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Seasonal cycle of emissions of CO inferred from MOPITT profiles of CO: Sensitivity to pyroconvection and profile retrieval assumptions

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[1] We estimate monthly continental-scale CO emissions for 2006 by optimally fitting prior emissions used by the GEOS-Chem chemistry transport model to retrieved profile measurements of CO from the Measurement Of Pollution In The Troposphere (MOPITT) satellite instrument. We focus on the range of emission estimates obtained by using different versions of the MOPITT profile data, and by better describing enhanced vertical mixing of emissions from wildfires. We find that annual posterior CO emissions estimates for 2006 range from 1003 to 1180 Tg CO, within the range of prior estimates (1243 ± 617 Tg CO). We generally find larger differences in posterior CO emissions from using different versions of the MOPITT data than from improving the description of wildfires, with the exception of fires over Indonesia. Posterior emissions over regions with wildfires have a large seasonal cycle, as expected, which can be substantially different from prior emission estimates. We find GFDL v.2 prior emissions underestimate the duration of the biomass burning season for North Africa by as much as 1 month. We also find posterior emissions over Indonesia are a factor of 2 higher than prior emissions (83 ± 42 Tg CO) in 2006 due to widespread fires during July–December. Posterior emissions over Canada during 2006 are a factor of 2–3 higher than prior emissions (9 ± 4.6 Tg CO). We also find a seasonal cycle of CO emissions over North America and Europe, in agreement with previous studies, which is not described by prior emissions. Citation: Gonzi, S., L. Feng, and P. I. Palmer (2011), Seasonal cycle of emissions of CO inferred from MOPITT profiles of CO: Sensitivity to pyroconvection and profile retrieval assumptions, Geophys. Res. Lett., 38, L08813, doi:10.1029/2011GL046789.

1. Introduction

[2] Carbon monoxide (CO), a tracer of incomplete combustion, is the major sink for the hydroxyl radical (OH), the main tropospheric oxidant, and consequently is important for determining the oxidizing capacity of the troposphere. The main direct sources of CO include the incomplete combustion of biomass, fossil fuels and biofuels. There is also a substantial source of CO from the oxidation of volatile organic compounds (VOCs) co-emitted with CO as products of incomplete combustion. The largest contribution to the CO budget via the oxidation chain is from methane, isoprene, methanol, monoterpenes, and acetone [Müller and Stavrakou, 2005]. The atmospheric lifetime of CO, determined largely by OH oxidation, is typically a couple of months but is a function of latitude and season. There remains substantial uncertainty in year to year changes in regional emission estimates, determined from surface, aircraft, and space-borne instruments, particularly where the seasonal cycle is determined primarily by wildfire and agricultural burning emissions.

[3] Here, we use profile retrievals from the MOPITT satellite instrument to infer the seasonal cycle of surface emissions of CO over major regions of continental outflow during 2006. In particular, we quantify the sensitivity of posterior emission estimates to changes in MOPITT data products and to changes in the underlying assumptions about the vertical transport of wildfire emissions.

2. MOPITT Data

[4] The MOPITT instrument, launched in December 1999 aboard the NASA EOS Terra satellite, has measured global CO concentrations with a local overpass time of 10:30 since 2000. Global coverage is achieved within 3 days, with typically more than 10^5 profile measurements/day. CO profiles are retrieved from radiance measurements in the thermal infrared channel at 4.7 μm [Deeter et al., 2003]. We use two versions of the level 2 data: 1) v3 which include CO concentrations, averaging kernels and error covariances on 7 pressure levels retrieved in linear space (surface, 850, 900, 500, 350, 250, and 150 hPa); and 2) v4 which includes the same variables as in v3 but on 10 pressure levels retrieved in logarithmic space (surface, 900, 800, 700, 600, 500, 400, 300, 200, and 100 hPa) [Deeter et al., 2009]. The logarithmic space MOPITT data product has been developed to capture large profile variations of CO, e.g., over wildfires, which are often not well captured by the linear product resulting in a bias (M. Deeter, NCAR, private communication, 2009). There are typically 1.5 pieces of information in the MOPITT CO profile, but over intense wildfires where there is a thermal contrast between lower and upper troposphere there is enough information to independently distinguish 2 crude vertical regions [Deeter et al., 2004].

[5] For our analysis, we only use cloud-free retrievals that are not predominately determined by prior information. We also adopt the quality control procedures outlined by the MOPITT documentation [Deeter et al., 2009]. Previous work, using independent aircraft CO measurements during 2006, showed that MOPITT has a bias between 10–20% at 700 hPa [Emmons et al., 2007, 2009], which we do not
include in our study. This bias is unlikely to be globally constant, and it is unclear how to extrapolate the regional bias to large regions as would be necessary for our study.

3. Forward Model Description

[6] The forward model includes 1) the global 3-D GEOS-Chem transport model (v7.04.10, www.geos-chem.org) that relates surface CO emissions to global 3-D CO concentrations, 2) interpolation to the time and location of each MOPITT measurement, and 3) the convolution of the interpolated model CO profile with scene-dependent MOPITT averaging kernels, which take into account the vertical sensitivity of MOPITT to CO.

[7] The GEOS-Chem model, driven by meteorological analyses from the NASA Goddard Earth Observing System v4 (GEOS 4), has a horizontal resolution of 2 × 2.5º and 30 sigma levels that span the surface to 0.01 hPa, 12 of which are below 10 km. We use monthly mean biomass burning emissions estimates from the Global Fire Emission Database (v2) [van der Werf et al., 2006]. Fossil fuel distributions are based on values that have been scaled to 2006 values using CO2 emissions from fossil fuel burning, cement manufacture, and gas flaring [Palmer et al., 2006], with emission estimates over eastern Asia taken from more recent work [Streets et al., 2006]. Biofuel emission estimates are taken from Yevich and Logan [2003]. We account for the secondary source of CO from the oxidation of VOCs by using empirical scaling factors based on oxidation yields determined by laboratory studies [Duncan et al., 2007], and applying them to prior emission estimates of methane, isoprene, methanol, monoterpenes, and acetone. We use pre-computed monthly OH fields from a full-chemistry simulation relevant to this simulation, enabling us to linearly decompose CO contributions originating from individual sources and geographical regions, which greatly simplifies the inverse model calculation. Figure 1 shows the 9 geographical regions used in this study for which we resolve monthly total direct CO emissions (the sum of biomass burning, and the combustion of fossil fuels and biofuels) for 2006. We further include 9 background tracers that describe emissions prior to 2006, and 1 tracer describing the annual secondary production of CO from CH4 and non-CH4 VOCs. This results in 9 × 12 + 9 + 1 (=119) tracers for 2006. We determine CO concentrations on 1st January 2006, after spinning up through 2005 to remove initial conditions, by inferring emissions estimates from MOPITT (v3 or v4) measurements (see below for method) during October–December 2005.

[8] We address the sensitivity of our emission estimates to assumptions about the enhanced vertical mixing of emissions from surface heating of the atmosphere from fires (known as pyroconvection). The control calculation assumes emissions are confined to the boundary layer. We account for pyroconvection by using a regional climatology for 2006 that combined independent satellite measurements of CO at thermal and microwave wavelengths [Gonzi and Palmer, 2010]. This study showed that generally only 10–20% of boreal and tropical fire emissions reached the free and upper troposphere during June–October 2006 but this led to a disproportionate change in CO concentrations, depending on the region.

4. Inverse Model Description

[9] We use a maximum posterior (MAP) optimal estimation approach to fit the surface CO emission estimates to MOPITT CO profile measurements. For brevity, we only provide the essential details of the approach here and refer the reader elsewhere for a more detailed description of the method [Rodgers, 2000].

[10] The ultimate objective of the MAP approach is to maximise the posterior probability density function describing the state vector x (monthly CO emission estimates for the 9 continental regions as described above) given the measurement vector y (MOPITT CO profile measurements), and their respective prior error estimates Sx and observation error Sr. The general iterative approach used to achieve this objective with the linear and logarithmic space MOPITT data is summarised by:

\[ x_{i+1} = x_i + (K_i^T S^{-1}_r K_i + S_i^{-1})^{-1} K_i^T S^{-1}_r (y - F(x_i)) + S_i^{-1}(x_i - x_a) \]  

(1)

\[ \hat{S} = (S_x^{-1} + K_i^T S^{-1}_r K_i)^{-1} \]

(2)

\[ S_r = S_{MOPITT} A^T + S_{\text{noise}} \]

(3)

where xi is the i\(^{th}\) iteration of the state vector; xa is the prior state vector; Ki is the i\(^{th}\) iteration of the Jacobian matrix that describes the sensitivity of the measurement vector to changes in the state vector (i.e., ğy/ġx); F(x) is the forward model; S is the posterior error covariance matrix; and all other variables are as previously defined. The superscripts -1 and T denote matrix inverse and transpose operations, respectively. K describes the posterior Jacobian.

[11] The diagonal matrix S_{MOPITT} represents the sum of forward model error and representation error. We assume a combined error of 50% of observed concentrations. We smooth S_{MOPITT} by the MOPITT averaging kernels A to account for the correlations between different retrieval levels. Finally, we add operational MOPITT retrieval error covariance matrices S_{\text{noise}} to build the observation error matrix Sr. In the prior error covariance matrix Sx we assume a uniform 20% uncertainty for direct continental emissions in the state vector and a 20% uncertainty for the secondary source of CO from the oxidation of CH4 and non-CH4 VOCs. We thin the MOPITT data, reflecting the large number available and to avoid correlations between neighbouring observations, by
using only one observation per $2 \times 2.5^\circ$ model grid box per day.

We find that our problem is near-linear, with typically only one iteration required to satisfy our convergence criteria, as expected \cite{Rodgers2000} ($x_i - x_{i+1} S(x_i - x_{i+1}) < n$, where $n$ is the number of state vector elements. We have performed a number of sensitivity tests in which we perturbed the forward model and prior errors and we find that our results are robust.

5. Results

Figure 2 shows monthly prior and posterior emission estimates inferred from v3 and v4 MOPITT CO profile data during 2006. For the main text, we focus on NAF, INDO, NA1, and EU; results for SAF, SAM, NA2, BA, and AS are mentioned in the text but presented in the auxiliary material. Emissions from ROW are negligible (3 Tg CO/yr, originating mainly from New Zealand) and are not discussed further.

Table 1 shows the annual CO emission estimates for each of the 9 study regions. We find that posterior emissions are significantly higher than prior values over NA2, BA, and INDO; and lower than prior values over AS, SAM. Over NA1, EU, NAF, and SAF there is agreement, within uncertainty, between prior and posterior emission estimates. We generally find that posterior emission estimates are less sensitive to the prescribed vertical transport of emissions than the differences resulting from using v3 and v4 of the MOPITT data, except over regions experiencing extensive fires, e.g., Indonesia during October 2006.

Relative to our control calculation, we find that the GEOS-Chem model prior CO profiles in MOPITT space that account for fire-related vertical mixing, only slightly a) improve the mean Pearson correlation with MOPITT data (from 0.76 to 0.77), and b) decrease the mean positive bias with MOPITT data (typically by 1–3 ppb) with an insignificant change to the associated mean standard deviations (which remain 20 ppb, 13 ppb and 20 ppb in the lower, free, and upper troposphere, respectively). The corresponding posterior CO emissions are generally only a few percent larger than without this additional vertical mixing, which is partly due to the typically small amount of injected mass relative to the existing CO mass above the boundary layer and partly due to the vertically broad MOPITT averaging kernels. We anticipate that instruments with better vertical resolution will be more sensitive to changes to pyroconvection. The differences arising from using CO profile data retrieved in linear and logarithmic-space are largest over NA1, EU, NAF, INDO, and SAM, with posterior emissions largest for the linear v3 data product; this is consistent with our analysis of v3 and v4 MOPITT column observations (not shown). The model has a negative global CO bias of $-20\%$–$5\%$ relative to v3 MOPITT data and a

Table 1. Regional Annual CO Emissions [Tg CO ± Prior Uncertainty] Inferred From MOPITT Level 2 Profile Concentration Data (v3 and v4) and the Sensitivity to Enhanced Vertical Mixing (EVM) Due to Surface Fires

<table>
<thead>
<tr>
<th>Region</th>
<th>Prior</th>
<th>v3 + EVM</th>
<th>v3</th>
<th>v4 + EVM</th>
<th>v4</th>
</tr>
</thead>
<tbody>
<tr>
<td>NA1</td>
<td>162.9 ± 77.0</td>
<td>161.3</td>
<td>155.9</td>
<td>110.4</td>
<td>107.7</td>
</tr>
<tr>
<td>NA2</td>
<td>9.1 ± 4.6</td>
<td>17.2</td>
<td>16.3</td>
<td>28.0</td>
<td>21.0</td>
</tr>
<tr>
<td>EU</td>
<td>175.9 ± 87.9</td>
<td>153.9</td>
<td>153.4</td>
<td>116.3</td>
<td>114.8</td>
</tr>
<tr>
<td>BA</td>
<td>34.7 ± 17.4</td>
<td>54.7</td>
<td>52.8</td>
<td>68.5</td>
<td>59.6</td>
</tr>
<tr>
<td>AS</td>
<td>386.5 ± 193.2</td>
<td>241.9</td>
<td>232.4</td>
<td>271.3</td>
<td>270.2</td>
</tr>
<tr>
<td>NAF</td>
<td>141.1 ± 70.6</td>
<td>171.8</td>
<td>169.8</td>
<td>104.0</td>
<td>107.4</td>
</tr>
<tr>
<td>SAF</td>
<td>113.0 ± 56.5</td>
<td>112.6</td>
<td>107.9</td>
<td>134.5</td>
<td>130.9</td>
</tr>
<tr>
<td>SAM</td>
<td>83.3 ± 41.6</td>
<td>182.4</td>
<td>180.5</td>
<td>152.8</td>
<td>131.7</td>
</tr>
<tr>
<td>Total</td>
<td>136.3 ± 68.1</td>
<td>82.0</td>
<td>71.4</td>
<td>68.3</td>
<td>59.3</td>
</tr>
</tbody>
</table>

1Auxiliary materials are available in the HTML. doi:10.1029/2011GL046789.
global bias of −10% to +5% relative to v4 MOPITT data (auxiliary material).

[16] Regions where CO emissions are dominated by fire have the largest seasonal cycle, e.g., NAF and INDO, as expected. We find that over NAF, where emissions peak in winter and spring months, posterior CO emissions are generally larger than prior estimates in April and May, although there is substantial differences between results that use v3 and v4 MOPITT data. We acknowledge that we estimate only total CO emissions, but we believe this discrepancy is most likely due to error in biomass burning emissions that could simply reflect the temporal resolution of the inventory. Our results for NAF are broadly consistent with recent work [Chevallier et al., 2008] that used MOPITT CO columns to infer surface CO emissions, but our posterior results for SAF (auxiliary material) are 10–30% higher [Chevallier et al., 2008].

[17] We also found that MOPITT data supports a seasonal cycle of CO emissions over North America (NA1) and Europe (EU) with a summertime minimum, a result supported by surface CO concentration measurements [Holloway et al., 2000; Miller et al., 2008]. These regions are thought to be determined by emissions from fossil fuel and biofuel combustion although recent analysis of aircraft observations of outflow from the contiguous US showed that the summertime CO is dominated by the oxidation of VOCs [Hudman et al., 2008]. Studies have attributed the seasonal variation to lower summertime emissions when OH concentrations are higher [Holloway et al., 2000] and to underestimating emissions from cold starts from cars and wood burning [Kopacz et al., 2010].

[18] We estimate an anterior CO source from the oxidation of CH4 and non-CH4 VOCs of 1248 Tg CO and 1550 Tg CO for MOPITT v3 and v4 data, respectively, compared to our prior source of 1242 ± 249 Tg CO. These values are within the range of published estimates but sit at the lower end: 1200–1400 Tg CO inferred using MOPITT CO columns from 2000 and 2001 [Stavrakou and Müller, 2006]; 1650 Tg CO inferred using MOPITT CO columns from 2000–2001 [Petron et al., 2004]; 1500 ± 40 Tg CO inferred from surface concentration measurements 1990–1996 [Petron et al., 2002]; and 1290 Tg CO inferred using MOPITT in combination with AIRS, SCIAMACHY, and TES observations [Kopacz et al., 2010].

[19] We acknowledge that errors resulting from a Bayesian analysis are artificially small, reflecting the common assumption that prescribed errors are random and Gaussian; as a consequence, we have chosen not to report them. Sources of systematic error (e.g., measurement bias or aggregation error due to coarse spatial and temporal resolution) are difficult to quantify accurately and neglected in this study.

6. Concluding Remarks

[20] We found that regional posterior emission estimates inferred from MOPITT are generally more sensitive to recent changes in the formulation of the level 2 CO profile measurements than to a more realistic (albeit still crude) description of pyroconvection except for large fires, e.g., Indonesia, October 2006. We acknowledge that this is not a result that can necessarily be applied to all CO sensors, especially those instruments that measure complementary spectral regions (e.g., short-wave and thermal) and/or that have better spectral resolution.

[21] Our results are broadly consistent with previous studies that show large seasonal cycles of emissions over regions with burning seasons, e.g., Africa, Canada, and Southeast Asia. We found unexpected but previously reported seasonal cycles over the contiguous US and Europe, and to a lesser extent Asia (Figure S1). We also found a longer than expected burning season over North Africa, which could be explained by the temporal resolution of the biomass burning emission inventory. These results are robust and not significantly affected by the MOPITT data product.

Appendix A

[22] The auxiliary material gives a short description of the implementation of fire-related elevated vertical mixing in the forward model; the posterior CO emission estimates for SAF, SAM, NA2, BA, and AS; and a comparison of MOPITT v3 and v4 CO columns and the corresponding GEOS-Chem model columns.

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