetime of CO, most CO will be destroyed in free troposphere rather than boundary layer. Thus, free tropospheric CO will be more susceptible to the discrepancies in OH, after long-range transport in free troposphere. The discussion has been clarified.

Two new panels are added to Figure 6 to show the vertical structure of OH.

(15) p. 5 line 99. . . . Are section 4 results using pseudo data still?Á

Thanks for catching this! The OSSE is only employed in Section 3. This has been changed and the discussion containing the OSSE moved to Appendix A.

(16) p. 6 line 114-118. . . .true (actual) atmospheric state . . . please qualify that z

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here is in fact the true layer averaged CO state at MOPITT grid levels.

Changed.

(17) p. 7 line 151-152. Please elaborate on biogenic vocs (i.e. MEGAN versions) since this is discussed later on.

Thanks for your suggestion! The description of MEGAN inventory has been added.

(18) p. 8 line 162-164. Please make N and i in y_i italic (consistent with eq 2). $F_i(x)$? y_i is a vector of observed concentrations at a given time (does this mean also at a given space \hat{A} horizontal and vertical?)

Yes, at a given MOPITT observation location in space and time. The description of Equation (2) has been rewritten.

(19) p. 8 line 164. . . .which represents the transport of the CO emissions . . . suggest qualifying this since $F(x)$ represents not only transport but also chemistry.

Changed.

(20) p. 8 line 165-167. . . . is the a priori estimate . . . (of what?). also, please add dimensions of S_e and S_a so it becomes clearer.

It is the a priori estimate of CO emissions. Changed.

(21) p. 8 line 169. . . .but is a set of scaling factors S such that $x = \sum x_a$. Is S σ ?

Thanks! The scaling factor is expressed as σ here. Changed. This discussion has been moved to Appendix A.

(22) p. 8 line 175-178. Why does eq 3 assume that the uncertainty in the emissions is normally distributed about scaling factor one? Please elaborate. Is this part of S_a ? What is S_a ? Why is there a mention of statistical distribution when in fact the previous discussion is about a cost function? Is x considered a random variable?

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Thanks for your suggestion! The description about Equation (2) has been rewritten to provide more information about the inversion approach. We also refer to reader to Henze et al. (2007) and Kopacz et al. (2009, 2010) for more details about the GEOS-Chem 4D-var approach.

Inherent in the 4-D variational data assimilation framework is the assumption that the error statistics (for the state and observation vectors) are Gaussian. Because we use scaling factors rather than emissions in the inversion, the errors on the scaling factors is normally distributed about the a priori scaling factor (which is assumed to be 1).

(23) p. 8 line 177. . . .because it allows (the) negative emissions. . . .

Changed.

(24) p. 8 line 183. . . .reduce negative gradients effectively . . . please elaborate the meaning of 'negative gradients' and 'effectively'. p. 9 line 184-190. Why is there a problem with partially offsetting the decrease in gradient? Would this just be increasing the number of iterations to find the minimum? Please clarify. Also, it might be good (for the ms to be more concise) to move the discussion of this transformation and OSSE to a supplement or appendix.

An example has been added to elaborate the difference between two methods. As shown in the OSSE, the convergence speed for negative gradient is much slower in the LOG scaling factor optimization. Ideally, more iteration could solve this problem.

We believe that it is important to keep this discussion in the paper to ensure that it is documented in the literature for other users of the model. We agree with the reviewer, however, that would be best to move it to an appendix, which we have done.

(25) p. 9 line 186-190. . . . Please elaborate on OSSEs. What do you mean by CO emission unchanged? And . . .we reduced the CO emission by 50%. What do you mean by ...whether the scaling factors can return to true state (1.0). Scaling factors are not exactly the state.

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The “CO emission unchanged” means the model was run with the default CO emission inventory. “Emission reduced by 50%” means that the emissions were reduced to 50% of default value. More description has been added.

(26) p. 9 line 191-199. Why would there be different treatment of minimization? Should there be consistency in this regard? The reviewer is concerned (as also noted by the authors) that there is inconsistency in the error statistics and assumed error covariances and basic assumption of Gaussian distribution (if this methodology is viewed as similar to Bayesian inversion framework rather than purely variational scheme).

A very good question! The major reason is that both linear and LOG optimization approaches have limitations in application, although they have consistent error statistics.

For the linear optimization, we have to add the lower bound, otherwise it will lead to negative scaling factor, which is unphysical. The cost function is minimized with BFGS method. When there is no bound, it will optimize the strong gradient regions first and the optimization of weak gradient regions depends on the adjustment of strong gradient regions, as shown in the OSSE.

However, when there is a bound, all grids with positive gradient will be adjusted to the bound, zero, and then be adjusted backward gradually. It is reasonable under ideal conditions, however, because the grid boxes with weak gradient have the same a priori constraints as those with strong gradient, the a priori penalty will be too large in the beginning of optimization, which will become an obstacle during future iterations.

For the LOG optimization, we have shown that they do not reduce the negative gradients effectively in the OSSE. We also observed this phenomenon in actual inversion: the inversion kept trying to reduce the large negative gradient but could not, which led to a “false convergence”, because the cost function could not be reduced furthermore.

The LOGX2 method is the result of our effort to mitigate the issues of the linear and LOG optimization in the analysis. We understand the issue with the error statistics.

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However, because of the numerical limitations of the linear and LOG approach, and the performance of LOGX2 method in both the OSSE and the true inversion, we feel that it is an acceptable compromise, in the context of the BFGS algorithm. It may not be needed in the future if a more robust optimization algorithm is found to replace BFGS.

(27) p. 9 line 198 . . .stati(sti)cs.

Changed.

(28) p. 9 line 203. Please qualify the rationale behind 5×10^{17} threshold.

Given the variability of CO, such low column abundances are rare (see Fig 5 of Kopacz et al., ACP, 2010). Typically, such low CO columns might be found over the Antarctica or over the Tibetan Plateau. For example, for May 2005 the minimum CO column estimated in our model was 6×10^{17} cm⁻², located over Antarctica. The global mean column was 1.5×10^{18} cm⁻². The 5×10^{17} cm⁻² threshold was simply chosen to filter out retrievals with unrealistically low CO that might adversely impact the analyses over the source regions. We now state this rationale in the revised manuscript.

(29) p. 10 line 207-212. . . .which assumes that the mean differences between the model and observations are due to discrepancies in the emissions . . . The reviewer disagrees. The reviewer argues that the mean differences can be also attributed to systematic bias (especially this study on vertical transport) of the model. In fact, the treatment of the observation error here should be improved to account for this systematic bias. And if represented accurately can account for most of the differences in the top-down estimate discussed in this manuscript.

p. 10 line 212. ...we expect assumption of a uniform observation error to have a negligible impact on our inversion results. . . The reviewer disagrees. The reviewer thinks that misrepresentation of observation error is the crux of the problem. Note that observation error here should also represent errors in $F(x)$.

A very good question! We agree that systematic errors are an issue. This is why we

didn't use the Relative Residual Error (RRE) method in our work. Constructing the observational error covariance using this approach will convert systematic bias into emission bias, which could be a contributor to the spread in the a posteriori source estimates among different inversion analyses.

As the reviewer noted, to account for these errors they must be “represented accurately.” At present, we do not have a good means for doing so. We have done some work using a weak-constraint 4D-var to try to quantify the systematic errors. It is a promising approach and will be described in Keller et al., (Quantifying Model Biases in CO Emission Estimation Using Weak Constraint 4D-Var, manuscript in preparation). However, for our analysis period, we have no reliable information as to the systematic errors in the analysis. We now explain this in the text.

As regards, the assumption that the observation errors are uniform, Heald et al. (JGR, 2004), in their inversion analysis of MOPITT data, showed that it will impact the source regions that are less well constrained. The large source regions in North America, Europe, and Asia, and the major biomass burning source regions in the tropics should be less influenced by our treatment of the observation error. Clearly, the a posteriori uncertainties (the precision) will be sensitive to the spatial distribution of the observation errors. Our statement in the manuscript was too strong. We have modified the discussion. The observations errors are a combination of the measurement errors and the model errors, which will be dominant. Unfortunately, we do not have a reliable way of characterizing the model errors. We believe the best approach is still the so-called NMC method (Parris and Derber, 1992), as applied by Jones et al. (2003) for CO inverse modeling. This approach uses pairs of model forecasts, of different length, but which are valid for the same time, to characterize the model errors. We do not have such forecasts available to use for this version of GEOS-Chem during this analysis period. Consequently, rather than assuming an ad hoc distribution for the errors, we assume that they are uniformly distributed.

(30) p. 10 line 215. What is the rationale behind assuming uncorrelated errors? Several

papers have reported the importance of this term in the inversion.

Spatial correlation is important. Unfortunately, this version of the adjoint cannot accommodate non-diagonal covariance matrices. In addition, there is the problem of how to obtain a reasonable estimate of the correlation length scale. For example, Heald et al. (2004) used a length scale of 147 km, based on the NMC approach described in Jones et al. (2003); Mukherjee et al. (2011) suggested the correlation in MOPITT data should be larger than 5000 km. Given such vastly different estimates for the correlation length scale, a separate study is clearly required to address this.

(31) p. 10 line 218-224. Please elaborate on how initial conditions (from KF assimilation) are used in the inversion.

We archived the optimized CO distributions from the KF assimilation. The optimized initial condition was read at the beginning of the assimilation period for each monthly inversion. More description has been added.

(32) p. 10 line 225 . . .we will show(n) below. . .

Changed.

(33) p. 12 line 253. What do you mean by free run model?

The free run is the standard GEOS-Chem simulation without Kalman filter assimilation. The initial condition for the free model run is the model original initial condition on June 1 2004, without optimization. More description has been added.

(34) p. 12 line 257. . . .MOPITT data (are these profiles?).

It is the tropospheric profiles. Changed

(35) p. 12 line 267. . . There's a difference between this inversion and Hooghiemstra et al 2012 since the latter used V4 column CO.

We find this comment unclear. Yes, the inversions are different and Hooghiemstra et

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al (2012) used V4 data. The point of the discussion was to note that previous studies suggested a high bias in the southern hemisphere with V3 data. Hooghiemstra et al. (2012) suggested a high bias with V4 data. We are seeing a posteriori CO source estimates that also suggest a high bias with V5J data. We have different inversion analyses, using different versions of the MOPITT data, all pointing to a similar problem. Hopefully, the modified text is easier to follow.

(36) p. 12-13. It can also be argued that the differences (relative to GMD) are due to issues in sub-optimal Kalman filter (i.e. error covariance used to update the surface concentrations).

This is a good point! They are two difference approaches, with different strengths and weaknesses. We note, however, that because the Kalman filter is adjusting the CO distribution, it should better correct errors due to transport.

(37) p. 13 line 292-293. . . .as shown in Figure 5c, the a posteriori emissions. . . . These are scaling factors not emissions.

A note has been added for clarification.

(38) p. 14 306 . . . reduced (by) 32%.â€”

Changed.

(39) p. 14 305-314. How about fires? Is there a compensating effect of fires and biogenic emissions? What is the impact of inaccurate injection heights?

As our answer for question 6, we believe biogenic emissions were overestimated. An example is the emission reduction in southeast US, where we have strong biogenic emission but low biomass.

We agree with the reviewer that biomass burning play an important role in South America, Central Africa and Indonesia. If the biomass burning source and the biogenic emissions have similar spatial distributions, it will be impossible to distinguish them by

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using only CO. Consequently, it will be possible for there to be compensating effects, as indicated by the reviewer. This was discussed in Jiang et al. (2011) and, as a result, we do not try to independently optimize the biogenic and biomass burning sources.

The effect of injection height is interesting. Because of lacking of enough data about plume heights, an accurate description of the spatial-temporal variation of injection height is still challenging. In a recent paper, Gonzi et al. (2011) showed that “enhancing the vertical mixing for the biomass burning emissions had a small impact on the a posteriori regions source estimates”, compared to the differences they obtained with different versions of the MOPITT data.

The discussion has been modified.

(40) p. 15 line 320-321. How about transport and mixing?

Using GOSAT XCO₂ column data, we have found that the discrepancy on different boundary mixing schemes has only negligible influence on the CO₂ flux inversion. We assume the response of CO inversion should also be small. However, it would be interesting to check the actual response of CO in future.

(41) p. 15 line 323. What version of MEGAN would this be?

It is MEGAN 2.0. Changed.

(42) p. 15 line 321-322. OH fields are biased high in summer when the CO lifetime is short. CO lifetime is based on loss rate by OH isn't it (or is this residence time)? Please elaborate, especially when compared to line 315-317.

The shorter CO lifetime in summer is because OH concentration is greater than that in winter. In summer, if the lifetime is 30 days, a 25% or 50% bias in the lifetime is significant in the context of a monthly inversion. However, if the lifetime is 200 days in winter, a 25% or 50% bias will have less of an impact on the results because the lifetime is already so long. This was unclear and the text has been removed.

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(43) p. 15 line 330-336. What is the implication of this issue? Please elaborate on the importance of this paragraph.

The biomass burning sources in South America and Southern Africa are both dominant in only in summer and fall, which means the bias in the biogenic emissions will have large influence on tropospheric CO throughout the rest of the year. This text was removed to help shorten the manuscript.

(44) p. 16 line 357. . . .significant(ly) greater.

Changed.

(45) p. 16-17 line 353-371. Please rephrase or simplify. It is currently hard to follow. The reviewer suggests comparing relative changes rather than magnitude since the priors in Kopacz et al 2010 priors and this manuscript are different.

p. 17 line 368-371. Please elaborate as to why there is discrepancy between the results and Kopacz et al. 2010.

Thanks for your suggestion! Not only are the priors different, but we are also using different versions of the data. Kopacz et al. (2010) used MOPITT V3 data (together with AIRS and SCIAMACHY data), whereas we are using V5J. Gonzi et al., (2011) found large differences in the source estimates obtained from differences versions of the data. For example, for North America the estimate a total source of 172. Tg CO with V3 data, and a source of 128.7 Tg Co with V4 data. Furthermore, Kopacz et al used column MOPITT data, whereas we are using the profile. We do believe that the vertical sensitivity of the retrieval influences the inversions, given the information on the sources that is reflected in the vertical structure of the CO distribution. That is the motivation for our analysis. We have rewritten this part to make the manuscript clear and shorter.

(46) p. 17 line 383. ..meteo(tor)rological.

Changed.

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(47) p. 17 line 384. .. Here (the) the biogenic source(s) are combined with the combustion sources and optimize(d) a the resol(u)tion of the model.

Changed.

(48) p. 17 line 387 . . .and optimize(d). . . p. 18 line 403. . . .We beli(e)ve

Changed.

(49) p. 18 line 401-410. Please rephrase or simplify. The reviewer suggests having description of convection and how this would increase emissions from the profile inversions.

Thanks for your suggestion. This figure has been removed.

(50) p. 19 line 418. . . .We (we) performed . . .

Changed.

(51) p. 20 line 439-441. Please elaborate as to why this is a valid conclusion. As noted earlier, this can be just issues with model representation of synoptic and meso-scale meteorology. Unless this is corroborated by observations, this may not be a valid conclusion.

As we discussed above, we do not believe that is unique to GEOS-Chem. We have added a citation to the work of Stohl et al. (2002), who obtained the same results using a Lagrangian particle dispersion model.

(52) p. 20 line 448-451. It would be informative to show vertical structure of OH.

Thanks for your suggestion! Two new panels are added to Figure 6.

(53) p. 23 line 504-506. . . .due to model discrepancies in the free tropospheric abundance of CO. . . how are these discrepancies evaluated? Discrepancies relative to? It might just be model errors that are unaccounted for.

The discussion has been changed.

(54) p. 23 line 523-529. How about using full-chemistry in the inversion rather than pre-scribed OH?

A very good suggestion! We believe full-chemistry inversion, by assimilating multiple species, is the best approach. We are already working on this with GEOS-Chem model.

(55) Figures: Please look at the figures once again and see if they can be deleted (not necessary) or combined.

Thanks for your suggestion! Four figures have been removed and two have been moved to the appendices.

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