GEOPHYSICAL RESEARCH LETTERS, VOL. 31, L01104, doi:10.1029/2003GL018609, 2004

Correction published 22 June 2004

Top-down estimates of global CO sources using MOPITT measurements

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Received 11 September 2003; revised 12 November 2003; accepted 24 November 2003; published 8 January 2004.

[1] We present a synthesis inversion of CO emissions from various geographical regions and for various source categories for the year 2000 using CO retrievals from the MOPITT (Measurements of Pollution in the Troposphere) instrument. We find a large discrepancy between our top-down estimates and recent bottom-up estimates of CO emissions from fossil fuel/biofuel (FFBF) use in Asia. A key conclusion of this study is that CO emissions in East Asia (EAS) are about a factor of 1.8–2 higher than recent bottom-up estimates. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 3210 Mathematical Geophysics: Modeling; 3260 Mathematical Geophysics: Inverse theory; 3360 Meteorology and Atmospheric Dynamics: Remote sensing. **Citation:** Arellano, A. F., Jr., P. S. Kasibhatla, L. Giglio, G. R. van der Werf, and J. T. Randerson (2004), Top-down estimates of global CO sources using MOPITT measurements, *Geophys. Res. Lett.*, *31*, L01104, doi:10.1029/2003GL018609.

1. Introduction

[2] Carbon monoxide (CO), an important tropospheric chemical constituent, has natural and anthropogenic sources [*Logan et al.*, 1981]. Since CO is a ubiquitous by-product of various combustion processes, atmospheric CO measurements can provide potentially valuable information on the intensity of various anthropogenic activities. This feature has been exploited in various inverse modeling studies using surface and airborne CO measurements [*Bergamaschi et al.*, 2000; *Kasibhatla et al.*, 2002; *Pétron et al.*, 2002; *Palmer et al.*, 2003]. All of these CO inversion studies point to significant differences between top-down (i.e., derived using inverse modeling approaches) and bottom-up (i.e., derived using traditional inventory-based approaches) estimates of regional CO sources. In particular, these studies consistently suggest that bottom-up approaches underestimate anthropogenic CO emissions in the northern hemisphere, and point to Asian emission estimates as being particularly uncertain. However, the limited spatial and/or temporal scope of the surface and airborne CO measurements (coupled with uncertainties in the numerical chemical transport models used in the inversions) has significantly limited the amount of information that can be extracted with regards to CO emissions at regional scales. In this context, newly available space-based measurements of tropospheric CO from the MOPITT instrument [*Deeter et al.*, 2003] have the potential to provide improved estimates of CO sources due to the global extent of the dataset.

[3] Here, we use CO measurements from the first year of operation of MOPITT (year 2000) to derive top-down estimates of CO emissions from major geographical regions for key source categories. Our analysis is carried out under a range of methodological assumptions, and comparisons with other relevant studies are presented to assess the robustness of the derived CO source estimates.

2. Methodology

2.1. Inverse Approach

[4] In the present application, the modeled CO concentration field is described in terms of a linear combination of the individual source categories or basis functions. Measured CO concentrations $\mathbf{y}$ can be related to individual CO source strengths $\mathbf{x}$ according to

$$\mathbf{y} = \mathbf{K}\mathbf{x} + \mathbf{e}, \tag{1}$$

where $\mathbf{K}$ is the Jacobian matrix of response functions derived from a chemical transport model (CTM) and $\mathbf{e}$ is a vector of errors in both the modeled and measured concentrations. Assuming multivariate normal distributions for the prior of $\mathbf{x}$ ($\mathbf{x_a}$) and $\mathbf{e}$, the maximum a posteriori solution ($\mathbf{x}$) to the inverse problem, is given by

$$\hat{\mathbf{x}} = \mathbf{x_a} + \left(\mathbf{K^T S_e^{-1} K} + \mathbf{S_a^{-1}}\right)^{-1} \mathbf{K^T S_e^{-1}} (\mathbf{y} - \mathbf{K x_a}) \tag{2}$$

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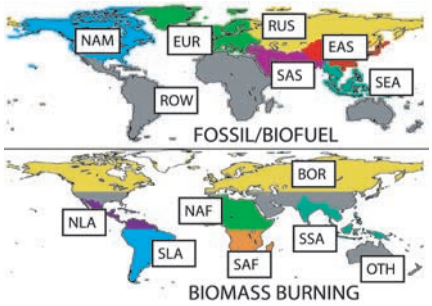

Figure 1. Definition of fossil fuel/biofuel (FFBF) and biomass burning (BB) source regions.

and the a posteriori error covariance matrix ($\hat{\mathbf{S}}$) is given by

$$\hat{\mathbf{S}} = \left(\mathbf{K}^{\mathbf{T}}\mathbf{S}_{\mathbf{e}}^{-\mathbf{1}}\mathbf{K} + \mathbf{S}_{\mathbf{a}}^{-\mathbf{1}}\right)^{-1}, \quad (3)$$

where $\mathbf{S_a}$ is the a priori error covariance matrix, and $\mathbf{S_e}$ is the observation error covariance matrix.

2.2. Definition of Basis Functions

[5] In this study, 15 individual CO source categories are considered, and consist of fossil fuel/biofuel (FFBF) combustion in 7 geographical regions, biomass-burning (BB) in 7 geographical regions, and the global CO source from the oxidation of isoprene and monoterpenes (BIOG). Figure 1 shows the definition of the geographical source regions for the FFBF and BB categories. CO from methane oxidation is calculated on-line in the CTM (see section 2.4) and is presubtracted from the measurements in the inversion.

2.3. Specification of a priori Emissions, $\mathbf{x_a}$

[6] The a priori FFBF and BIOG sources are specified as in our previous work [*Kasibhatla et al.*, 2002]. The FFBF source is based on the EDGARV2/GEIA CO source inventory [*Olivier et al.*, 1996], with an added component to account for the CO source from the oxidation of nonmethane hydrocarbons (NMHC) [*Kasibhatla et al.*, 2002]. The BIOG source is prescribed by scaling emissions of isoprene and monoterpenes [*Guenther et al.*, 1995] using CO yields as in *Kasibhatla et al.* [2002]. In addition, a time-of-day variation based on light availability is imposed on the CO source from isoprene. Monthly-varying CO emissions from biomass burning in 2000 are specified using an improved methodology which incorporates satellite observations of fires, biogeochemical modeling of available biomass, and biome-specific CO emission factors [*Van der Werf et al.*, 2003].

[7] The matrix $\mathbf{S_a}$ is assumed to be diagonal, with the a priori error set equal to 50% of the corresponding a priori source estimate to provide a weak constraint on the inverse solution.

2.4. Calculation of Jacobian Matrix, K

[8] Response functions for each source category are calculated using the GEOS-CHEM CTM driven by NASA/GMAO assimilated meteorological fields [*Bey et al.*, 2001]. To linearize the model, the chemical loss of CO is calculated using prescribed monthly-mean OH fields from a full tropospheric chemical simulation [*Bey et al.*, 2001]. The OH fields are scaled down by 20% so that the corresponding global lifetime of methyl chloroform against the tropospheric OH sink is 6.6 years, which is consistent with recent estimates by *Prinn et al.* [2001].

[9] The chemical production of CO from methane (CH_4) oxidation is calculated in the CTM using prescribed OH fields and prescribed CH_4 concentration fields (90°S–30°S: 1706 ppbv; 30°S-EQ: 1716 ppbv; EQ-30°N: 1760 ppbv; 30°N–90°N: 1814 ppbv) representative of the late 1990s [*Dlugokencky et al.*, 2001]. The CO yield from CH_4 oxidation is assumed to be 0.95 [*Kasibhatla et al.*, 2002].

2.5. Processing of MOPITT CO Retrievals

[10] The Level 2 V3 MOPITT dataset used in this study consists of retrieved CO mixing ratios for 7 vertical levels in the atmosphere nominally corresponding to the surface, 850, 700, 500, 350, 250 and 150 mb [*Deeter et al.*, 2003]. However, the number of independent pieces of information in each retrieved profile is less than 2 [*Heald et al.*, 2003]. The MOPITT retrieval averaging kernels typically peak in the middle troposphere (700 to 350 mbar) and have a very low sensitivity to surface concentrations.

[11] In the present application, a subset of MOPITT CO retrievals from the first year of measurements (April–December 2000) is selected using a quality control procedure that is based on the MOPITT data quality statement. In particular, we restrict our analysis to CO retrievals: (i) containing all 7 standard levels between 50°S and 50°N, (ii) having <50% a priori contribution at the 350, 500, and 700 mb levels, and (iii) with retrieved 500 mb mixing ratio >40 ppbv. The first criterion accounts for most of the data that is discarded and is based on the data quality statement that high latitude data should be used with caution. The second and third criteria ensure that the measurement vector is representative of observed CO rather than MOPITT a priori information and is not contaminated with physically unrealistic observations. CO columns derived from the MOPITT CO profile retrievals are used in the inversion analysis. The individual CO columns are averaged over the $4° \times 5°$ horizontal CTM grid and over a daily time period. A monthly average is then calculated for each model grid box with at least 15 days of data in the month. A total of 11,432 elements are used in the analysis from a maximum of 16,848 elements that can be constructed if the 15-day criterion was satisfied for all grid boxes in each month. Sensitivity analysis showed that this criterion did not significantly affect the key results of the inversion. In addition to column CO, the 500 mb and 700 mb retrievals are also used in separate inversions as an independent check on the sensitivity of the a posteriori source estimates to the choice of dataset.

[12] To construct the matrix $\mathbf{K}$, the averaging kernels from the MOPITT retrievals are applied to the model CO profiles corresponding to each basis function. The vector $\mathbf{y}$ is constructed by removing the contribution of the a priori CO profile used in the MOPITT retrieval algorithm from each retrieved profile.

2.6. Specification of Observation Error Covariance, $\mathbf{S_e}$

[13] The matrix $\mathbf{S_e}$ is constructed as the sum of a model plus representation error covariance matrix ($\mathbf{S_m}$) and a retrieval error covariance matrix ($\mathbf{S_r}$). $\mathbf{S_r}$ is prescribed using the error covariance matrices provided as part of the MOPITT retrievals. $\mathbf{S_m}$ is assumed to be diagonal, with

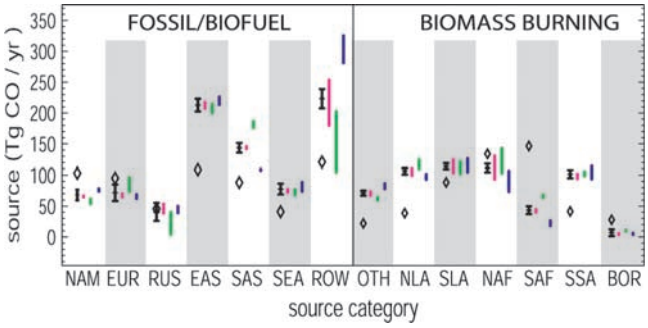

Figure 2. Top-down estimates of CO sources derived using column (magenta lines), 500 mb (green lines), and 700 mb (blue lines) MOPITT CO retrievals. The vertical extent of the lines denotes the range of estimates for the 4 error scenarios considered. The a priori source estimates (black diamonds) and mean a posteriori 2-σ uncertainties (black error bars) are also shown for each source category. The a posteriori estimate for CO from global biogenic oxidation is 150–240 Tg CO/yr compared to the a priori source of 460 Tg CO/yr.

elements specified using the variance of daily-averaged CO fields about the corresponding monthly-mean fields. In each inversion, all elements of the resulting matrix are uniformly scaled such that the a posteriori goodness-of-fit parameter is close to unity. The underlying assumption is that spatial structure of the temporal variance field can be used as a surrogate for the spatial structure of the model error field, a concept that has been widely used in many inverse modeling studies [*Enting*, 2002, and references therein]. Recognizing the ad hoc nature of this assumption, we carry out the inversions using the following 4 approaches to specify the temporal variance at each model grid box for each month: (1) variances derived from the retrievals only, (2) variances derived from the model fields only, (3) the larger of the model- and retrieval-derived variances (4) variances derived from the residual (MOPITT-a priori model) fields. Our approach thus represents an attempt, albeit limited, to explore the issue of sensitivity to error specification. For the cases considered here, the model plus representation error is typically 5–30% of the corresponding monthly-mean, and accounts for 98–99% of the specified total observation error.

3. Results and Discussion

[14] Source and error estimates[1] from the inverse analysis are summarized in Figure 2 for 12 case scenarios corresponding to the use of 3 retrieval datasets (column, 700 mb, and 500 mb CO) and 4 different assumptions for constructing $\mathbf{S_m}$. Figure 3 shows comparisons of modeled CO with independent surface CO measurements from the NOAA/CMDL Cooperative Air Sampling Network [*Novelli et al.*, 1998] at several key stations.

[15] It can be seen from Figure 2 that the a posteriori errors derived from the inverse analysis are small for all source categories, a finding that is expected given the large number of data points in the MOPITT dataset. Consequently, the well-defined model resolution matrices (not shown here) indicate that the data provide strong constraints on source magnitudes. However, it is worth noting the range of the maximum a posteriori estimates (i.e., $\hat{\mathbf{x}}$) for some source categories (e.g., FFBF/SAS, FFBF/ROW, and BB/NAF) is much larger than the corresponding mean a posteriori error, indicating that the latter quantity does not provide a true indication of the uncertainty in the derived source estimates. As shown in Figure 2, the range of these source estimates is largely driven by the choice of dataset and partly influenced by the different error specifications. In this context, it is also worth noting that the 12 cases represent a limited exploration of the sensitivity to methodological assumptions. Thus, the true uncertainties are likely larger than the ranges shown in Figure 2. Nevertheless, it is worth comparing a posteriori and a priori source estimates, especially for source categories where the a posteriori errors are similar in magnitude to the range of estimates derived from the sensitivity analysis.

[16] For the FFBF source categories, there are striking differences between the a priori and a posteriori estimates in Asia. In all 3 Asian sub-regions, the FFBF top-down estimates are significantly higher than the corresponding bottom-up estimates. It is evident from Figure 3 that the larger a posteriori source magnitudes result in a better agreement of model CO with the independent NOAA/CMDL surface measurements at sites downwind of Asia (GMI, MID, MLO). However, it should be noted that the estimated FFBF/SAS source magnitude is sensitive to the methodological assumptions used. This is also true for the FFBF/ROW source, where our estimates range from 165 to 327 Tg CO/yr, indicating that further investigation is needed to better quantify this potentially large source.

[17] Several recent studies have focused on CO sources in Asia in general, and East Asia in particular. The FFBF/EAS CO source is estimated to be 115 Tg CO/year in the most up-to-date inventory for this region [*Streets et al.*, 2003], again significantly smaller than our estimate of 195–

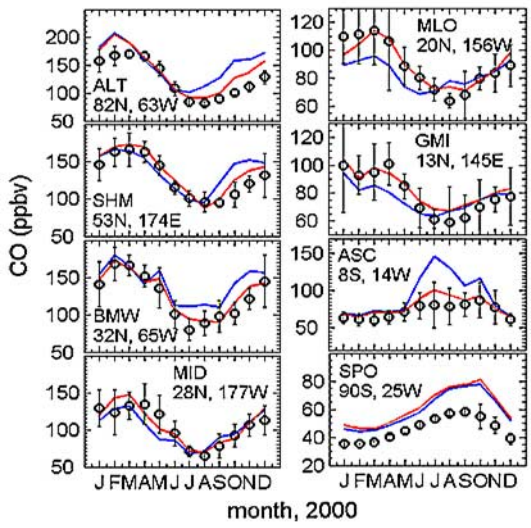

Figure 3. Comparison of modeled CO using a priori (blue solid line) and a posteriori (red solid line) source estimates with observed monthly-mean (circles) at selected NOAA/CMDL sites. Error bars represent the standard deviation of individual measurements between 1999 and 2001 about the monthly mean.

[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2003GL018609.

215 Tg CO/year. *Carmichael et al.* [2003] noted a large discrepancy between modeled CO using the *Streets et al.* [2003] inventory and the Spring 2001 TRACE-P aircraft observations over the Yellow Sea. They suggested that CO emissions from the domestic sector are possibly underestimated in the *Streets et al.* [2003] inventory by a factor of 2 or more. Scaling the *Streets et al.* [2003] FFBF domestic emissions by a factor of 2 to 3 would yield a FFBF source strength of 160–200 Tg CO/yr in East Asia, which is in closer agreement with our analysis. It is also worth noting that a similar scaling applied to South and Southeast Asia would increase the *Streets et al.* [2003] FFBF estimates for these regions to 100–140 and 50–75 Tg CO/year, which are comparable to the results of this study. Our estimate of the FFBF/EAS source magnitude is also very consistent with results from a recent inversion study using TRACE-P (Feb–Apr 2001) aircraft measurements by *Palmer et al.* [2003]. They estimated that about 190 Tg CO/yr is emitted due to fossil fuel and biofuel use in China, Korea and Japan, which is very similar to the estimates presented here (195–215 Tg CO/year). Our results regarding the Asia FFBF source are also consistent with inversions based on ground-based CO measurements that suggest that anthropogenic Asian CO sources are significantly underestimated in bottom-up inventories [*Pétron et al.*, 2002; *Kasibhatla et al.*, 2002].

[18] Among the BB source categories, Figure 2 shows significant differences between the bottom-up and top-down estimates for the BB/OTH, BB/NLA, BB/SAF, and BB/SSA categories. While it is difficult to compare these findings to other studies in these regions due to the potentially large interannual variability in regional biomass-burning emissions, some aspects of these results are worth noting. The comparisons with surface CO measurements at Ascension Island shown in Figure 3 support our finding of a lower BB/SAF source. Our estimate of 95–120 Tg CO/year for the BB/SSA source is significantly higher than the *Streets et al.* [2003] climatological estimate of 48 Tg CO/year. More significantly, our total anthropogenic source estimate for Southeast Asia (FFBF source of 70–90 Tg CO/year plus a significant portion of the 95–120 Tg CO/year estimated for the BB source in South Asia and Southeast Asia combined) is much higher than the corresponding estimate of 35 Tg CO/year by *Palmer et al.* [2003] based on TRACE-P CO measurements. However, the analysis of TRACE-P ethane, propane, and CO measurements by *Carmichael et al.* [2003] using a different biomass burning emissions inventory does not support the *Palmer et al.* [2003] estimate, but rather suggests that total anthropogenic CO emissions in Southeast Asia are likely higher than the *Streets et al.* [2003] estimate of 66 Tg CO/year. The wide range of estimates for CO emissions from Southeast Asia clearly indicates a need for better characterizing the magnitude and variability of anthropogenic emissions in this region.

[19] Finally, it should be noted that our estimate of the global biogenic CO source ranges from 150 to 240 Tg CO/year. This is significantly smaller than the bottom-up estimate of 460 Tg CO/year. Again, it is difficult to evaluate this aspect of our analysis. As seen in Figure 3, the lower biogenic source combined with a lower BB/BOR source results in a better agreement of modeled CO with surface measurements in the mid- and high latitudes of the Northern Hemisphere (ALT, SHM, and BMW) but not in the high latitudes of the Southern Hemisphere (SPO). An added complication that may influence these results is that the MOPITT CO retrievals during 2000 may be biased in low CO regions [*Deeter et al.*, 2003].

4. Summary and Future Directions

[20] This study represents a first attempt to use remote sensing CO measurements to quantify regional CO sources. The discrepancies between the top-down source estimates derived here and the bottom-up source estimates derived using traditional inventory-based approaches highlights the uncertainties in CO source estimates on regional scales. Our study adds to the growing body of evidence that suggests that anthropogenic CO emissions in Asia are significantly higher than generally assumed. To the extent that this is true, an important next step will be to determine whether the discrepancy is due to an underestimate of the magnitude of fuel use in general or due to an underestimate of emission factors of CO from certain source categories in particular.

[21] In terms of future inverse modeling studies using MOPITT CO measurements, there is a need to explore the sensitivity of the inverse results to (i) the specified a priori time- and spatial-dependence of sources by using different emission inventories, (ii) the CTM used to calculate the Jacobian matrix, (iii) the OH fields and assumed model linearity, and (iv) the prescribed structure of the a priori error covariance matrices. Furthermore, a better characterization of MOPITT CO retrieval errors is needed in light of the biases evident in limited comparison with in situ data. Lastly, sources such as the FFBF/ROW source, which seem to show significant differences in estimates, warrant further investigation.

[22] **Acknowledgments.** We acknowledge the NCAR MOPITT and NOAA CMDL Carbon Cycle Groups for providing CO measurements. We thank Bob Yantosca for help with the GEOS-CHEM model. This work was supported by NASA ESE/IDS Grants NAG5-9605 and NAG5-9462.

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