Global estimates of CO sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS, SCIAMACHY, TES)

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Abstract

We combine CO column measurements from the MOPITT, AIRS, SCIAMACHY, and TES satellite instruments in a full-year (May 2004 - April 2005) global inversion of CO sources at 4°x5° spatial resolution and monthly temporal resolution. The inversion uses the GEOS-Chem chemical transport model (CTM) and its adjoint applied to MOPITT, AIRS, and SCIAMACHY. Observations from TES, surface sites (NOAA/GMD), and aircraft (MOZAIC) are used for evaluation of the a posteriori solution. Global intercomparison of the different satellite datasets using GEOS-Chem as a common intercomparison platform shows consistency between the satellite datasets and with the in situ data. The majority of the differences between the datasets can be explained by different averaging kernels and a priori information. The global CO emission from combustion as constrained in the inversion is 1350 Tg a^{-1}, with an additional 217 Tg a^{-1} from oxidation of co-emitted VOCs. This is much higher than current bottom-up emission inventories. Consistent with both the satellite and in situ data, a large fraction of the correction results from a seasonal underestimate of CO sources at northern mid-latitudes and suggests a larger-than-expected CO source from vehicle cold starts and residential heating. A posteriori emissions also indicate a general underestimation of biomass burning relative to the GFED2 inventory. However, the tropical biomass burning constraints are not consistent across the different datasets. Although the datasets reveal regional inconsistencies over tropical biomass burning regions, we find the global emission estimates to be a balance of information from all three instruments.

1. Introduction

Carbon monoxide (CO) is a product of incomplete combustion and atmospheric oxidation of volatile organic compounds (VOCs). It has an atmospheric lifetime of about two months against oxidation by the OH radical. It is of interest as a sink for OH, the main tropospheric oxidant [*Logan et al.*, 1981], as an indirect greenhouse gas [*Forster et al.*, 2007], as a tracer of long-range transport of pollution [*Staudt et al.*, 2001], and as a correlative constraint for inverse analyses of CO_2 surface fluxes [*Palmer et al.*, 2006]. Understanding CO sources also places constraints on emissions of other pollutants released during combustion and whose emissions are often referenced to CO [*Andreae and Merlet*, 2001]. CO has strong absorption lines in the thermal infrared spectral region and the first overtone has reasonably strong absorption features in the solar shortwave infrared, making it readily observable from space. A number of satellite instruments have been measuring tropospheric CO globally over the past decade including MOPITT (2000-) [*Edwards et al.*, 2006b; *Emmons et al.*, 2007; *Emmons et al.*, 2009], SCIAMACHY (2002-) [*Bovensmann et al.*, 1999; *Buchwitz et al.*, 2007; *Burrows et al.*, 1995; *de Laat et al.*, 2007], AIRS (2002-) [*McMillan et al.*, 2005; *McMillan et al.*, 2008a; *Warner*, 2007; *Yurganov et al.*, 2008], ACE-FTS (2003-) [*Clerbaux et al.*, 2005; *Clerbaux et al.*, 2008], TES (2004-) [*Lopez et al.*, 2008; *Luo et al.*, 2007a; *Rinsland et al.*, 2006], and IASI (2007-) [*Fortems-Cheiney et al.*, 2009; *Turquety et al.*, 2009]. These satellite data expand the perspective offered by in situ observations, such as the NOAA/GMD surface monitoring network [*Novelli et al.*, 2003] and those from aircraft [*Nedelec et al.*, 2003].

Our objective here is to combine information from four different satellite sensors (MOPITT, SCIAMACHY, AIRS, TES) to provide global high-resolution constraints on CO sources using an adjoint inverse modeling method. The four instruments all observe in the nadir from sun synchronous polar orbits. MOPITT, AIRS, and TES observe thermal emission in the 4.7 μm absorption band and thus are most sensitive to the mid-troposphere. SCIAMACHY observes backscattered solar radiation upwelling from the top of the atmosphere in the 2.3 μm absorption band and is thus sensitive to the full depth of the atmosphere. AIRS and TES are on the same orbit (A-train) with equator crossing time within 8 minutes of 01:30. MOPITT and SCIAMACHY are on different orbits with equator crossing times of 10:30 and 10:00.

A central component of our work is to assess the consistency and complementarity of the data from the different satellite instruments. This is challenging because of the differences in sensitivity of the measurement and in retrieval techniques and the differences in the observed atmospheric scenes. Some limited intercomparisons between satellite pairs have been reported in the literature [*Buchwitz et al.*, 2007; *Luo et al.*, 2007b; *Turquety et al.*, 2008; *Warner*, 2007; *Yurganov et al.*, 2008]. Near simultaneous aircraft vertical profiles provide accurate validation but are sparse. A more general approach that we exploit here is to use a chemical transport model (CTM) as an intercomparison platform. The CTM provides a global, continuous, and consistent 3-D representation of CO concentrations, albeit with some error. Comparison of the observed and modeled CO concentrations sampled for the different orbits, overpass times, and retrievals of the individual instruments are used to examine the consistency of the

observations relative to the model. This is particularly useful in an inverse modeling framework where, as here, the CTM serves as the forward model for the inversion.

Despite long-standing interest in atmospheric CO and the abundance of data, our understanding of the CO budget remains inadequate, as illustrated by a recent CTM comparison exercise showing significant disagreements between models and observations [*Shindell et al.*, 2006]. Simulation of the spatial, seasonal, and interannual variability of CO involves a complex interplay of sources, transport, and chemistry [*Duncan et al.*, 2008]. Errors in sources can exceed a factor of two on continental scales [*Bian et al.*, 2007; *Hudman et al.*, 2008]. A number of inverse modeling studies have used MOPITT satellite data as constraints on CO sources [*Arellano et al.*, 2004; *Arellano et al.*, 2006; *Heald et al.*, 2004; *Pétron et al.*, 2004; *Pfister et al.*, 2005], including several by the adjoint method [*Chevallier et al.*, 2009; *Kopacz et al.*, 2009; *Stavrakou and Müller*, 2006; *Yumimoto and Uno*, 2006]. The study by *Fortems-Cheiney et al.* [2009] combined MOPITT and IASI data. Results of these and other inverse studies using surface CO measurements as constraints [e.g. *Kasibhatla et al.*, 2002; *Pétron et al.*, 2002] are often not quantitatively consistent, which could reflect insufficient constraints from observations, errors from model transport, and unrecognized errors in the inverse modeling approach. The adjoint method is particularly efficient at extracting the information content from observations by retrieving sources at the resolution of the underlying CTM, thus overcoming large-region aggregation errors in the more standard analytical method [*Kopacz et al.*, 2009]. Exploitation of multi-sensor satellite data in a global inversion by the adjoint method holds the potential for significant advance over previous studies and we follow that approach here.

We use a full year (May 2004-May 2005) of satellite data from MOPITT, SCIAMACHY, AIRS, and TES. This time period corresponds to the best overlap of data from these instruments. The Short Wave Infrared channels of SCIAMACHY were experimental and the first of their kind to fly in space. The 2.3 μm channel suffered most from the growth of the ice layer in 2003 and later also from an increasing number of bad and dead detector pixels (2005-) arising from radiation damage [*Buchwitz et al.*, 2007], making 2004 the year with best quality of SCIAMACHY data, while the TES record begins in October 2004. We use the GEOS-Chem CTM as the forward model for the inversion and apply its adjoint [*Henze et al.*, 2007; *Kopacz et al.*, 2009] to optimize the CO sources on a 4^{o}x5^{o} horizontal grid with monthly temporal resolution. We begin by describing the satellite datasets (section 2) and the GEOS-Chem CTM (section 3). In section 4 we intercompare the data from the different satellite instruments using GEOS-Chem as the intercomparison platform. The inverse analysis is described in section 5 and results are presented in section 6. Testing of the optimized sources with independent datasets including in situ data from the surface (NOAA/GMD network) and aircraft (MOZAIC) is presented in section 7.

2. Satellite data

2.1 MOPITT

The Measurements Of Pollution In The Troposphere (MOPITT) instrument was launched aboard EOS Terra in December 1999. The equator crossing time is 10:30/22:30 local with global coverage every 3 days. MOPITT measures thermal emission in the 4.7 μm absorption band, which results in highest vertical sensitivity in the mid-troposphere but also provides some boundary layer information [*Deeter et al.*, 2003; *Deeter et al.*,

2007; *Kar et al.*, 2008]. The sensitivity of the retrieval (to the true profile) is defined by its averaging kernel matrix **A**:

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

where $\hat{\mathbf{z}}$ is the retrieved vertical profile vector consisting of mixing ratios on a fixed pressure grid [*Deeter et al.*, 2003], $\mathbf{z}$ is the true profile on the same grid, and $\mathbf{z}_a$ is a globally uniform a priori profile derived from an ensemble of observations [*Deeter et al.*, 2003]. Only cloud-free scenes are retrieved. The Degrees Of Freedom (DOF) for signal, representing the number of pieces of information in the vertical profile and estimated as the trace of the averaging kernel matrix, are typically about 1.5 [*Deeter et al.*, 2004]. Therefore we only use the altitude-weighted CO column $\hat{y}$ obtained by summing the vertical profile $\hat{\mathbf{z}}$ with the corresponding pressure weights. MOPITT version 3 data for $\hat{y}$ and **A** are collected from ftp://l4ftl01.larc.nasa.gov/MOPITT/MOP02.003/. MOPITT daytime observations have been validated against aircraft data from several campaigns (mostly in the northern hemisphere), indicating a positive bias of about 5±11% on the column, with an uncertain increasing trend [*Emmons et al.*, 2004; *Emmons et al.*, 2007; *Emmons et al.*, 2009; *Jacob et al.*, 2003]. Nighttime observations have not been validated and appear subject to larger bias [*Heald et al.*, 2004]. We use the daytime data only.

2.2 AIRS

The Atmospheric Infrared Sounder (AIRS) instrument was launched aboard EOS Aqua in May 2002. The equator crossing time is 01:30/13:30 local with daily global coverage due to a 1650 km cross-track scanning swath. AIRS measures thermal emission in the 4.7 μm absorption band, as does MOPITT [*McMillan et al.*, 2005; *Warner*, 2007].

However, unlike MOPITT and other instruments in this comparison, AIRS possesses a cloud clearing capability [*Susskind et al.*, 2003; *Susskind et al.*, 2009] that enables it to retrieve partly cloudy scenes and thus achieve 70% effective daily coverage. Profile retrieval of partial columns $\hat{\mathbf{z}}$ is described by the following equation [*Olsen*, 2007]:

$$\ln \hat{\mathbf{z}} = \ln \mathbf{z}_a + \mathbf{FAF}'(\ln \mathbf{z} - \ln \mathbf{z}_a) \qquad (2)$$

where $\mathbf{z}$ is a vertical profile of partial columns on the 100 levels of the radiative transfer model, **F** is a matrix that defines the nine trapezoidal layers on which AIRS CO is retrieved, **F'** is its pseudo inverse, **A** is a 9x9 averaging kernel matrix in the trapezoidal space, and $\mathbf{z}_a$ is an a priori profile of partial columns, which is the same as for MOPITT for the common levels and AFGL standard atmosphere above that. AIRS retrievals have DOF for signal on average about 0.8, with higher values over land than ocean and typically higher in daytime than at night. We use the columns $\hat{y}$ obtained by summing the vertical profiles $\hat{\mathbf{z}}$ of partial columns. AIRS version 5 data for $\hat{y}$, **F** and **A** are collected from ftp://airspar1u.ecs.nasa.govdata/s4pa/Aqua_AIRS_Level2/AIRX2SUP.005/. The version 5 data are expected to represent significant improvement over the previously documented version 4 [*McMillan et al.*, 2008a; *Warner*, 2007; *Yurganov et al.*, 2008], with validation ongoing [*McMillan et al.*, 2008b]. For consistency, we use daytime CO column data only as for MOPITT. For best quality, we subsample for retrievals with surface temperature greater than 250K.

2.3 TES

The Tropospheric Emission Spectrometer (TES) instrument was launched aboard EOS Aura in July 2004 (observations available starting October 2004). The overpass time lags 8 minutes behind AIRS. TES measures thermal emission at 4.7 μm, as do MOPITT and AIRS. It obtains global coverage every 16 days and has no cross-track scanning capability, yielding a much sparser dataset than MOPITT or AIRS [*Rinsland et al.*, 2006]. The retrieval provides vertical profiles $\hat{\mathbf{z}}$ of logarithms of mixing ratios:

$$\ln \hat{\mathbf{z}} = \ln \mathbf{z_a} + \mathbf{A}(\ln \mathbf{z} - \ln \mathbf{z_a}) \qquad (3)$$

Unlike AIRS and MOPITT, the TES a priori profiles $\mathbf{z_a}$ vary by region and season [*Osterman et al.*, 2007]. As for MOPITT and AIRS, we only use daytime column data $\hat{y}$ computed from the vertical profile $\hat{\mathbf{z}}$. TES V002 data for $\hat{y}$ and **A** were collected from http://eosweb.larc.nasa.gov/PRODOCS/tes/table_tes.html. Limited validation of these data with aircraft show no consistent bias [*Lopez et al.*, 2008; *Luo et al.*, 2007a] The quality of the TES CO data improved greatly (four-fold increase in signal-to-noise ratio) following a warm-up of the optical bench in early December 2005 [*Rinsland et al.*, 2006]. Therefore we consider here not only the period October 2004 - April 2005 overlapping with the other satellite datasets, but also the period May 2005 - April 2006 (with available data starting in July 2005), which includes data after the December 2005 bench warm-up.

2.4 SCIAMACHY

The SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument was launched aboard ENVISAT in March 2002 with an equator crossing time of 10:00 local [*Bovensmann et al.*, 1999; *Burrows et al.*, 1995].

SCIAMACHY measures solar backscattered radiation at 2.3 μm, which allows for nearly uniform sensitivity through the tropospheric column though with no vertical resolution [*Buchwitz et al.*, 2004; *Buchwitz et al.*, 2005; *de Laat et al.*, 2006]. Global coverage is obtained by SCIAMACHY for its nadir measurements in 6 days at the equator. However, the selected data, achieving the necessary fit goodness criteria, which depends on the signal to noise ratio, are significantly reduced because of the low reflectivity of ocean and the presence of clouds in the relatively large SCIAMACHY ground scene (30 km x 120 km) [*Buchwitz et al.*, 2007; *Gloudemans et al.*, 2005; *Gloudemans et al.*, 2008]. We consider version 0.6 retrieval from the University of Bremen [*Buchwitz et al.*, 2004; *Buchwitz et al.*, 2005; *Buchwitz et al.*, 2007]. The retrieval provides CO columns, $\hat{y}$, with vector averaging kernels **a**, related to the true vertical profile (**z**) by the following equation:

$$\hat{y} = \mathbf{z}_a + \mathbf{a}(\mathbf{z} - \mathbf{z}_a) \qquad (4)$$

where both **a** and the fixed a priori profile $\mathbf{z}_a$. Ocean data are indirectly discarded due to low signal to noise ratio over dark surfaces. *Buchwitz et al.* [2007] found that the Bremen retrieval was on average 10% higher than MOPITT CO columns with 20% standard deviation.

SCIAMACHY data have considerable noise, i.e. 10-100% of the total column, and the effective useful resolution is monthly on a 3º x 2º grid [*de Laat et al.*, 2007]. Here we use daily averaged data weighted by the reported instrument error and use available quality flags for data screening. We select data with the quality flag, which is part of the data product, and which to some extent corrects for cloud effects using simultaneously retrieved methane. We further sample using the cloud-free flag (as also done by

205 *Tangborn et al.* [2009]), which corresponds to a cloud fraction of no more than 0.1. *Buchwitz et al.* [2007] used data with maximum cloud fraction of 0.3 in their comparison with MOPITT. The cloud-free screening significantly reduces the number of measurements, especially over the oceans [*Khlystova et al.*, 2009].

210 **3. CO simulation in the GEOS-Chem CTM**

GEOS-Chem is a global 3-D chemical transport model (CTM) driven by GEOS assimilated meteorological data from the NASA Global Modeling and Assimilation Office (GMAO) (http://www.as.harvard.edu/chemistry/trop/geos). The GEOS-Chem CO simulation was originally described by *Bey et al.* [2001] and more recently by *Duncan et*

215 *al.* [2007]. Here we use version 7-04-11 for the period spanning May 1, 2004 through April 30, 2005. We use GEOS-4 meteorological data with 1ºx1.25º horizontal resolution and degrade the resolution in GEOS-Chem to 2°x2.5° for the satellite data intercomparison and to 4°x5° for the inverse model analysis. Combustion sources of CO include fossil fuel, biofuel, and biomass burning emissions, augmented following *Duncan*

220 *et al.* [2007] by 19%, 19%, and 11% respectively to account for co-emitted nonmethane VOCs (NMVOCs). Additional CO sources include oxidation of methane, which produces CO in the atmosphere with an instantaneous yield of unity, and NMVOCs, which produce CO at the point of emission with a yield of 0.09-1.00 [*Duncan et al.*, 2007]. We compute CO loss and production from methane by using monthly mean 3-D OH

225 concentration fields archived from a detailed oxidant-aerosol GEOS-Chem simulation (version 5-07-08) [*Park et al.*, 2004]. Our global mean tropospheric OH concentration is 10.8 x 10^5 molec/cm^3, which compares well with the multimodel mean of 11.1+/-1.7 x

10^5 molec/cm^3 reported by *Shindell et al.* [2006]. Our corresponding tropospheric lifetime of methyl chloroform is 5.3 years, somewhat shorter than those reported by *Prinn et al.*[2005] and *Spivakovsky et al.* [2000], 6.0 (+0.5,-0.4) years and 5.7 years, respectively. We initialize our simulation with CO concentrations derived from a year-long GEOS-Chem spin-up simulation and subsequent rescaling to MOPITT CO columns (corrected for the 5% high bias), as done previously in *Kopacz et al.* [2009].

Previous versions of the GEOS-Chem CO simulation have been evaluated against observations from surface sites [*Duncan et al.*, 2007; *Goldstein et al.*, 2004; *Liang et al.*, 2004; *Weiss-Penzias et al.*, 2004], aircraft [*Heald et al.*, 2003; *Hudman et al.*, 2008; *Zhang et al.*, 2008], and satellites, including MOPITT and TES [*Arellano et al.*, 2006; *Heald et al.*, 2004; *Kopacz et al.*, 2009; *Zhang et al.*, 2006]. The comprehensive model evaluation by *Duncan et al.* [2007] showed biases relative to the NOAA/GMD [*Novelli et al.*, 2003] surface network data in the range +/- 10% in the northern hemisphere and up to -19% in the southern tropics.

Duncan et al. [2007] estimated a direct global emission of CO (excluding co-emitted NMVOCs) of 956-1086 Tg a^{-1} for 1988-1997, a period of downward emission trends in Europe and the U.S., but upward trend in Asia. They assigned an error of less than 25% on this global estimate. Their mean tropospheric OH concentration simulated for that period is 8.7-9.3 x 10^5 molec/cm^3, in agreement with CH_3CCl_3 lifetime [*Prather*, 2001]. The models in the *Shindell et al.* [2006] comparison included higher OH concentrations, and found a consistent underestimate of CO concentrations across models (including GEOS-Chem) of up to 40-60 ppb in spring at northern midlatitudes and in excess of 60 ppb over south-central Africa during the biomass burning season.

Figure 1, Figure 2 and Table 1 show seasonal and annual emissions in our current GEOS-Chem simulation for May 2004-April 2005, taken as a priori for our source inversion. CO emissions from combustion amount to 858 Tg a^{-1}, with an additional 140 Tg a^{-1} from oxidation of co-emitted VOCs. They are drawn from EDGAR 3.2FT2000 inventory [*Olivier et al.*, 1999; *Olivier and Berdowski*, 2001] for the year 2000, implemented in GEOS-Chem by *van Donkelaar et al.* [2008]. These were overwritten with the following regional inventories: the US Environmental Protection Agency National Emission Inventory for 1999 (EPA-NEI99) for the US with a 60% downward correction following *Hudman et al.* [2008] (NEI99_Hudman), the Big Bend Regional Aerosol and Visibility Observational (BRAVO) Study Emissions Inventory for Mexico [*Kuhns et al.*, 2003], the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) inventory for Europe in 2000 [*Vestreng and Klein*, 2002], as well as *Streets et al.* [2006] and *Streets et al.* [2003] anthropogenic emissions for Asia in 2000 and China in 2001. Biomass burning emissions are from the interannual GFED2 inventory with monthly resolution [*van der Werf et al.*, 2006]. The combustion emissions differ from those used by *Duncan et al.* [2007]. In the high summer fire season in the North American boreal region, we do not assume any emission injection above the boundary layer. This could cause an underestimate of vertical transport and thus an overestimate of surface emissions in the inversion. A recent analysis of the heights of plumes from these fires shows that at least 10% of plumes were injected above the boundary layer at local time of 11:00-13:00 [*Kahn et al.*, 2008; *Val Martin et al.*, 2009]. Additional CO sources come from oxidation of methane (853 Tg) and biogenic NMVOCs (426 Tg), which include isoprene, monoterpene, methanol and

acetone as described by in previous studies[*Arellano et al.*, 2006; *Heald et al.*, 2004; *Kopacz et al.*, 2009].

Figure 3 compares our CO simulation with a priori sources (in red) with monthly mean CO concentrations (climatological 1988-2001 in black, 2004-2005 in blue) from the same NOAA/GMD surface sites as in *Duncan et al.* [2007]. A posteriori model shown in green will be discussed in section 7. Our a priori comparison support conclusions from *Shindell et al.* [2006] and *Duncan et al.* [2007], including the model winter-spring underestimate in the extratropical northern hemisphere. One notable difference with *Duncan et al.* [2007] is our underestimate of Bermuda in winter-spring (2004-2005 not shown due to scarcity of data), reflecting our decrease of U.S. CO emissions following *Hudman et al.* [2008]. This will be discussed further in the context of the inverse model results. Our simulation in the southern tropics (Samoa) improves on *Duncan et al.* [2007], who found a larger underestimate for the seasonal maximum; this could reflect our use of GFED2 biomass burning emissions or different meteorological fields (GEOS-1) and year of data (1994), and again will be discussed further in the context of the inverse model results.

Figure 4 compares our a priori model results at 710, 480, and 305 hPa with 2002-2007 monthly mean MOZAIC aircraft observations over selected locations. The winter-spring model underestimate in the northern hemisphere is apparent at all altitudes, consistent with the data from surface sites, although it dampens with altitude. Here also, a posteriori model is shown in green and will be discussed in section 7.

4 Intercomparison of satellite datasets

Figure 5 shows annual mean (May 2004 - April 2005) CO columns from MOPITT, AIRS, SCIAMACHY Bremen. For TES, the mean is computed for October 2004 – April 2005. There are obvious differences, which could reflect differences in instrument/retrieval properties (as described by the averaging kernels and a priori), sampling, and actual biases. To separate these effects we use as an intercomparison platform the GEOS-Chem CTM, which provides a continuous 3-D concentration field and we take into account differences between instruments in sensitivities, a priori profiles, and sampling. This is done by applying retrieval equations (1)-(4) to the model vertical profiles for each observation scene.

Figure 6 shows scatterplots of satellite versus model CO columns for May 2004 – April 2005 (except for TES, where we show May 2005 – April 2006 observations and model). Individual points represent daily observations averaged over the 2^{o}x2.5^{o} grid of the model. We report the resulting correlation coefficient (r) and slope of the reduced-major axis (RMA) regression line, which allows for error in both datasets, as well as the mean model-observed percentage difference. Also included in Figure 6 is the model correlation with in situ measurements from the GMD and MOZAIC datasets (from Figure 3 and Figure 4) , which provides an absolute reference. It shows r = 0.84 with a slope of 0.75. The relative difference of annual mean model versus annual mean data is -12%, indicating a mean model underestimate as discussed previously.

Differences between the model and satellite observations in Figure 6 reflect model, retrieval, and instrument errors. The smoothing error described by the averaging kernel is applied to both the observations and the model and thus is not a cause of the

differences. In fact, variability of this smoothing error from scene to scene could lead to the appearance of strong correlation in cases where the DOF are low [*Luo et al.*, 2007b; *Rodgers*, 2000].

Figure 6 shows strong consistency between MOPITT and AIRS, based on their correlations with the model. The correlations reflect actual information from the instruments, as opposed to variability in the a priori, since the a priori is globally uniform and identical for both retrievals. We investigated whether the correlation could be driven in part by varying contributions from the a priori to the retrieval, as measured by the DOF for signal. DOF shown in Figure 6 for MOPITT and AIRS are about 0.5 in the polar regions but much larger than 0.5 otherwise, indicating that most of the information comes from the measurement as opposed to the a priori; average values are 1.1 for MOPITT (1.4 in extra-polar regions), and 0.78 for AIRS (0.81 in extra-polar regions). There is no indication from Figure 6 that variability in DOF contributes to the correlation of the observations with the model. Cloud screening is a likely reason for the higher DOF for MOPITT than AIRS. Further examination of MOPITT-AIRS comparisons with GEOS-Chem for individual hemispheres, land versus ocean, and individual seasons indicate statistics similar to the global values in Figure 6. MOPITT shows a stronger winter-spring maximum than AIRS as well as a larger interhemispheric difference (Figure 5). We elaborate further on regional differences between MOPITT and AIRS in the context of the inversion results in section 6.

TES shows stronger correlation and less difference with GEOS-Chem compared to MOPITT or AIRS (Figure 6). For the 2005-2006 data shown in Figure 6 (mean DOF of 0.99), the correlation coefficient is 0.91 and the regression slope is 0.88. We find

similar statistics for the 2004-2005 data (not shown) with mean DOF of 0.74. The high correlation reflects the variable a priori used by TES. To test the effect of the smoothing, we reprocess TES retrieved columns and their corresponding GEOS-Chem columns using the same a priori profile as used by MOPITT and AIRS. We find that the TES versus GEOS-Chem correlation coefficient drops to 0.81 and the slope drops to 0.74, yielding statistics similar to MOPITT and AIRS vs. GEOS-Chem. Although the model-TES correlation is very close to that of MOPITT and AIRS when TES and corresponding model are reprocessed with MOPITT a priori, the absolute values of the reprocessed TES CO columns and the corresponding model columns are much lower. Also the global annual mean model-data difference is -5% for TES vs. -16% for MOPITT and -13% for AIRS. In terms of a relative comparison with the model, the TES data appear consistent with the MOPITT and AIRS data once the effect of the variable a priori is removed, although in absolute values there appears to be an offset.

SCIAMACHY daily CO data have considerable noise and most assessments of these data have been on a monthly average basis to reduce the noise error [*Buchwitz et al.*, 2007; *de Laat et al.*, 2007]. Figure 6 shows slope and correlation coefficient relative to GEOS-Chem, which are 0.56 and 0.44, while mean model-data difference is -20%. The SCIAMACHY Bremen retrieval reproduces the northern hemispheric seasonal variation, clearly present in the thermal infrared instrument measurements and the model, and has similar model-data differences.

5. The inverse model

Our inverse problem consists of optimizing the sources of CO by minimizing the mismatch between simulated (GEOS-Chem) and observed CO columns, accounting for

constraints from a priori knowledge. Let the vector $\mathbf{y_o}$ represent the ensemble of CO column observations used in the inversion (as described in section 2), $\mathbf{y_m}$ the corresponding model values, $\mathbf{x}$ the ensemble of CO sources to be optimized (state vector), and $\mathbf{x_a}$ the a priori estimate (described in section 3 and shown in Figure 1). Bayesian optimization assuming Gaussian errors involves minimization of the least-squares scalar cost function $J(\mathbf{x})$:

$$J(\mathbf{x}) = (\mathbf{y_m} - \mathbf{y_o})^{\mathrm{T}} \mathbf{S}_{\Sigma}^{-1} (\mathbf{y_m} - \mathbf{y_o}) + (\mathbf{x} - \mathbf{x_a})^{\mathrm{T}} \mathbf{S_a}^{-1} (\mathbf{x} - \mathbf{x_a}) \quad (5)$$

where $\mathbf{S}_{\Sigma}$ and $\mathbf{S_a}$ are the observational and a priori error covariance matrices described below.

We use the GEOS-Chem model adjoint to solve the minimization problem $\nabla_{\mathbf{x}} J = \mathbf{0}$ numerically, as described previously by *Kopacz et al.* [2009] in an inverse analysis of CO sources in East Asia in spring 2001 using MOPITT data. The GEOS-Chem adjoint was originally developed by *Henze et al.* [2007]. We extend it here to include the adjoint of GEOS-4 convective transport, derived using the Tangent Linear and Adjoint Model Compiler (TAMC) software, and advective transport, using negative winds. We also include the satellite observation operators and their adjoints. The observation operators compute corresponding columns for the observation scene and apply retrieval equations (1)-(4).

We include in the inversion the observations for May 2004 – April 2005 from MOPITT, AIRS, and SCIAMACHY Bremen, averaged over the 4º x 5º resolution of GEOS-Chem used for the inversion. We exclude MOPITT and SCIAMACHY Bremen data in polar regions (> 60^{o} latitude), where they are of lower quality. Aircraft validation data (section 2) show a 5% MOPITT positive bias and for the purposes of the source

inversion, we correct for it. AIRS validation indicates a positive bias of 6-10% between 300 and 900 hPa in the northern hemisphere [*McMillan et al.*, 2008b]. Translating the MOPITT 5% bias via the model-data correlations in section 4, we derive a rough estimate of the AIRS bias. The northern hemispheric monthly bias ranges from a low bias of 2-10% in spring-summer to a high bias of 5-8% winter. Thus, based on the MOPITT bias and the model-data correlations, the need for bias correction in the AIRS data is unclear and we do not correct for it. Similarly, no correction for bias in SCIAMACHY data appears necessary. We thus have 305,484 observations from MOPITT, 923,234 observations from AIRS, and 25,773 observations from SCIAMACHY Bremen. Due to data scarcity, we do not include TES data in the source inversion, but instead use them as an independent set of observations to evaluate our a posteriori results.

We optimize the CO combustion sources at the 4º x 5º grid resolution of the GEOS-Chem model and monthly temporal resolution, over the whole year from May 1, 2004 to April 30, 2005. Optimization is only for grid squares with non-zero combustion sources in the a priori (Figure 1). We also optimize the global CO source from oxidation of methane and biogenic NMVOCs as a single variable with monthly temporal resolution. Our state vector **x** thus has 18420 elements.

The observational error covariance matrix $\mathbf{S}_{\Sigma}$ includes contributions from the measurement error, GEOS-Chem model error, and representation error. We estimate the latter with the Relative Residual Error (RRE) method [*Heald et al.*, 2004; *Kopacz et al.*, 2009; *Palmer et al.*, 2003]. This method attributes the mean of model-observation differences for a given grid square and season (month in the case of AIRS) to an error in CO sources, and the residual to observational error. We thus find that the highest

observational errors are for SCIAMACHY (up to 70-100% in high northern latitudes). MOPITT observational errors are in the 10-30% range, highest over pollution outflow regions. AIRS errors are similar to MOPITT but lower (as low as 5% in remote ocean regions), reflecting the lower DOF. Error correlations between observations can be neglected at the 4° x 5° resolution used for the inversion [*Heald et al.*, 2004], so that $\mathbf{S_\Sigma}$ is diagonal.

The a priori error covariance matrix $\mathbf{S_a}$ includes a uniform error of 50% for combustion sources and 25% for the global oxidation source, the latter as used in previous studies [*Heald et al.*, 2004; *Kopacz et al.*, 2009]. The monthly errors are assumed uncorrelated so that $\mathbf{S_a}$ is diagonal. The a priori terms in Equation (5) do not contribute substantially to minimization of the cost function. This, however, does not imply that the a posteriori sources are constrained entirely by the data and are independent of the choice of a priori sources.

6. Optimized monthly CO sources

6.1 General results

Figure 7 shows the global annual mean correction factors to the a priori emission estimates and Table 1 gives the annual total emissions for the largest source regions. The emission correction factors are ratios of a posteriori to a priori emissions. Emissions increase almost everywhere relative to the a priori. The global CO source from oxidation of methane and biogenic NMVOCs was derived as a global monthly estimate and its a posteriori change from a priori value of 1280 Tg was < 1%. Most of this source is from the oxidation of methane (Table 1). Our a posteriori annual global estimate for direct CO emissions is 1350 Tg a^{-1} (+217 Tg a^{-1} from oxidation of co-emitted VOCs), a 60%

increase from a priori. This is within 25% of results from previous (global and annual) inversions of satellite (MOPITT) measurements: 1091 Tg a^{-1} using the MOZART model [*Pétron et al.*, 2004], 1342-1502 Tg a^{-1} using GEOS-Chem [*Arellano et al.*, 2004; *Arellano et al.*, 2006] and 1695 Tg a^{-1} using the IMAGES model [*Stavrakou and Müller*, 2006]. We also compare our results to regional and seasonal studies as we describe details of our results in section 6.2.

The annual emission corrections in Figure 7 show an overall underestimate in large source regions of southern Africa, SE Asia (southeast of Bangladesh), equatorial Africa, China, S. America, southern Africa, India, northern Australia and Europe, in order of decreasing emission corrections ranging from 100% to 20%, with large seasonal variations. Figure 2 shows the seasonal correction to sources in northern midlatitudes, with the largest absolute correction in E. Asia. The cold months upward correction could be due to underestimated residential heating and transport ("cold starts" [*Parrish*, 2006]). We also see large positive corrections to biomass burning sources, especially in southern African and S. American. This indicates an overall emission underestimate by the GFED2 inventory, also reported in several past studies [*Chevallier et al.*, 2009; *Tanimoto et al.*, 2009; *Turquety et al.*, 2009]. The regional details (Figure 8) and comparison with previous findings are described in section 6.2. Since we find a substantial correction to the seasonality of sources, which varies with region, we discuss these results in more detail below.

Comparing a priori and a posteriori model bias with respect to each dataset, we find regional inconsistencies among the instruments. Figure 9 and section 6.3 compare model bias from the inversion using three datasets versus inversions performed with

individual datasets. As mentioned above and confirmed in the results, the datasets are overall consistent, but we find inconsistencies over the tropical biomass burning regions.

In addition to data inconsistencies, our estimates are subject to uncertainty in OH concentrations, particularly in northern extratropics, where OH concentrations are not well constrained by methyl chloroform lifetime, as well as unknown errors in meteorological data and errors in VOC concentrations. Lower OH concentrations would lower our emission estimates, while errors in meteorology and VOC concentrations could have a varied effect, the latter especially affecting CO seasonal cycle in some regions [*Arellano and Hess*, 2006].

6.2 Seasonal and regional results

A striking result of the inversion is the seasonal variation of the source correction at northern mid-latitudes. Figure 8a shows this seasonal variation for North America. We find no need for correction over the US in summer (of the NEI99_Hudman inventory), supporting the previous 60% downward correction to the NEI99 emission inventory derived by *Hudman et al.* [2008]. This correction was based on ICARTT summer aircraft measurements and we used it year round as a priori. In an independent analysis using aircraft and tower data, by *Miller et al.* [2008] found the NEI99 emissions to be too high by a factor of three in summer and two in spring. They suggest that spring emissions are higher because of source from domestic wood burning and less efficient combustion for mobile sources. We find that emissions are higher in seasons other than summer, in a way that is not properly represented by the seasonal variation in NEI99 inventory (Figure 2). A posteriori US emissions in winter (DJF) are on average 50% higher than in summer, while spring (MAM) and fall (SON) are 25% higher with the largest effects (exceeding a

factor of two) in the Northeast and Midwest. The spring estimate in *Miller et al.* [2008] may be larger than ours because they focused their analysis on the Midwest and Northeast. *Parrish* [2006] in his evaluation of NEI99 emission estimates against fuel-based inventory and surface measurements, suggests that while US on-road emissions are overestimated, as corrected by *Hudman et al.* [2008], a lot of uncertainties remain and could include, among others, "cold starts" during the cold months. Our analysis implies that the errors in original NEI99 inventory are larger in summer than in winter, and the cause of the dramatic emission overestimate in summer (60%) remains unclear.

The spring underestimate in the Yucatan Peninsula occurs during a period of large biomass burning in the region. Inverse model results for the boreal forest fire regions of Alaska and western Canada in summer 2004 indicate a 30% underestimate in the GFED2 biomass burning inventory, corresponding to an a posteriori emission estimate of 24 Tg. A previous inversion for that region and season by *Pfister et al.* [2005] using MOPITT data indicated an a posteriori estimate of 30Tg. A detailed bottom-up fire emission inventory for the region also found a total of 30 Tg [*Turquety et al.*, 2007].

Figure 8b shows a qualitatively similar picture for Europe. Summer a posteriori emissions are largely the same as a priori (the EMEP inventory), but we see relative underestimates in the fall, winter and spring, particularly in northern France, western Germany, the Benelux countries and northern Italy, where underestimates range from 30% to 70%. Winter emissions (January-February) are underestimated consistently throughout Europe by at least 30% in most gridboxes and up to 70% in northern Italy. Figure 2 demonstrates the seasonal correction is Europe is similar to, but not as strong as that in North America. Since the seasonal pattern of upward emission corrections

corresponds to urban areas and cold months, the likely underestimate possibly comes from residential heating and on-road vehicle emissions ("cold starts").

In Asia (Figure 8c), the inversion finds the *Streets et al.* [2006] inventory for China, with no seasonal variation for fossil fuel and biofuel, is underestimated in fall-winter-spring by 50-100%, with only a small underestimate in summer. The corresponding annual total is 267 Tg. Our previous estimate for China in 2001 for the same region [*Kopacz et al.*, 2009] was 142 Tg a^{-1} (much smaller in southern and western China), derived from MOPITT observations in March-April 2001 only. Figure 2 shows a strong seasonal correction in East Asia, which includes China, the Koreas and Japan as well as the northern half of India, and parts of several other countries. As with Europe and North America, we see a similar seasonality in the correction but with a much larger amplitude, here close to 100% in the winter (DJF), extending to March when it is partly due to biomass burning in SE Asia (included on the fringes of our domain). Since our most consistent underestimate corresponds to cold months and coastal regions with high population density, although not the only region of high emissions, we attribute part of its source to the transport sector. Chinese sources also include uncertain amounts from residential coal and biofuel heating [*Streets et al.*, 2006], which should be higher in the colder months, further contributing to the seasonal underestimate shown in Figure 2. Figure 8c also indicates a consistent underestimate of Indian emissions with little seasonal variation except for northern India in spring. The underestimate could be due to biomass burning, which in India is largely absent in the GFED2 inventory.

The large biomass burning areas in southeastern Asia, in particular Indonesia and Malaysia appear to be consistently underestimated by more than 100% with respect to

GFED2 inventory. Our previous work [*Kopacz et al.*, 2009] focused on spring 2001 using MOPITT data derived an annual source of 113 Tg in the SE Asia-Indonesia-Philippines region [*Kopacz et al.*, 2009]. Our current estimate for the region is 256 Tg. One reason for the difference is the ENSO cycle: 2001 was a La Niña year, while 2004 saw a weak El Niño with considerably more biomass burning [*Edwards et al.*, 2006a]. Also, the MOPITT v3 (unlike v4) retrieval algorithm is not applied to high signal values and thus high CO concentrations are not obtained, introducing a low bias over high emission regions like this one. Here both model-MOPITT and model-AIRS a priori differences are negative, indicating low model bias, but a posteriori model biases are positive (smaller with respect to AIRS), indicating overcompensation for the original bias. This overcompensation can be expected, given a large least-squares correction to a large source, and should be kept in mind when comparing a posteriori results to independent observations. *Fortems-Cheiney* [2009] applied MOPITT and IASI 700hPa CO concentrations individually to constrain global CO sources during July to November 2008 (outside the biomass burning season), using the adjoint of the LMDZ-INCA model. In their optimization, as in ours, they find in SE Asia that a priori model underestimate is overcompensated with a posteriori positive model bias. They obtain a 793 Tg global total (for 5 months) using IASI data and 566 Tg using MOPITT data. Our a posteriori estimate for the same months is comparable, 723 Tg, including 628 Tg of direct emissions and 95 Tg of co-emitted VOCs. The higher estimate derived from IASI CO measurements (also for the regional totals) could imply that MOPITT CO is too low in SE Asia (again, because of screening out high CO concentrations), which can be inferred from our a posteriori model-AIRS agreement as well (Figure 9).

In biomass burning dominated emission regions of Africa and S. America our a posteriori annual estimates are 343 Tg and 183 Tg (Table 1). The large biomass burning emissions in the Amazon and southern Africa are shown in Figure 8d to be largely underestimated in the GFED2 inventory, especially during the biomass burning season (August-October in S. America and July-October in southern Africa), with some underestimate seen as early as August and as late as March. Figure 9 shows that while the inversion improves model bias over eastern Brazil, it worsens it over the interior. The inversion points to the same difficulty in southern Africa, where model bias is reduced more over the source region than over the outflow. In the tropical biomass burning regions we find inconsistencies among the datasets as visible in the a posteriori model bias in Figure 9.

Chevallier et al. [2009] applied 2000-2006 MOPITT 700 hPa CO concentrations to an adjoint CO source inversion based on the LMDZ-INCA model [*Chevallier et al.*, 2005]. The aim of their study was to constrain African biomass burning emissions (bounded by 40S-40N, 25W-60E) over the years and seasons. Their a priori estimate was the same as ours, the GFED2 inventory. They also considered the GMD station Ascension, where their prior model bias of -5% was reduced to 0, as well as other stations with comparable a posteriori improvement. This contrasts with our a posteriori disagreement at that station. Their reported a priori and a posteriori comparisons of emissions and concentrations indicate the need for both increase and decrease of African emissions, depending on season and exact location. Their estimates for 2004 and 2005 are significantly lower, 255 Tg and 283 Tg, than our a posteriori value of 343 Tg. Our a posteriori model bias is positive with respect to MOPITT, confirming that MOPITT

retrieves lower concentrations (and hence emissions) over biomass burning source regions. In fact, the MOPITT version 3 algorithm specifically filters out spectra corresponding to very high CO values, as would be seen over the source regions, which could introduce a low bias.

6.3 Individual versus combined datasets

The largest improvement in model-data agreement with a posteriori sources is seen with respect to AIRS CO. This is not surprising, given the large number of AIRS measurements: three times as many as MOPITT and an order of magnitude more than SCIAMACHY. There is no objective way to weigh each dataset differently, other than through proper characterization of observational error.

To estimate the contributions of each dataset to the inversion and the value of combining them we performed individual dataset source inversions for the 3-month period of September-November 2004. This also tests the consistency among the inverse results and, by extension, the consistency of the datasets themselves. Figure 9 shows model a priori bias, model a posteriori bias from the three satellite inversion and model a posteriori bias derived from source inversion using individual datasets, all with respect to AIRS, SCIAMACHY and MOPITT data.

Globally, the model bias change from a priori to a posteriori in a joint inversion is as follows: an order of magnitude decrease with respect to AIRS data (-6% to -0.3%), an increase with respect to MOPITT (-4% to +10%), implying regional inconsistencies, and a reduction with respect to SCIAMACHY CO (-10% to +2%). In contrast, individual dataset inversions yield a larger a posteriori bias for AIRS (-0.7%), a smaller a posteriori bias with respect to MOPITT (+1%), and almost no change in bias with respect to

SCIAMACHY (a posteriori still -10%) due to the small amount of SCIAMACHY data along with a high observational error during that period. In summary, it is overall beneficial to combine the data to improve the model bias, but based on the model bias amounts, MOPITT column concentrations (with the correction for the 5% high bias) appear lower than AIRS or SCIAMACHY, especially in the southern hemisphere. The largest contribution to the cost function (78%) and largest difference comes from the model-AIRS discrepancy, which is much lower in the individual inversion. Figure 9 shows that the three satellite inversion best improves the model-AIRS disagreement, further suggesting that AIRS data tends to dominate the overall source estimates. Unless AIRS observational errors were much larger than those of MOPITT and SCIAMACHY, we expect AIRS CO to dominate the a posteriori source corrections, given the relatively large number of AIRS data and potential inconsistencies. If the datasets were perfectly consistent, improvement in model-AIRS agreement would perfectly map onto model-MOPITT and model-SCIAMACHY agreement (as seen over NH Pacific and Middle East). In fact, AIRS observational errors are lower than those of MOPITT and SCIAMACHY, but that reflects lower AIRS DOF and should not affect the information balance. Since the difference between model and observation for low DOF is also small, it prevents large contributions to the inversion from low signal data.

As much as the bulk calculations reveal overall consistency, regional discrepancies increase the model-data disagreement. Figure 9 shows an a posteriori model overestimate (joint inversion) with respect to MOPITT and SCIAMACHY throughout the southern hemisphere, which implies that AIRS is higher than MOPITT and SCIAMACHY (at least during September-November 2004), and that the difference

cannot be fully explained by lower AIRS DOF. Areas where the joint inversion did not improve model-data agreement are also not well constrained by individual dataset inversions (e.g. in S. America). Figure 9 also shows that using individual datasets to constrain CO sources can yield different results than combining the data. However, as each dataset has been thoroughly evaluated by its retrieval team, they should be combined together for a balance of information.

It follows then that the emission correction factors from the individual dataset inversions corresponding to model bias shown in Figure 9 are generally, but not entirely consistent. The three satellite inversion correction patterns are common in each of the individual dataset inversions. For September - November 2004, all datasets find a large (~100%) underestimate of southern African biomass burning and a similar pattern of underestimate and overestimate of biomass burning in the Amazon, but of different magnitudes. The only consistent difference is that MOPITT and SCIAMACHY corrections are more localized, while AIRS finds large areas to be underestimated. A few other regions also show opposite signs of corrections from different instruments, but generally, the differences are confined only to the magnitude of the correction.

7. Comparison with independent measurements

Figure 3 shows the a posteriori model CO compared against in situ observations from the GMD network. All stations in the northern hemisphere show considerable improvement in fitting the surface observations. The winter-spring underestimate is largely corrected. The phase and amplitude of the seasonal cycle in the model match the observations, supporting the seasonally varying corrections to the northern mid-latitude emissions and implying consistency between the satellite and surface data. The

inconsistency at Barrow in summer reflects the anomalous fire conditions in summer 2004, not reflected in the GMD data, which represent background conditions.

The a posteriori model comparison with MOZAIC aircraft observations in Figure 4 also shows large improvement at all extratropical locations and complete correction of the winter-spring underestimate at the different altitudes. The seasonal phase and amplitude are well reproduced.

No such improvement in fitting the surface observations is found for the southern hemisphere sites in Figure 3. The simulation with a posteriori sources fares generally worse than the a priori, although there is an improvement in the amplitude and phase of the seasonal cycle at all stations in the extratropics. This suggests an overestimate of the biomass burning source in the southern tropics constrained by the AIRS data, as suggested also in Figure 9 by the results for MOPITT.

Since we did not use TES CO data in the source inversion, we use it as an additional independent set of measurements to verify our a posteriori results. A global correlation against GEOS-Chem using a posteriori sources in the 2004-2005 period yields a correlation coefficient $r = 0.91$, same as the a priori, but the slope of the regression line increases from 0.89 to 1.04, indicating a better fit.

8. Conclusions

We applied the adjoint of the GEOS-Chem CTM to a global inversion of CO sources as constrained by three satellite datasets (MOPITT, AIRS, SCIAMACHY). The inversion used a full year of data (May 2004 to April 2005) and optimized CO combustion sources at a spatial resolution of 4º x 5º and monthly temporal resolution. The optimization also included a monthly global source from oxidation of methane and

biogenic NMVOCs. Results were evaluated with independent CO observations from surface sites (NOAA/GMD network), aircraft (MOZAIC), and satellite (TES).

An important first step was to evaluate the consistency of the satellite datasets used in the inversion. GEOS-Chem served as an intercomparison platform. We showed that MOPITT, AIRS, and TES (all observing in the 4.7 μm thermal infrared band) are consistent overall, and that apparent differences in the data are driven mainly by different averaging kernels and a priori information. SCIAMACHY (observing in the 2.3 μm solar IR band) is considerably noisier, but also consistent in suggesting a similar model a priori underestimate.

Our a posteriori estimate is 1350 Tg for direct emissions, with 217 Tg from oxidation of co-emitted VOCs. This represents a 60% underestimate of bottom-up inventories, but is within 25% of recent top-down estimates [*Arellano et al.*, 2004; *Arellano et al.*, 2006; *Pétron et al.*, 2004; *Stavrakou and Müller*, 2006]. CO source from oxidation of methane and biogenic NMVOCs changed by <1% from our a priori of 1280 Tg. GEOS-Chem driven by the a posteriori estimate from MOPITT, AIRS and SCIAMACHY improves the model bias against TES CO, an independent dataset.

A striking feature of our results is the larger-than-expected seasonal variation of CO emissions at northern mid-latitudes. Emissions in winter are 50% higher than in summer in the US and Europe, while up to 100% higher in winter in E. Asia. We mainly attribute our higher winter estimates in northern hemispheric midlatitudes mostly to residential heating (wood burning) in US and residential coal burning in China. Our annual a posteriori estimate is 49.5 Tg for the U.S. (48 states), 94.7 Tg for Europe, and

354 Tg for E. Asia (with 267 Tg for China alone). Our finding of increased seasonal amplitude is supported by independent observations from GMD and MOZAIC.

Our inverse model results indicate a large underestimate of tropical biomass burning in the GFED2 inventory [*van der Werf et al.*, 2006]. Annual a posteriori emission estimates are 343 Tg a^{-1} for Africa and 183 Tg a^{-1} for South America. However, the consistency among datasets is not as good in the southern hemisphere as in the north. In particular, AIRS implies larger biomass burning estimates than MOPITT or SCIAMACHY or the GMD surface sites, most likely due to AIRS high bias.

Our emission correction factors are a balance of information from the three datasets, even in the tropics and in the southern hemisphere, where a posteriori model bias suggests AIRS CO is higher than MOPITT and SCIAMACHY. We derived our conclusions from comparing results from the joint three dataset inversion with results from individual dataset inversions for a subset of three months. The a priori cost function contribution from model-AIRS differences is 78%, suggesting largest contribution to results from AIRS, but other than proper error characterization, there is no objective way to weigh the contributions from each dataset.

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Figures and tables

Table 1. Annual CO emissions: a priori and a posteriori estimates for selected regions Tg a^{-1}.

Figure 1. Seasonal a priori CO sources from fossil fuel, biofuel and biomass burning for May 1, 2004 - April 30, 2005. See text for details.

Figure 2. Seasonal variation of total CO combustion sources from the contiguous US (NEI99 region), Europe (EMEP region) and E. Asia (20-50ºN, 70-150ºE). A priori values for fossil fuel are from the NEI99 inventory for the US (with *Hudman et al.* [2008] 60% correction), EMEP inventory for Europe and *Streets et al.* [2006] for E. Asia. A posteriori values are from the inversion. Both a priori and a posteriori reflect total emission source, from direct emissions and rapid oxidation from co-emitted VOCs. Unit is Tg $month^{-1}$.

Figure 3. Seasonal variation of CO concentrations at remote surface sites. Climatological observations from NOAA/GMD (1988-2001) [*Novelli et al.* 2003] are shown in black, 2004-2005 observations are in blue. Vertical lines show interannual variability of monthly mean concentrations. GEOS-Chem model values are shown in red (a priori sources) and in green (a posteriori sources). Note the differences in scale between panels.

Figure 4. Seasonal variation of CO concentrations throughout the troposphere. Climatological aircraft observations from MOZAIC (2002-2007) [*Nedelec et al.* 2003] are shown in black, 2004-2005 observations are in blue. Vertical lines show interannual variability of monthly mean concentrations. GEOS-Chem model values are shown in red (a priori sources) and in green (a posteriori sources).

Figure 5**.** Annual daytime average CO columns observed by the MOPITT, AIRS, TES and SCIAMACHY satellite instruments over the period May 1, 2004 - April 30, 2005

(TES data starting September 2004). White space indicates lack of data. SCIAMACHY data include "cloud-free" data only, AIRS data include retrievals with corresponding temperature > 250K

Figure 6. Scatterplots of CO observational datasets vs. the GEOS-Chem model. Points represent daily observations averaged over the 2º x 2.5º grid of the model for the period May 2004 – April 2005, with the exception of TES (July 2005 – April 2006) and the GMD/MOZAIC data (monthly climatological averages as described in Figures 3 and 4). The green dashed line is the 1:1 relationship. The red solid line is a reduced-major-axis (RMA) fit. Correlation coefficients and slopes are given inset. Symbols on the top three panels are colored by their degrees of freedom (DOF) for signal. Units are 10^{18} molecules cm^{-2} for the satellite panels and 10^2 ppb for the GMD/MOZAIC panel.

Figure 7 Annual mean correction factors to the a priori combustion sources of CO from Figure 1 as derived from the adjoint inversion of MOPITT, AIRS, and SCIAMACHY CO columns for May 2004 - April 2005.

Figure 8. Ratio of a posteriori to a priori emission estimates for winter (DJF), spring (MAM), summer (JJA) and fall (SON) as derived in an adjoint inversion using MOPITT, AIRS and SCIAMACHY CO columns for May 1, 2004 to May 1, 2005. (a) N. America, (b) Europe and Middle East, (c) Asia, (d) Africa and S. America.

Figure 9. Fractional a priori and a posteriori model bias against MOPITT, AIRS and SCIAMACHY during September, October and November of 2004 from the three dataset inversion (top and middle rows); a posteriori model bias against MOPITT, AIRS and SCIAMACHY CO during the same months from individual dataset inversions (bottom row).

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