Sensitivity of top-down estimates of CO sources to GCTM transport

Avelino F. Arellano Jr. and Peter G. Hess

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[1] Estimates of CO sources derived from inversions using satellite observations still exhibit discrepancies. Here, we conduct controlled inverse analyses to elucidate the influence of model transport on the robustness of regional CO source estimates. We utilized Model of Ozone and Related chemical Tracers global chemical transport models (GCTMs) driven by National Centers for Environmental Prediction and European Centre for Medium-Range Weather Forecast reanalyses, and GEOS-Chem GCTM driven by Global Modeling and Assimilation Office assimilated meteorology to generate response functions for prescribed regional CO sources. We find that inter-model differences in CO due to differences in transport are within 10–30% of inter-model mean CO concentration. However, these differences can translate to regionally significant spread in source estimates. While we find that CO source estimates for East Asia and North Africa are reasonably robust, we find inconsistencies and inter-model spread of greater than 40% in our source estimates for Indonesia, South America, Europe and Russia. This indicates the need for rigorous assessment on uncertainties in top-down source estimates through model inter-comparisons and ensemble approaches.


1. Introduction

[2] The availability of near-global and long-term CO observations, in conjunction with global chemical transport models (GCTMs), has allowed the use of inverse methods in constraining regional CO sources [Arellano et al., 2004; Heald et al., 2004; Petron et al., 2004]. This has shed light on our understanding of fossil-fuel and biofuel use and on the patterns of biomass burning activity. However, the use of Bayesian inverse methods to estimate regional sources intrinsically involves assessing the robustness of the estimates by characterizing methodical and model errors. Current approaches to characterize these errors are typically ad hoc leading to discrepancies in the inverse estimates well beyond the error bound attributable to model and data errors specified in the analysis. This is especially true for CO source inversions where persistent discrepancies across top-down estimates are still present, precluding an accurate estimation of some poorly-known yet chemically important CO sources. The errors associated with such discrepancies are mainly attributed to errors in GCTMs and in addition, there are errors due to sampling, aggregation and instrumentation. While there has been model inter-comparisons relating to differences in modeled distribution of chemical species as well as research efforts to characterize GCTM errors [e.g., Mallet and Sportisse, 2006], the impact of such errors on the top-down estimates of CO sources has not been explored in detail. This is in contrast to the comparison of CO₂ constituent inversions conducted within the TransCom framework [Gurney et al., 2003].

[3] For the use of inverse estimates of CO sources to become meaningful to chemistry and regulatory studies, a more rigorous assessment of its uncertainties is deemed appropriate. Here, we demonstrate the need for such an assessment by illustrating the impact of model transport, among many other sources of error, on regional CO source estimates. The main objective of this paper is to conduct a sensitivity analysis on the differences in the model treatment of transport on top-down estimates of CO sources. This allows us to assess our confidence in emission estimates for various regions. Additionally, we show the inadequacy of current inverse methodology to represent model transport error which is currently estimated using a single GCTM.

2. Methodology

[4] Our inverse analysis is based on a Bayesian synthesis inversion approach, in which observations of CO concentration (y) are expressed as linear combinations of modeled CO concentration responses (K), resulting from a set of prescribed source functions (xₐ). With prior knowledge of the source distributions, typically derived from bottom-up emission inventories, we solve for a vector of optimal source strengths (x̂) to best match the modeled CO concentration (Kx̂) with the observation (y). In probability statistics notation, we want to determine in general, the conditional distribution p(x|y) ∼ N(μ, Σ) assuming a prior p(x) ∼ N(x₀, S₀) and a likelihood p(y|x) ∼ N(Kx, Sₑ) derived using a GCTM, where S₀, Σ and Sₑ are error covariances associated with the prior and posterior source functions and the model/data, respectively. This type of statistical method entails reasonably characterizing these error covariances, which at present are approximated in a variety of ad hoc ways.

[5] The sensitivity of inverse estimates of CO sources to model transport is examined by applying different offline GCTMs and meteorological input to estimate the CO concentration responses (K). In particular, we run for 2 years with 1 year spin-up: 1) the Model of Ozone and Related chemical Tracers (MOZART) v4 http://gctm.acd.ucar.edu with meteorology from National Centers for Environmental Prediction (NCEP) reanalysis at T21 (~5.6°×5.6°) horizontal resolution with 28 vertical levels (L. Emmons et al., “Sensitivity of chemical budgets to meteorology in MOZART-4,” manuscript in preparation, 2006),
Figure 1. Percentage difference of zonal-averaged modeled CO for July 2000 relative to the mean of the 3 models. Vertical levels are in hPa. The plots correspond to: (a) MZ4 NCEP, (b) MZ4 ERA40, and (c) GEOS-Chem.

2) MOZART v4 with meteorology from European Centre for Medium-Range Weather Forecast (ECMWF/ERA-40) reanalysis at T21 with 60 vertical levels, and 3) GEOS-Chem v5.5.03 http://www.as.harvard.edu/chemistry/trop/geos) with Global Modeling and Assimilation Office (GMAO/GEOS-3) assimilated meteorology at $4^\circ \times 5^\circ$ horizontal resolution with 30 vertical levels.

[s] MOZART rediagnoses the convective transport and boundary layer transport from Hack [1994] and Zhang and McFarlane [1995] convective parameterizations and the Holtslag and Boville [1993] boundary layer parameterization. GEOS-Chem inputs the mass-fluxes using Allen et al. [1996] convective parameterization and assumes full mixing within GEOS-diagnosed atmospheric boundary layer. In practice, both models use the same tracer advection scheme [Lin and Rood, 1996] with a pressure fixer. Otherwise, the model setup is the same in the 3 simulations. We prescribe the same OH fields for the CO sink from Fiore et al. [2003] and use the same prior CO source distributions $x_a$ and monthly-averaged CO column observation vector $y$ from Measurement of Pollution in The Troposphere (MOPITT) Phase 1 period (April 2000 to April 2001) as described in Arellano et al. [2006]. Briefly, our prior source for fossil/biofuel is taken from EDGARv2/GEIA inventory and includes chemical production of CO from NMHC oxidation due to fossil/biofuel use. Our biomass burning source is taken from Global Fire Emission Database v1 [van der Werf et al., 2003] while our prior biogenic CO source is calculated from a standard tagged-CO scheme of GEOS-Chem [Heald et al., 2004] and represents surface CO emissions due to oxidation of biogenic hydrocarbons. The observation vector $y$ is composed of selected MOPITT retrievals mainly based on the MOPITT data quality statement (e.g. we limit the profiles to those located between 50°S to 50°N with an a priori contribution <50% and with the 500 hPa mixing ratio > 40 ppbv). The CO columns were further re-gridded to $4^\circ \times 5^\circ$ horizontal resolution to facilitate comparison.

[7] Using the 3 sets of modeled response matrices ($K$), transformed accordingly using MOPITT averaging kernels at $4^\circ \times 5^\circ$, and with all other inverse components constructed in the same manner (i.e. $y$, $S_e$, $x_a$, $S_a$), we carry out 3 sets of time-independent inverse analysis to solve $\hat{x}$, where $\hat{x}$ consists of 13 geo-political regions of annually-averaged and combined fossil/biofuel and biomass burning CO sources and 1 global biogenic CO source (please refer to Figure S1 of the auxiliary material for definition of regions). CO from methane is computed in the respective models and is presubtracted in the observation vector. Its response function is very similar in all models and appears to be insensitive to model transport. The model/data error $S_e$ is specified for each set of analysis as a high frequency variance of the model and observation residuals in addition to a small error supplied from the retrieval [Palmer et al., 2003]. This approach implicitly assumes that the overall model-measurement bias is due to the errors in emissions. We use a minimum error threshold of about 8% of global mean CO column for each element of $S_e^{1/2}$ to reduce the larger impact of regions with low background variability and of data points with insufficient retrievals to estimate the sample statistics [Arellano et al., 2006]. Previous inverse studies estimate the model/data error to be globally about 20–30% of the observation with relatively higher errors within the continental regions [Palmer et al., 2003; Jones et al., 2003; Arellano et al., 2004]. The prior source error covariance $S_a$ is approximated in this work to be 50% of the corresponding source magnitude $x_a$.

[s] With this experimental design, differences in posterior estimates $\hat{x}$ can be solely attributed to model transport uncertainty. Representing CO transport in the model out of the boundary layer is particularly critical for inversions using MOPITT CO columns (and other satellite retrievals) where retrieval information is reported to be limited to the free troposphere [Deeter et al., 2004]. Hence, in this paper, inter-model differences in the transport of CO from the free troposphere are subsequently translated to differences in posterior estimates of the sources.

3. Results

3.1. Forward Model Results

[s] First, it is informative to look at the simulated differences in the CO distribution. Shown in Figure 1 is a comparison of the differences from the inter-model mean in zonally-averaged CO from the 3 simulations (MZ4 NCEP, MZ4-ERA40 and GEOS-Chem) during July 2000 (see Figure S2 of the auxiliary material for inter-model mean CO distribution). In the Northern Hemisphere mid-latitude troposphere, the CO signal in MZ4 NCEP (a) and MZ4 ERA40 (b) are similar, in contrast to GEOS-Chem (c), which has more CO near the surface and less above. All three models differ significantly in the tropics, with MZ4 ERA40 showing the largest convective signal. However, MZ4 ERA40 has much less CO in the stratosphere com-
pared to the other two simulations. Clearly, the differences in the treatment of transport results in systematic differences in modeled vertical structure of CO. Differences in spatial patterns of CO columns are also evident. As a particular example, we show in Figure 2 the differences between the 3 simulations in the column Indonesian and European CO sources as weighted with the MOPITT averaging kernels (see Figure S3 of the auxiliary material for corresponding MOPITT CO and inter-model mean total CO column distributions). The differences are most pronounced in the tropics and subtropics, apparently due to differences in convection between the simulations.

Quantitatively, the model spread in both CO zonal means and CO columns is within 10–30% of the mean CO concentration which is consistent with typical estimates of model/data error. A limited model inter-comparison of 7 regional and global models from Kiley et al. [2003] within the TRACE-P domain gives a model spread (in terms of RMS difference) of about 15% in all levels, 16% in the free troposphere, and 25% at the surface. When compared to selected National Oceanic and Atmospheric Administration (NOAA ESRL/GMD) CO surface measurements (see Figure S4 of auxiliary material for sample comparisons), the 3 model simulations vary, in terms of its relative distance from the observation, by about 17 to 27% of the mean difference, which is similar to results from Kiley et al. [2003].

### 3.2. Inverse Model Results

Shown in Figure 3 are results of the inverse analysis for each of the 3 simulations. For each source, we arbitrarily categorized each estimate according to its degree of consistency within the 3 simulations. Defining the relative model spread as the difference between the maximum and minimum of the 3 estimates divided by the mean, category A refers to posterior estimates where the model spread is <25%, category B refers to estimates where model spread is >25% and where the trend in the posterior estimates relative to the prior are moderately consistent between model simulations, and category C refers to estimates where the model spread is >25% and where the trend in posterior estimates relative to the prior are not consistent.

We first focus on category A and in particular on the estimates of anthropogenic CO source in East Asia. This region has been the subject of several inverse studies which consistently report that the current emission inventory is significantly underestimated [Arellano et al., 2004; Allen et al., 2004; Heald et al., 2004; Pétron et al., 2004]. All three model estimates (Figure 3) are consistent with those studies, suggesting that the magnitude of the emission estimate in East Asia is reasonably robust and the uncertainty in the published estimates (summarized by Heald et al. [2004]) can be mainly attributed to model transport error. Our estimate for Northern Africa is also consistent with previous inverse estimates by Arellano et al. [2006] and Pétron et al. [2004]. On the other hand, although our estimates for Southeast Asia, Central America and Oceania are consistent
across model simulations, they are difficult to compare to previous estimates since the regions defined in previous studies are slightly different from this work.

The model spread in category B, ranging from 38% (Southern Africa) to 50% (North America), is significantly higher than can be expected from a global model/data error of 10–30%, although the difference in the posterior estimates relative to the prior is still consistent between simulations. In the case of North American (NAM) region, all three models reduce the a priori source (representative of the 1990s), consistent with a decrease in the NAM emissions in recent decades [e.g., Parrish et al., 2002]. However, an analysis of the simulations show that MZ4 NCEP exhibits a stronger lifting of the NAM outflow over the North Atlantic, relative to MZ4 ERA40 and GEOS-Chem, and consequently has a higher CO column in the MOPITT space (i.e. higher CO response). As a result, the inverse analysis reduces the MZ4 NCEP NAM emissions the most so as to match the MOPITT observations in the North Atlantic. While it is possible that the NAM emission is decreasing in recent decades and is lower relative to our prior estimate which is representative of the 1990s, our posterior estimate using MZ4 NCEP seems unreasonable low. In the case of Southern Africa (SAF), our estimate is mainly constrained by MOPITT observations over the source region rather than downwind. Although the 3 simulations show a similar convective pattern over the source region, the convection in MZ4 NCEP is deeper, leading to increased CO transport away from the source region. The consequent reduction in the local CO response leads to a comparatively small reduction in SAF emissions.

For category C, it appears that the inferred source estimates are not robust based on the model spread, which ranges from 39% (BIOG) to 81% (IND). As shown in Figure 2, the CO response for the Indonesian source using MZ4 NCEP is enhanced over Indonesia compared to the other 2 simulations. This difference, which can be attributed to stronger convection in this region using NCEP (Figure 1), produces a prior CO distribution that closely matches MOPITT observations. This difference explains the large model spread in the estimates for Indonesian source. It is also likely that the differences in the South American regions are linked to convection. Perhaps more surprising are the differences in the mid-latitude sources. Over both Europe and Russia, GEOS-Chem suggests much lower emissions relative to the prior than the other 2 simulations. We note from Figure 1 that the mid-latitude free tropospheric simulated concentrations in GEOS-Chem are significantly different than the 2 models (see also Figure 2).

Finally, the largest difference in these simulations is in the global biogenetic (BIOG) CO source, where both MZ4 NCEP and MZ4 ERA40 suggest a much lower reduction in the estimates. Both MZ4 simulations, which show stronger convection in the tropics and subtropics relative to GEOS-Chem, have stronger global CO responses to the BIOG emissions resulting in a reduction of the BIOG emissions in the inversion. This results to higher global posterior CO total emissions for GEOS-Chem (2550 Tg) compared to MZ4 NCEP (2400 Tg) and MZ4 ERA40 (2361 Tg). It is not clear however if this large impact can be observed when BIOG emissions are constrained on a regional basis rather than globally as is done in this work.

4. Discussion

In summary, we find that estimates of sources from regions of East Asia and Northern Africa are robust regardless of the GCTM (and/or meteorological fields) used. On the other hand, we find that differences of 10–30% in simulated CO translate to differences in source estimates of greater than 40% for the regions of Indonesia, South America, Europe and Russia. Our results concur with those of Baker et al. [2006], under the TransCom framework, that agreements between regional CO2 flux estimates across 13 models only occur at few regions that are well-observed and/or exhibit highly distinct CO2 response. Our source estimates are sensitive to the treatment of transport specifically in regions of strong convection and transport out of the boundary layer. Here, the sensitivity is further amplified relative to reports from CO2 studies, since our estimates are largely constrained by observations in the free troposphere. It should be noted that due to the limited number of models used, these comparisons should be interpreted as indicative rather than definitive of the true sensitivities. In some cases, the spread in modeled response would be larger given different a priori emissions, different OH distributions, and the use of different observations in current CO inversions.

Nevertheless, it appears that the emission uncertainties presented above are not currently accounted for in our ad hoc procedure to estimate model/data error covariances, which is based on model-measurement statistics. In the absence of observations of regional sources, we need to characterize the uncertainties as probability distribution of all errors. To a first order, our procedure does not account for correlation of these model/data error in both space and time and assumes that the distribution is fully Gaussian. More importantly, uncertainties of the second kind (e.g. errors in model parameters) can only be accounted for, to a large extent, using ensemble-based approaches. Such bias is difficult to identify using a single GCTM. In fact, this sensitivity study clearly shows that model-measurement biases are not solely due to emission uncertainties, which is a main assumption for this type of inversion. Our results support the conclusion by Arellano et al. [2004, 2006] that the posterior error, based on the inverse analysis and shown in Figure 3 as 2-σ uncertainty, is significantly underestimated in most analysis particularly those using satellite observations, suggesting that the posterior error estimate is not an appropriate measure of source estimate uncertainty. We therefore recommend that current and future results from CO inverse analysis be interpreted with a more rigorous assessment of its associated uncertainties, perhaps by exploring ensemble inversion approaches in conjunction with independent data comparisons.
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References


A. F. Arellano Jr. and P. G. Hess, Atmospheric Chemistry Division, National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO 80307-3000, USA. (arellano@ucar.edu)