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.

Reviewer #1

I think the paper can be published but should be shortened and checked for spelling mistakes. I thought the contrasting results on emission estimates of two different OH fields were quite interesting. It is not an overly long paper but sometimes it is not very concise.

(1) *Line 10: "the CO emissions in the GEOS-Chem model and accounts for the vertical smoothing of the MOPITT retrieval".*

You mean equation (1) is applied to model profiles and smoothed by the averaging kernels? If so, please make a reference to equation (1).

You should also include a single line sentence or short reference why the adjoint does not give you error bars in the posterior emissions estimates.

Thanks for your suggestion! The expression for the observation operator is analogous to equation (1). We have added the expression for the observation operator. We are also modified the discussion on the a posteriori errors.

(2) *Figure 2: Maybe you should put forward a sentence or two about how the OSSE should ideally look. As far as I understand it, in a perfect world the grid boxes would have a color of yellow which corresponds to the number 1. But it seems to me that there are still a considerable number of grid boxes which are either much lower than 1 or much higher, respectively. It further looks like the OSSE does a better job at the equatorial regions. I think this needs particular consideration as your main results in the following sections adjust emissions to higher values in the extratropics. Did you include the distribution of the MOPITT observations in your pseudo observations and therefore the OSSE reflects the less stringent constraint at higher latitudes? If so please clarify in the text.*

In the adjoint of GEOS-Chem, the cost function is minimized in regions where the gradients are the largest. According to Equation (A1-A3), grids with strong CO emissions, such as East Asia, India, equatorial Africa and South America, will have large initial gradients and thus will be optimized preferentially in the OSSE, which leads to more

yellow color in the equatorial regions. The adjoint model will optimize the weak emission regions, after reducing the model bias due to the strong emission regions. We will see the effect (more yellow color) if we have more iterations in the OSSE.

We didn't include the distribution of MOPITT observations in the OSSE. The pseudo observations are the complete CO fields from model.

(3) *Results and discussion: You need to go through the text again and make it shorter by at least 30%. The text does not have a very good focus.*

Thanks for your suggestion! Four figures were removed to make the paper shorter.

(4) *3.1 Figure 3: Just a comment. Please tell the reader how are you smoothing the GEOS-Chem model output on the vertical grid. Are you just using 1 model grid box that corresponds to the actual altitude of GMD observation?*

We didn't consider the actual altitude of GMD observations. Because the model resolution is coarse (4°x5°), there is clearly going to be large representativeness errors associated with the actual altitude of the GMD site vs. the model altitude. In this figure, we compare GEOS-Chem surface level CO concentrations with the GMD data. This figure has been moved to appendices.

(5) *3.2. Figure 4: Is this Figure 4 really needed here? Also, one could probably guess but there is no explanation if the "difference between two types of scaling factors" is the left panel minus middle panel or vice versa.*

This figure has been removed.

(6) *3.3. Figure 5 and Figure 6: I do not know if this would be easy to do. If so, you should consider merging Figure 5 with Figure 6. Figure 5 is essentially the summary of Figure 6 on an annual scale. Again Figure 6 does not say if the "difference" is based on surface inversion minus profile inversion or vice versa.*

Thanks for your suggestion! These two figures have been merged.

(7) *3.4 Figure 8: Is this Figure essential? I think everything is being said already in Table 1. However, you could expand Table 1 so that it includes all the seasonal values instead of Figure 8. On another note: I would also multiply the values in the color table of Figure 8 by 100. You also need to state the unit in the Figure legend. Table 1 shows the "Relative difference between surface and profile inversion" with the % unit?*

Thanks for your suggestion! This figure has been removed.

Table 1 already has quite a bit of information. We feel that expanding it to include all the seasonal values as well would make it difficult to read.

(8) *4. Section 4.3 Ideal Tracer Experiments: The prior two sections 4.1 and 4.2, respectively are much too long but this section is rather short. You need to explain a little bit more what you exactly did here. You are saying you emit 3.33 Tg CO/day in each of the defined tagged regions, and 1/30 of that is being destroyed after each simulation day?*

As the reviewer indicated, we emitted 3.33Tg CO/day and imposed a loss rate that is given as [CO]/tau, where [CO] is the concentrations in molecules/cm^3 and tau is the timescale for loss, which is 30 days. The description has been modified.

(9) *4.1 I think (if easy to do) Figure 10a and Figure 10b could be combined into 2 useful single Figures for P>700 hPa and 700>P>250 hPa, respectively. As far as I understand every subfigure in Figure 10 tells the story for each individual tagged tracer. But it is also obvious from the Figures that the partial columns of the tagged tracers are confined in the individual tagged regions (although there may be some overlap through to transport).*

Thanks for your suggestion. We decided to remove Figure 10b. It was not critical to the discussion in that section.

(10) *4.2. Figure 10a and 10b It is not really clear from that Figure or from the text in section 4.3 what the actual units of the color bars are. 4.3. Table 2*

Again how do units in Table 2 relate to Figure 10a and 10b, respectively? And what is "Upper fraction (%)"?

The unit in the figure is 10^{18} molec/cm^2. In Table 2, we calculated the monthly mean mass of continental CO tracers. The "upper fraction" is calculated by Mass_upper / Mass_total. The captions have been changed.

Reviewer #2

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

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 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°x0.667°, the native resolution of GEOS-5, to mitigate some of the issues that might arise as a result of the course 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?) . . .*

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?*

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 here is infact 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 yi italic (consistent with eq 2). Fi(x)? yi is a vector of observed concentrations at a given time (does this mean also at a given space —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 Se and Sa 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=sigma xa. Is S sigma?*

Thanks! The scaling factor is expressed as sigma 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 Sa? What*

is Sa? 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?

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.*

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 leads 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. 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 5x10ˆ17 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 x 10^{17} cm^{-2}, located over Antarctica. The global mean column was 1.5 x 10^{18} cm^{-2}. The 5 x 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 Hooghiesmstra et al 2012 since the latter used V4 column CO.*

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

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 using only CO. Consequently, is 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 XCO_2 column data, we have found that the discrepancy on different boundary mixing schemes has only negligible influence on the CO_2 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.

(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.

(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 . . .*

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(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 mesocale 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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Sensitivity of top-down CO source estimates to the modeled vertical structure in atmospheric CO

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Abstract

We assessed the sensitivity of regional CO source estimates to the modeled vertical CO distribution by assimilating multi-spectral MOPITT V5J CO retrievals with the GEOS-Chem model. We compared the source estimates obtained by assimilating the CO profiles and the surface layer retrievals from June 2004 to May 2005. Because the surface layer retrievals are less sensitive to CO in the free troposphere, it is expected that they should provide constraints in the CO source estimates that are less sensitive to the vertical structure of CO in the free troposphere. The inferred source estimates all suggest a reduction in CO emissions in the tropics and subtropics and an increase in the extratropics over the a priori estimates. The tropical decreases were particularly pronounced for regions where the biogenic source of CO was dominant, suggesting an overestimate of the a priori isoprene source of CO in the model. We found that the differences between the regional source estimates inferred from the profile and surface layer retrievals for 2004-2005 were small, generally less than 10% for the main continental regions, except for estimates for South Asia, North America, and Europe. Because of discrepancies in convective transport in the model, the CO source estimates for India and Southeast Asia inferred from the CO profiles were significantly higher than those estimated from the surface layer retrievals during June-August 2004. On the other hand, the profile inversion underestimated the CO emissions from North America and Europe compared to the assimilation of the surface layer retrievals. We showed that vertical transport of air from the North American and European boundary layer is slower than from other continental regions and thus air in the free troposphere from North America and Europe in the model is more chemically aged, which could explain the discrepancy between the source estimates inferred from the profile and surface layer retrievals. We also examined the impact of the OH distribution on the source estimates and found that the

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discrepancies between the source estimates obtained with two OH fields were larger when using the profile data, which is consistent with greater sensitivity to the more chemically aged air in the free troposphere. Our findings indicate that regional CO source estimates are sensitive to the vertical CO structure. They suggest that diagnostics to assess the age of air from the continental source regions should help interpret the results from CO source inversions. Our results also suggest that 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.

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1. Introduction

The emissions of greenhouse gases and other atmospheric pollutants have been significantly increased since the industrial revolution. Their influences on atmospheric chemical composition, local air quality and climate are the subject of increasing numbers of studies. In this context, inverse modeling has been widely used to provide better understanding of the emissions of these atmospheric constituents. In particular, in the past decade there has been expanded use of inverse modeling to better quantify the emissions of atmospheric CO (e.g., Pétron et al., 2004; Heald et al., 2004; Arellano et al., 2006; Jones et al., 2009; Kopacz et al., 2010; Fortems-Cheiney et al., 2011; Gonzi et al., 2011). Tropospheric CO is produced from incomplete combustion and is a byproduct of oxidation of hydrocarbons. As the primary sink of OH, tropospheric CO has significant influence on the oxidative capacity of the atmosphere. The lifetime of tropospheric CO is a few months, which is long enough to track within intercontinental scale pollution plumes but short enough to provide strong signals over background distribution (Jiang et al., 2010). Previous studies (Palmer et al., 2006; Wang et al., 2009) have demonstrated that CO can be included in the inverse analyses of CO_2 sources and sinks to reduce the influence of model

transport errors.

Remote sensing from space-based instruments provide valuable global observational coverage to enable us to better constrain CO emissions. There are now several satellite sensors from which abundances of CO in the troposphere have been retrieved using measurement of thermal infrared (TIR) radiation near 4.7 μm: MOPITT (Measurements of Pollution In The Troposphere), on EOS-Terra, launched December 1999 (Deeter et al., 2003); AIRS (Atmospheric InfraRed Sounder), on EOS-Aqua, launched May, 2002 (Warner et al., 2007); TES, (Tropospheric Emission Spectrometer) on EOS-Aura, launched July, 2004 (Luo et al., 2007); and IASI (Infrared Atmospheric Sounding Interferometer), on METOP-A, launched October, 2006 (George et al., 2009). The TIR radiances are sensitive to CO concentrations from the middle to the upper troposphere. The lack of global observations of CO near the surface has implications for the use of inverse modeling to quantify CO emissions because the modeled CO distribution in the free troposphere is affected by discrepancies in the parameterization of convective transport in models (e.g. Ott et al., 2009), the simulated chemical sink of CO (e.g. Jiang et al. 2011), and long-range transport (e.g. Arellano et al. 2006b; Jiang et al., 2013).

The multispectral MOPITT version 5 CO product (V5J, where J indicates joint retrievals) are the first retrievals to exploit simultaneous near infrared (NIR) and TIR measurement to provide greater sensitivity to CO in the lower troposphere over land (Deeter et al., 2011). Recently, Jiang et al. (2013) showed that lower tropospheric MOPITT V5J CO retrievals can be used to study the influence of convective transport error on CO source estimates. They compared the CO source estimates in June-August 2006, inferred from MOPITT surface layer retrievals, the profile retrievals, and the column amounts. They found that there were large discrepancies in the inferred source estimates obtained with the surface layer and profile retrievals in Asian

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142 monsoon regions where strong emissions are co-located with significant vertical mass flux due to
143 convection. The discrepancies in the CO source estimates were also used to assess the impact of
144 vertical transport error on the CH_4 emission estimates from Indonesian peat fires in fall 2006,
145 estimated from TES CH_4 observations (Worden et al., 2013).

146 The study by Jiang et al. (2013) was carried only for summer 2006 and focused mainly
147 on discrepancies in convective transport. The work presented here complements and extends that
148 analysis. Reflecting its long lifetime, CO is destroyed mainly in the free troposphere rather than
149 in the boundary layer. Thus, free tropospheric CO will be more susceptible to discrepancies in
150 OH, and in long-range transport. One way to mitigate the potential impact of discrepancies in
151 transport and OH on CO inversion analyses is to use surface observations, near the CO source
152 regions. However, the current surface-observing network is sparse, whereas MOPITT provides
153 significantly greater observational coverage. Therefore, we focus here on the use of the surface
154 layer retrievals from MOPITT for inverse modeling CO sources. We expect that the source
155 estimates inferred from the surface layer retrievals will be less sensitive to errors in OH and
156 model transport. We estimate and compare monthly CO source estimates for June 2004 to May
157 2005 using MOPITT tropospheric profiles and surface layer retrievals to observe the influence of
158 the OH distribution and the vertical structure in CO, as observed by MOPITT, on the inferred
159 source estimates. The updated global CO distributions will be used as boundary conditions in our
160 companion paper to constrain the North America CO emission at a horizontal resolution of
161 0.5°x0.67° (Jiang et al., 2014). The objective of that study is to assess the extent to which we can
162 further reduce the impact of model transport and chemistry errors on CO source estimates in a
163 regional inverse modeling context.

164 This paper is organized as follows: in Section 2 we describe the MOPITT instruments

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Deleted: (2013) focused only on summer 2006. The work presented here complements and extends that analysis.

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Deleted: with 0.5°x0.667° resolution (Jiang et al., 2014). We also examine the impact of discrepancies in OH on the source estimates. The concentration of tropospheric OH peaks in the middle troposphere (e.g., Spivakovsky et al., 2000), therefore, errors in CO due to discrepancies in OH will accumulate in the free troposphere through the influence of long-range transport. Consequently, we expect that the source estimates inferred from the surface layer retrievals will be less sensitive to errors in OH

and the GEOS-Chem model used in this work. In Section 3 we outline the inverse method. We then discuss the annual and seasonal variations of the estimated CO emissions in Section 4. The discrepancies in the CO source estimates are interpreted in the context of the CO vertical structure and the OH distribution. Our conclusions follow in Section 5. In Appendix A we present the results of an indirect validation of the MOPITT data that was conducted to guide the filtering of the data used in the assimilation, and in Appendix B we have included a discussion of the optimization scheme used in the assimilation.

2. Observations and Model

2.1. MOPITT

The MOPITT instrument was launched on the Terra spacecraft on December 18, 1999. The satellite is in a sun-synchronous polar orbit of 705 km and crosses the equator at 10:30 local time. With a footprint of 22 km *x* 22 km, the instrument makes measurements in a 612 km cross-track scan that provides global coverage every three days. The MOPITT data used here were obtained from the joint retrieval of CO from TIR (4.7μm) and NIR (2.3μm) radiances using an optimal estimation approach (Worden et al., 2010; Deeter et al., 2011). The retrieved volume mixing ratios (VMR) are reported as layer averages 10 pressure levels (surface, 900, 800, 700, 600, 500, 400, 300, 200 and 100 hPa) and the relationship between the retrieved CO profile and the true atmospheric state can be expressed as:

$$\hat{\mathbf{z}} = \mathbf{z}_a + \mathbf{A}(\mathbf{z} - \mathbf{z}_a) + \mathbf{G}\varepsilon \quad (1)$$

where $\mathbf{z}_a$ is the MOPITT a priori CO profile (expressed as log(VMR)), $\mathbf{z}$ is the true atmospheric state averaged at MOPITT grid levels (also as log(VMR)), $\mathbf{G}\varepsilon$ describes the retrieval error, and $\mathbf{A} = \partial\hat{\mathbf{z}}/\partial\mathbf{z}$ is the MOPITT averaging kernel matrix, which gives the sensitivity of the retrieval to

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the actual CO in the atmosphere. The MOPITT V5 data have been evaluated by Deeter et al. (2012, 2013) using aircraft measurements from the National Oceanic and Atmospheric Administration (NOAA). For the V5J multi-spectral retrievals, they found a small positive bias of 2.7% at the surface and a much larger positive bias of 14% at 200 hPa. As a result of the high bias in the upper troposphere, in our analysis we do not use the retrievals at altitudes above 200 hPa. We conduced an indirect validation of the MOPITT V5J data (see Appendix A) using NOAA Global Monitoring Division (GMD) in situ observations, which suggested that there is a high-latitude positive bias in the MOPITT data, possibly associated with the lower degrees-of-freedom-for-signal (DFS) at higher latitudes. Consequently, in this work, we omitted MOPITT data that are polarward of 40° over oceans and 52° over land.

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2.2. GEOS-Chem

The GEOS-Chem global chemical transport model (CTM) [www.geos-chem.org] is driven by assimilated meteorological fields from the NASA Goddard Earth Observing System (GEOS-5) at the Global Modeling and data Assimilation Office. We use version v34 of the GEOS-Chem adjoint, which is based on v8-02-01 of the forward GEOS-Chem model, with relevant updates through v9-01-01. Our analysis is conducted at a horizontal resolution of 4°x5° and employs the CO-only simulation in GEOS-Chem, which uses archived monthly OH fields from the full chemistry simulation. The standard OH fields used in this work are from GEOS-Chem version v5-07-08, with a global annual mean OH concentration of 0.99×10^6 molec/cm^3 (Evans et al. 2005). We use this as our standard OH field to facilitate comparison of our results with those of Kopacz et al. (2010). We also conduct a sensitivity analysis using OH fields from the full chemistry simulation of v34 of the adjoint model, run in forward mode. This simulation produces a global annual mean OH concentration of 1.24×10^6 molec/cm^3.

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The anthropogenic emission inventories are identical to those used in Jiang et al. (2013). Anthropogenic emissions are from EDGAR 3.2FT2000 (Olivier et al., 2001), but are replaced by the following regional emission inventories: the US Environmental Protection Agency National Emission Inventory (NEI) for 2005 in North America, the Criteria Air Contaminants (CAC) inventory for Canada, the Big Bend Regional Aerosol and Visibility Observational (BRAVO) Study Emissions Inventory for Mexico (Kuhns et al., 2003), the Cooperative Program for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) inventory for Europe in 2000 (Vestreng et al., 2002) and the INTEX-B Asia emissions inventory for 2006 (Zhang et al., 2009). Biomass burning emissions are based on the Global Fire Emission Database (GFED3), with a three-hour temporal resolution (van der Werf et al., 2010). Additional CO sources come from oxidation of methane and biogenic volatile organic compounds (VOCs,) as described in previous studies (Kopacz et al., 2010; Jiang et al., 2013). The biogenic emissions are simulated using the Model of Emissions of Gases and Aerosols from Nature, version 2.0 (MEGANv2.0) (Guenther et al., 2006). The distribution of the annual mean CO emissions for June 2004 to May 2005 is shown in Figure 1. The annual global sources are 928 Tg CO from fossil fuel, biofuel and biomass burning, 661 Tg CO from the oxidation of biogenic NMVOCs, and 884 Tg CO from the oxidation of CH_4.

3. Inversion Approach

We use the 4-dimensional variational (4D-var) data assimilation system in GEOS-Chem (e.g., Henze et al., 2007; Kopacz et al., 2009, 2010; Singh et al. 2011; Jiang et al., 2011, 2013; Parrington et al., 2012) to estimate the CO sources. Details of the 4D-var scheme are given in Henze et al. (2007) and Kopacz et al. (2009, 2010). In this approach, we minimize the cost function of the form,

$$J(\mathbf{x}) = \sum_{i=1}^{N} (\mathbf{F}_i(\mathbf{x}) - \mathbf{z}_i)^{\mathrm{T}} \mathbf{S}_{\Sigma}^{-1} (\mathbf{F}_i(\mathbf{x}) - \mathbf{z}_i) + (\mathbf{x} - \mathbf{x}_a)^{\mathrm{T}} \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) \quad (2)$$

where $\mathbf{x}$ is the state vector of CO emissions, N is the number of MOPITT observations that are distributed in time over the assimilation period, $\mathbf{z}_i$ is a given MOPITT profile (or surface level retrieval), and $\mathbf{F(x)}$ is the forward model which represents the transport and chemistry of CO in the GEOS-Chem model and accounts for the vertical smoothing of the MOPITT retrieval,

$$\mathbf{F}_i(\mathbf{x}) = \mathbf{z}_a + \mathbf{A}(\mathbf{H}_i(\mathbf{x}) - \mathbf{z}_a) \quad (3)$$

Here $\mathbf{z}_a$ and $\mathbf{A}$ are the MOPITT a priori profile and averaging kernel, respectively, introduced in Equation (1), and $\mathbf{H}_i(\mathbf{x})$ is the GEOS-Chem profile of CO at the MOPITT observation location and time. The definition of the cost function assumes that the distribution of the errors for both the state vector $\mathbf{x}$ and the a priori constraint on the CO emissions $\mathbf{x}_a$ are Gaussian, and these errors are given by $\mathbf{S}_{\Sigma}$, the observational error covariance matrix, and $\mathbf{S}_a$, the a priori error covariance matrix, respectively. Minimization of the cost function provides the a posteriori CO emissions $\hat{\mathbf{x}}$, corresponding to the maximum of the conditional probability density function ($P(\mathbf{x}|\mathbf{y})$), with the a posteriori error covariance matrix $\hat{\mathbf{S}}$. However, because the 4D-var optimization scheme does not store the full Hessian matrix, it is difficult to construct the a posteriori error covariance matrix, which is the inverse of the Hessian. Details of the optimization approach are given in Appendix B.

We employ a similar procedure for data processing and quality control as in our previous study, Jiang et al. (2013). Since MOPITT V5J CO retrievals have a positive bias at high altitudes (Deeter et al. 2013), our analysis is restricted to CO retrievals below 200 hPa. Following Jiang et

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al. (2013), we also reject MOPITT data with CO column amounts less than $5x10^{17}$ molec/cm^2 and use only daytime data. The threshold of $5x10^{17}$ molec/cm^2 was selected to prevent unrealistically low CO columns from adversely impacting the inversion analyses.

The observation error $\mathbf{S}_\Sigma$ represents a sum of the retrieval errors, representativeness errors, and random model errors. Using the Relative Residual Error (RRE) approach (Palmer et al., 2003; Heald et al., 2004), which assumes that the mean differences between the model and observations are due to discrepancies in the emissions while the residual reflects the observation error, Kopacz et al. (2010) estimated that the observation errors for the MOPITT columns are 10% - 30%. Although the RRE approach does not account for systematic model errors, it provides a possible estimate of the random component of the observation errors. Accurately characterizing the systematic errors (in the model and observations) is a challenge. Keller et al. (Quantifying Model Biases in CO Emission Estimation Using Weak Constraint 4D-var, manuscript in preparation) have assimilated MOPITT V5J data using a weak-constraint 4D-var scheme to characterize the systematic component of the observation error. Their results suggest that the weak-constraint 4D-var is a promising approach for accounting for systematic errors, but it is still challenging. In the absence of meaningful information about the systematic errors in the model for the period considered here, we do not account for systematic errors in minimizing the cost function. Following Jiang et al. (2011, 2013), we assume a uniform observation error of 20%. Our assumed 20% error likely overestimates the observation error in the upper troposphere and underestimates it near the surface.

As described in Jiang et al. (2013), we combine the combustion CO sources (fossil fuel, biofuel and biomass burning) with the CO from the oxidation of biogenic NMVOCs and solve for the total CO emissions in each grid box, assuming a 50% uniform a priori error and that the

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errors are uncorrelated. We optimize the source of CO from the oxidation of methane separately as an aggregated global source, assuming an a priori uncertainty of 25%. As in Jiang et al. (2013), we produce initial conditions at the beginning of each monthly assimilation window by assimilating MOPITT V5J data using a sequential sub-optimal Kalman filter (Parrington et al. 2008). For the results presented here, the Kalman filter assimilation was carried out from January 1, 2004, to May 1, 2005, and to optimize the CO distribution, which was archived at the beginning of each month. In the monthly inversion using the 4D-var system, the optimized CO distribution from the Kalman filter was read at the beginning of each month to obtain initial conditions. Consequently, the initial conditions for the model simulation are independent of the inverse analyses. Although we use a one-month assimilation window, it is possible that a longer window of two or three months would lead to greater constraints on the CO source estimates. However, as we will show below, the inversion is sensitive to the specified OH distribution and thus with a longer assimilation window would be more susceptible to discrepancies in the CO chemical sink.

4. Results and Discussion

4.1. CO Source Estimates for June 2004 – May 2005

Figures 2a and 2f show the annual mean emission scaling factors for June 2004 to May 2005, obtained using the MOPITT surface layer and profile retrievals, respectively. Both analyses suggest that CO emissions in the tropics should be reduced, whereas the emissions in middle and high latitudes should be increased. However, as shown in Figure 2k, the a posteriori scaling factors from profile inversion is higher in India and Southeast Asia. As discussed in Jiang et al. (2013), these descrepancies over India and Southeast Asia are likely due to model errors in convection transport. The profile inversion also produces larger emissions in parts of tropical

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Africa and northern South America. In general, however, the a posteriori emissions from the profile inversion are lower than those obtained from the surface layer inversion, particularly at middle and high latitudes.

Table 1 shows the annual mean regional CO emissions from June 2004 to May 2005, inferred from the surface layer and profile retrievals. In this work, only the total CO emission is optimized in each grid box, but because the different CO source types have different spatial and temporal distributions, we apply the scaling factors in each grid box to each source type, which can provide useful information on the individual source types. As shown in Table 1, the emission reductions in the tropics and subtropics reflect large reductions in the biogenic source of CO, suggesting that our a priori biogenic emissions are too high. For example, in South America, with the profile inversion the biogenic source was reduced by 32%, whereas the combustion source was reduced by 13%. In northern Africa the biogenic source was reduced by 26% and the combustion source was reduced by 20% with the profile inversion. In the 48 contiguous United States the biogenic source was reduced by 31%, whereas the combustion source was increased by 5%. The reductions in the biogenic emissions were smaller in the surface layer inversion, but were still large for South America and northern Africa, 27% and 28%, respectively. We note that although there are large differences between the regional source estimates inferred from the profile and surface layer retrievals, the global total a posteriori CO emissions estimated from the two sets of retrievals are similar, 1513 Tg CO and 1555 Tg CO, respectively.

The seasonal mean scaling factors are shown in Figure 2. The main seasonal feature in the figure is that the inversions tend to decrease CO emission in the summer hemisphere and increase them in the winter hemisphere, with the profile inversion producing larger reductions (2b and 2g) and smaller increases (Figures 2d and 2i). Consequently, the differences between the

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scaling factors from the surface and profile inversions are smaller in winter. This pattern is consistent with an overestimate of isoprene emissions and a possible underestimate of wintertime fossil fuel combustion (Stein et al. 2014). The overestimate of biogenic emissions in GEOS-Chem by MEGANv2.0 has been reproted by previous studies (Barkley et al., 2008; Millet et al., 2008; Liu et al., 2010). Millet et al., (2008) found that North American isoprene emissions estimated by MEGAN were greater than those inferred from observations of formaldehyde (HCHO) from the Ozone Monitoring Instrument (OMI) by as much as 23%. Liu et al. (2010) used a newer version of MEGAN, version 2.1, which simulates lower isoprene emissions than version 2.0 (which is emloyed in our analysis), and found that it also produced an overestimate of CO from isoprene oxidation, particulalry in eastern South America. Marais et al. (2014) found that MEGANv2.1 ovestimated African isoprene emissions for 2005 – 2009 by 26% relative to those inferred from OMI data, primarily over the equatorial forests and the northern savannas.

Figure 3 shows the timeseries of the monthly mean source estimates for the 48 contiguous United States, Europe, East Asia, and India/Southeast Asia. For India/Southeast Asia, the dominant source of CO is biomass burning from Indonesia, which peaks in August - October, and from southeast Asia, which peaks in February - April. For the other regions, combustion of fossil fuels and biofuels provides the main annual source of CO. As we noted above, the tendency is for the inverse model to reduce the emissions in summer and increase them in winter, particularly in the United States and East Asia. In the profile inversion, the North American combustion emissions were reduced by about a factor of two in July and August 2004, whereas they were increased by 48% in January – March 2005. The summertime reduction of the North American combustion emissions was smaller than that obtained with the surface layer retrievals, whereas the wintertime increase was similar in both inversions. In Asia, both inversions

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produced comparable summertime reductions and wintertime increases in the combustion emissions, with the emission estimates from the profile inversion being slightly lower in summer and higher in winter. The seasonality of the European source estimates obatined from the surface layer retrievals was much less pronounced than that obtained for North America and Asia, and was consistently higher than those obatined from the profile inversion.

The seasonal variation of the a posteriori combustion emissions shown in Figure 3 is consistent with the results of Kopacz et al. (2010). Using data from MOPITT, SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY), and AIRS, Kopacz et al. (2010) showed that the CO emissions from North America, Europe and East Asia should be significantly increased in winter. There is also good agreement between the two studies in the aggregated emissions in the extratropical northern hemisphere. The total combined a posteriori combustion source from the United States, Alaska, Canada, Europe, and East Asia was 515 Tg and 548 Tg from the profile and surface inversions, respectively. The corresponding a posteriori estimate from Kopacz et al. (2010), was 520 Tg.

However, there are large differences in the region source estimates between our analysis and that of Kopacz et al. (2010). For example, our annual combustion emission estimate for the contiguous United States was 100 Tg from the profile inversion, whereas Kopacz et al. (2010) inferred 50 Tg. We note that our total a posteriori combustion source estimates for North America of 173 Tg CO and 156 Tg CO for the surface layer and profile inversions, respectively, is comparable to the a posteriori estimate of 206 Tg CO obtained by Fortems-Cheiney et al. (2012) from their inversion analysis of the MOPITT data for 2005. A significant difference between our inverson and that of Kopacz et al. (2010) is that their a priori combustion source for the United States was 40 Tg, whereas ours was 112 Tg. Their low a priori estimate was based on

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the results of Hudman et al. (2008), who suggested a 60% reduction in anthropogenic emissions in the United States as a result of an analysis of aircraft data in July – August 2004. The discrepancies in the regional source estimates between the results here and those of Kopacz et al. (2010) could also be related to differences in the configuration of the inversion analyses, such as the treatment of the initial conditions or vertical transport in the models. Our inversion analyses employed the GEOS-5 meteorological fields, whereas Kopacz et al. (2010) used GEOS-4. A significant factor could be the treatment of the biogenic source of CO. Here the biogenic sources are combined with the combustion sources and optimized at the resolution of the model. In contrast, Kopacz et al. (2010) aggregated the biogenic source with the methane source and optimized the global mean source from methane and VOC oxidation. As shown in Jiang et al. (2011), optimizing the VOC source at a lower resolution than the combustion emissions could result in an overadjustment of the combustion sources.

In general, we find that the regional source estimates inferred from the surface layer and profile retrievals are consistent, with relative differences of less than 10%, except for source estimates for North America (the United States, Alaska and Canada), Europe, and India/southeast Asia (see Table 1). The discrepancy between the source estimates for India/southeast Asia from the two inversions is linked to vertical transport by the Asian monsoon and was discussed by Jiang et al. (2013). In the next section, we present a passive tracer analysis to provide insight into the discrepancies between the source estimates from North America and Euope.

4.2. Ideal Tracer Experiments

It is surprising that Europe and North America (the United States and Canada) are the two regions, after India/southeast Asia, with the largest discrepancies between the source

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estimates inferred from the profile and surface layer inversions. To better understand how the vertical transport of CO from these region could impact the inversions, we conducted an analysis using an idealized CO-like tracer. We performed a tagged-CO simulation for the period June 2004 – May 2005 in which we imposed a constant source of CO of 3.33 Tg CO/day from each of the continental source regions shown in Figure 4, with a constant and uniform timescale for loss of 30 days (i.e., the lost rate was given as [CO]/30 molec cm^{-3} day^{-1}, where [CO] is the CO concentration). We ran separate tracers for each of the continental regions, with each tracer emitted only in that region but chemically destroyed everywhere.

The tracers were initialized to a uniformly low abundance of 1 pptv and the model was run for 17 months prior to June 2004 to spinup the tracer distributions. Shown in Figure 5 are the boundary layer (defined here as the surface – 700 hPa) and free tropospheric (700 – 250 hPa) partial columns of the continental tracers for June 2004. In the extratropical northern hemisphere, a larger fraction of the Asian surface emissions are exported to the free troposphere, compared to the North American and European emissions. We find that transport of the Asian emissions to the free troposphere is faster even in winter. In the tropics, transport of surface emissions to the free troposphere is slowest for South America (not shown), most likley due to the fact that in boreal summer the ITCZ is located in northern South America (in the northern hemisphere) and hence transport of south American emissions to the southern subtropics and extratropics is facilated instead by the influence of mid-latitude cyclones (Staudt et al., 2002). In fall, the ITCZ moves south and convection over South America intensifies (Liu et al., 2010); as a result, we find that, in December, the fraction of South American emissions in the free troposphere is greater, and is comparable to that from northern Africa (not shown).

The monthly mean fraction of the global mass of each continental tracer that is in the

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boundary layer and the free troposphere is listed in Table 2. North America and Europe have the smallest mass fraction in the free troposphere, 26% and 21%, respectively. This suggests that, relative to the other continental regions, the air in the free troposphere from Europe and North America is older and more chemically aged. This is consistent with the results of Stohl et al. (2002), who examined the transport of idealized tracers from continental source regions using a Lagrangian particle dispersion model. They found that the European tracer was more confined to the lower troposphere, relative to the North American and Asian tracers. They also noted that "in terms of vertical transport, the North America tracer… behaves intermediately between the Asia and Europe tracers." This suggests that the surface layer and profile inversions are sampling sufficiently different air masses that they obtain different constraints on the North American and European source estimates. The surface layer inversion is sampling air that is less aged and should, therefore, be less susceptible to discrepancies in the OH abundance.

4.3. Influence of the OH distribution

In this section we compare the impact on the source estimates of the OH distribution from v8-02-01 of GEOS-Chem with that from our standard inversion (which is based on v5-07-08 of GEOS-Chem). As shown in Figure 6, v8-02-01 OH is significantly higher than on v5-07-08 in the Northern hemisphere, while it is much lower over South America and Indonesia.

Using the v8-02-01 OH fields, we repeated the profile and surface inversions for June – August 2004. Shown in Figure 7 are the scaling factors and their differences, based on the two versions of the OH fields. With v8-02-01 OH, the a posteriori emissions in the tropics changed only slightly, while the inferred emission estimates in the extratropics, mainly for North America and Europe, were much greater that those obtained with v5-07-08 OH. The regional source estimates are given in Table 3. For the contiguous United States, with v5-07-08 OH we inferred a

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June-August source of 25.4 Tg CO using the profile retrievals, whereas with v8-02-01 OH we estimated a source of 49.2 Tg CO. Similarly, for Europe the source estimates inferred from the profile inversion with v5-07-08 OH was 47.3 Tg, whereas with v8-02-01 OH it with was 68.3 Tg.

To help understand the differences in the regional source estimates shown in Table 3, the mean CO lifetime in the tropics and in the northern midlatitudes, for August 2004, are plotted in Figure 8. Throughout the lower and middle troposphere in the northern midlatitudes, the CO lifetime is about 30% shorter with v8-02-01 OH, decreasing to less than 30 days between 900 – 400 hPa.. The shorter lifetime resulted in a reduction of the CO burden in the midlatitude free troposphere. Consequently, greater extratropical a posteriori source estimates, relative to the v5-07-08 OH inversions (see Table 3), were required to bring the model into agreement with the MOPITT data. In Jiang et al. (2014), this change in the free tropospheric distribution of CO is discussed further in the context of a regional inversion analysis for North American source estimates. In the tropics, the CO lifetime increased by about 15% with v8-02-01 OH. However, as shown in Figure 6, this reflects reductions in OH over source regions such as South America and Indonesia, which are partially offset by increases in OH over northern tropical Africa and the remote tropics. In general, we find that the relative differences between the source estimates from the v8-02-01 and v5-07-08 OH inversions are smaller for the surface inversion compared to the profile inversion, reflecting the fact the surface layer inversion is more strongly influenced by fresh emissions and less by background CO in the free troposphere.

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5. Summary

We presented a global inversion analysis to quantify monthly mean CO source estimates during the period of June 2004 – May 2005 using the version 5 MOPITT retrievals. Building on the work of Jiang et al. (2013), we conducted a comparative analysis of the influence of the

MOPITT profile and surface layer retrievals on the inferred CO source estimates. The inversions suggest a reduction in CO emission in the tropics, possible due to an overestimate of the biogenic source of CO, and an increase in emissions at middle and high latitudes. In the northern extratropics, we found that the inferred source estimates are typically much greater in winter than in summer, consistent with the seasonality in CO emissions inferred by Kopacz et al. (2010). With our standard OH distribution, we inferred source estimates of 148 Tg, 180 Tg, and 284 Tg for the contiguous United States, Europe, and East Asia, respectively, using the surface layer retrievals. Using the profile retrievals, the inferred source estimates were lower, 131 Tg, 158, and 282 Tg, respectively.

In general, we find that the annual mean, regional source estimates inferred from the surface layer retrievals and those from the profile retrievals are in agreement to better than 10%, with the exception of the North American (United States and Canada), European, and Indian/southeast Asian estimates. The difference in the Indian/southeast Asian estimates is due to discrepancies in vertical transport associated with the strong convective transport over the Southeast Asian region (Jiang et al., 2013). For Europe and North America, we argue that the differences in the source estimates from the profile and surface inversion are due to model discrepancies in the free tropospheric abundance of CO from these regions. We conducted an ideal tracer experiment and showed that transport of surface emissions from Europe and North America to the free troposphere is slower than from other continental regions. Consequently, compared to the inversion using the surface layer retrievals, the profile inversion is sampling older, more chemically age air from North America and Europe in our simulation, and is, therefore, more susceptible to discrepancies in long-range transport and in the chemical sink of CO. This suggests that diagnostics to assess the age of air from the continental source regions

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should be useful for interpreting the results from CO source inversions.

We examined the impact of the OH distribution on the inferred CO source estimates, using OH fields from versions v5-07-08 and v8-02-01 of GEOS-Chem. We found that changing OH from v5-07-08 (used in our standard inversions) to v8-02-01 produced large differences in the extratropical source estimates. The relative differences in the source estimates from the profile inversion using v5-07-08 and v8-02-01 OH were 64%, 33%, and 36% for source estimates from the contiguous United States, East Asia, and Europe for June – August 2004, when the CO lifetime is short. In the inversions using the surface layer data we found that the impact of the OH fields was reduced, but was still large: 40%, 20%, and 24%, respectively. The smaller impact of the OH fields in the surface layer inversion is due to the fact that the OH sink is at a maximum in the middle troposphere, while the surface layer retrievals have maximum sensitivity near the boundary layer.

The results presented here clearly demonstrate the challenge of inverse modeling of CO emissions. Although the CO chemistry is relatively simple, the sensitivity to tropospheric OH is a major issue. Accurate OH fields are essential for constraining CO reliably. In recent studies, Fortems-Cheiney et al. (2011) introduced Methyl Chloroform (MCF) in their CO inversion to provide a constaint on the OH abundnace. However, MCF is observed at only a few surface sites, hence, although a MCF inversion might give a good global mean OH constraint, it will not help mitigate discrepancies in the regional distribution of OH. A better method to improve the OH would be to assimilate tropospheric ozone and it precursors, together with CO, as was done by Miyazaki et al. (2012). They showed that in such a multispecies assimilation, the adjustment in the monthly mean, zonal OH abundance could be as large as 20%.

Our inversion results also highlight the need to better quantify the isoprene source of CO.

Previous studies (e.g., Abbot et al., 2003; Shim et al., 2005; Millet et al., 2008) have used space-based observatios of HCHO to inferred isoprene emissions. Since isoprene impacts the tropopsheric abundance of OH and ozone, it may be that the most reliable constraint on the isoprene source will be obtained by jointly assimilating HCHO data together with observations of CO and other ozone precursors. In that context, Fortems-Cheiney et al. (2012) conducted a joint inversion analysis using CO, HCHO, methane (CH_4), and MCF, and found that the biogenic a priori source of CO was overestimated, whereas the a priori combustion source was underestimated. Our results and those of Fortems-Cheiney et al. (2012) suggest that the way forward will require exploiting a broader range of composition measurements, besides just that of atmospheric CO, to better quantify the regional CO budget.

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Appendix A: Indirect Validation of the MOPITT V5J Data

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Although Deeter et al. (2012, 2013) showed that the bias in the V5 MOPITT data relative to aircraft observation is small in the lower troposphere, we note that the aircraft data are limited in space and time. Therefore, we conducted an indirect validation of the MOPITT data by assimilating the data to optimize the modeled CO distribution and compared it with independent data. A better understanding of potential bias in the data is critical for properly quantifying the source estimates. Comparison of the CO distribution obtained with the a posteriori source estimates can reveal potential bias in the inversion, but in that approach it is difficult to determine whether the bias is in the data or the model. By constraining the modeled CO to match the observations, we can more easily identify potential biases in the data. For example, recent inversion studies (Arellano et al., 2006; Jones et al., 2009; Hooghiemstra et al. 2012) have shown that the a posteriori CO emissions, inferred from MOPITT data, resulted in an overestimate of CO abundances relative to surface in situ measurements. Hooghiemstra et al. (2012) suggested

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that the overestimate of surface CO was due to a bias in the V4 MOPITT data that they employed. However, Arellano et al. (2006) and Jones et al. (2009) used the V3 MOPITT product in their inversion analyses. Jiang et al. (2013) suggested that the bias seen by Hooghiemstra et al. (2012) could be due to discrepancies in vertical transport. We also note that MOPITT validation comparisons (Deeter et al., 2010; 2013) over land rely on NOAA aircraft in situ CO profiles that are concentrated in North America with only two out of 15 locations at latitudes higher than 50°N.

To assess potential bias in the MOPITT data set, we assimilated the MOPITT V5J CO profile data into the GEOS-Chem model using the sequential sub-optimal Kalman filter and compared the resulting CO field with GMD in situ surface CO observations. Figure A1 shows the comparison of the assimilated CO with monthly mean CO concentrations at selected GMD sites. We first compared the free model simulation (the standard GEOS-Chem simulation without Kalman filter assimilation) with GMD data. The initial condition for the free model run is the model original initial condition on June 1 2004, without optimization. In the northern hemisphere, the CO concentration of the free run model is higher than that of GMD in summer and fall, and significantly lower than that of GMD in winter and spring. In the southern hemisphere, the free run model generally overestimates the observed CO, which is consistent with previous studies (Shindell et al. 2006; Kopacz et al. 2010). In our assimilation, we first assimilated the MOPITT profile data between 60°S to 60°N. The result shows that the assimilated MOPITT data (dark blue dotted line) are highly consistent with the GMD data between 0°N to 30°N. However, the analysis has a positive bias in the mid-latitudes of the southern hemisphere and in the high latitudes of the north hemisphere, such as at Cold Bay (CBA), Alaska, and Mace Head (MHD), Ireland. In the southern hemisphere, at Crozet Island

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(CGO), the a priori is biased high and the assimilation exacerbated the bias. Although Hooghiemstra et al. (2012) used V4 MOPITT data, our results suggests that the V5J data may also be biased high in the southern hemisphere. To reduce the potential impact of this high latitude bias in both hemispheres, we omitted MOPITT data in the assimilation that are polarward of 40° over oceans and 52° over land. As shown in Figure A1, this improved the agreement between the assimilated CO and the GMD data, but it did not completely remove the positive high-latitude bias at MHD and CGO. The results in Figure A1 show the value in the optimized initial conditions prior to the source estimation. The initial condition biases are much smaller than using original initial conditions from the free running model, particularly in winter and spring.

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Appendix B: Optimization of the Cost Function

For the results presented here, the state vector in Equation (2) is not the CO emissions, but is a set scaling factors σ such that $\hat{\mathbf{x}} = \sigma \mathbf{x}_a$. Consequently, the optimization is conducted by minimizing the gradient of the cost function with respect to the scaling factors, with errors in the emission inventories assumed on a relative basis rather than on an absolute basis. In this approach, the gradient of the cost function as described in Equation (2) is usually scaled as follows:

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$$\frac{\partial J}{\partial (x / x_a)} = \frac{\partial J}{\partial x} \cdot x_a \quad \text{(B1)}$$

This method is referred to as the linear scaling factor optimization. It assumes that the uncertainty in the emissions is normally distributed about scaling factor one. Henze et al. (2009) indicated that the normal distribution about one is nonphysical because it allows for negative emissions. An alternative method is the logarithm (LOG) scaling factor optimization (Henze et

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al., 2009):

$$\frac{\partial J}{\partial \ln(x/x_a)} = \frac{\partial J}{\partial x} \cdot x_a \cdot \frac{x}{x_a} \quad \text{(B2)}$$

It represents a log-normal distribution of scaling factors about zero. One advantage of LOG scaling factor optimization is that it can prevent negative scaling factors (Henze et al. 2009). However, it does not reduce negative gradients effectively because the increase in the factor x/x_a will partially offset the decrease of $\partial J/\partial x$. For example, assuming a negative gradient due to the model being lower than measurements (for example, $\partial J/\partial x = -100$), the inversion will increase emission (for example, $x/x_a = 1.5$) to reduce the negative gradient (for example, to $\partial J/\partial x = -66.7$). Using linear scaling factor optimization, we will see 33% improvement (reduction) of the gradient. However, using LOG scaling factor optimization, there is no improvement of the gradient because $\partial J/\partial x \times x/x_a = -66.7 \times 1.5 = -100$.

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Figure B1 shows the results of the linear scaling optimization and the LOG optimization in a simulation experiment for April 2006. In the experiment, we created pseudo-observations by archiving the model output with the CO emissions unchanged (the default CO emission inventory). In the inversion analysis of the pseudo-data, we then reduce the CO emission by 50% so that the objective of the experiment is to produce scaling factors that can return the source estimate to the default emissions (i.e., scaling factors of 1.0). According to Equation (B1-B3), grids with strong CO emissions, such as those in East Asia, India, equatorial Africa and South America, will have a large initial gradient. Because the cost function is minimized in regions where the gradients are the largest, these strong emission regions will be optimized preferentially. After 30 iterations, the a posteriori estimate with linear method (Figure B1a) converges to the true state in all major emission regions. The results with LOG method are clearly worse (Figure

B1b).

To better reduce the negative gradient, and avoid negative scaling factors, we developed the following modification to the LOG method:

$$\frac{\partial J}{\partial \ln(x/x_a)} = \frac{\partial J}{\partial x}\cdot x_a \cdot \frac{x}{x_a} \qquad when: \quad \frac{x}{x_a} \le 1$$
$$\frac{\partial J}{\partial \frac{1}{2}[(x/x_a)^2 - 1]} = \frac{\partial J}{\partial x}\cdot x_a / \frac{x}{x_a} \qquad when: \quad \frac{x}{x_a} > 1 \qquad \text{(B3)}$$

This new method is referred to as "LOGX2". It can minimize the positive and negative gradients with comparable efficiency. As shown in Figure B1c, the optimization effect of the LOGX2 method is slightly better than that of the linear method. However, it should be noted that although the LOGX2 approach improves the optimization efficiency and minimizes the potential systematic errors, it impacts the statistics of the solution. With the linear or LOG approaches the errors are Gaussian or log-normal, respectively, but with the LOGX2 scheme they are neither.

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Acknowledgments.

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Tables and Figures

Table 1. Annual total CO emission in different regions, from June 2004 to May 2005, constrained by MOPITT surface level and tropospheric profile retrievals. The relative difference on total (combustion + oxidation from biogenic VOCs) CO emission estimates is calculated by 2 * (CO_surface – CO_profile) / (CO_surface + CO_profile). The region defition is shown in Figure 1.

Table 2. Monthly mean mass of continental CO tracers (Tg) in the boundary layer (lower column) and the free troposphere (upper column). The upper fraction is calculated by Mass_upper / Mass_total. The region defition is shown in Figure 4.

Table 3. Total CO emission in different regions, in Jun-Aug 2004, constrained by MOPITT surface level and tropospheric profile retrievals. The region definition is shown in Figure 1.

Figure 1. Annual mean CO emissions from combustion sources and the oxidation of biogenic NMVOC and CH_4, averaged from June 2004 to May 2005. The unit is 10^{12} molec/cm^2/sec. The continental domains are defined with black boxes. The sub-continental domains in North America (US, Mexico, Alaska and Canada) are seperated based on the country boundaries.

Figure 2. (a) – (e) Annual/Seasonal mean scaling factors, using MOPITT V5J surface level data; (f) – (j) Annual/Seasonal mean scaling factors, using MOPITT V5J tropospheric profile data; (k) – (o) Difference between two scaling factors, calculated by middle panel (e, f, g, h) minus left panel (a, b, c, d).

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Moved down [15]: OSSE scaling factors for April 2006. The scaling factors represent the ratio of the estimated to true emissions. The ratio for the first guess is 0.5. The actual value is 1.0. Shown are the scaling factors obtained with: (a) the linear scaling factor optimization, (b) the LOG scaling factor optimization, (c) the LOGX2 scaling factor optimization.

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Moved down [17]: Black solid line shows the GMD monthly mean CO. Red solid line shows the free model simulation with original initial condition. The blue dash line is the assimilation result using MOPITT from 60°S to 60°N. The green dash line is the assimilation result from excluding the high latitude data.

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Deleted: 4. (a) – (c) Scaling factors without high latitude MOPITT data. (d) – (f) scaling factors with high latitude MOPITT data. (g) – (i) difference between two types of scaling factors.
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Figure 3. Monthly variation of regional combustion CO emission estimates.

Figure 4. Distribution of emissions used for the idealized 30-day tracer. The unit is 10^{13} molec/cm^2/sec.

Figure 5. 30-day tracer partial columns in the extratropics for June 2004. The unit is 10^{18} molec/cm^2. Note the difference in scales between the lower and upper tropospheric columns.

Figure 6. (a, b): Mean tropospheric OH column (10^{12} molec/cm^2) in July 2004; (c,d): Meridional mean OH concentration (10^6 molec/cm^3) between 20°N-40°N in July 2004.

Figure 7. Scaling factors with MOPITT surface level retrievals and their difference. (a) – (c) Scaling factors, using v5-07-08 OH; (d) – (f) Scaling factors, using v8-02-01 OH; (i) – (l) Difference between two scaling factors.

Figure 8. Atmospheric CO lifetime averaged zonally at 30°N-50°N and 10°S-10°N for August 2004, estimated using v5-07-08 (black solid line) and v8-02-01 (red dash line) OH fields.

Figure A1. Annual variation of monthly mean CO concentration at selected GMD sites and surface level CO in GEOS-Chem, sampled at the GMD sites. Black solid line shows the GMD monthly mean CO. Red solid line shows the free model simulation with original initial condition. The blue dash line is the assimilation result using MOPITT from 60°S to 60°N. The green dash line is the assimilation result from excluding the high latitude data.

Figure B1. OSSE scaling factors for April 2006. The scaling factors represent the ratio of the estimated to true emissions. The ratio for the first guess is 0.5. The actual value is 1.0. Shown are the scaling factors obtained with: (a) the linear scaling factor optimization, (b) the LOG scaling factor optimization, (c) the LOGX2 scaling factor optimization.

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Deleted: Figure 14. Relative difference of monthly mean tropospheric CO column, calculated as 2(CO_profile – CO_surface) / (CO_profile +CO_surface). (a) – (c) with v5-07-02 OH; (d) – (f) with v8-02-01 OH.