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Measurement of low-altitude CO over the Indian subcontinent by MOPITT

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[1] We show that the dayside MOPITT retrievals in the lower troposphere can provide useful information on surface sources of atmospheric CO over the Indian subcontinent. We find that MOPITT retrievals at 850 hPa show localized enhancements over the Indian subcontinent, which correlate with similar enhancements seen in the tropospheric NO_2 columns from the SCIAMACHY instrument. In particular, high concentrations of CO over the Indo-Gangetic basin and some prominent cities are captured in the lower-tropospheric retrievals in spring. MOPITT averaging kernels (normalized to take into account the absorber amounts in the layers) indicate that the retrievals are sensitive to CO in the lower troposphere. In winter, MOPITT retrievals at 850 hPa can detect the strongest source areas over the eastern states of Bihar and West Bengal, thus confirming the so-called ''Bihar pollution pool,'' which was detected earlier in the aerosol measurements by the multiangle imaging spectroradiometer (MISR) aboard Terra. The pollution features are consistent with the spatial distribution of CO emissions in India, as reflected in the GEOS-Chem simulation of CO. Furthermore, these lower-tropospheric features in the simulation are still present after smoothing the modeled fields using the MOPITT averaging kernels and a priori profile, demonstrating that the retrievals do have sensitivity in the lower troposphere. This work indicates that although MOPITT retrievals are often most sensitive to CO in the middle and upper troposphere, they do provide information on lower-tropospheric CO in selected continental regions with strong thermal contrast and could be useful for pollution studies.

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1. Introduction

[2] Carbon monoxide (CO) is an important tropospheric species inasmuch as it influences the atmospheric chemistry through its reaction with the OH radical. It is a by-product of combustion and is a useful tracer of atmospheric transport, because of its long lifetime (few weeks). In particular, emission and subsequent long-range transport of CO from Asia has been the focus of much attention in recent years, because of strong industrial activities and biofuel burning in this area [*Bey et al.*, 2001; *Liu et al.*, 2003; *Heald et al.*, 2003]. These efforts have relied on a combination of ground-based, aircraft, and space-based measurements. While intensive field campaigns using several platforms can greatly enhance our knowledge of the targeted species, long-term changes and trends over a large area can only be obtained through space-based remote sensing observations. Such measurements of CO have been made by the Measurements of Pollution in the Troposphere (MOPITT) since 2000 [*Drummond*, 1992]. MOPITT measurements have been used effectively to study CO plumes from forest fires [*Liu et al.*, 2005], intercontinental transport of pollution [*Edwards et al.*, 2003, 2006; *Heald et al.*, 2003], frontal lifting and convective transport of pollution to the upper troposphere [*Liu et al.*, 2006; *Kar et al.*, 2004, 2006; *Li et al.*, 2005], and for inverse modeling to quantify surface sources of CO [*Arellano et al.*, 2004, 2006; *Heald et al.*, 2004; *Kopacz et al.*, 2007].

[3] While MOPITT measurements have been useful in providing a global picture of the distribution of CO, their application for near-surface studies has been limited. Since the MOPITT operational retrievals employ radiances measured near 4.7 μm, they are sensitive to CO mostly in the middle and upper troposphere with generally less sensitivity in the lower troposphere, as compared to the CO measurements in the solar channel at 2.3 μm by SCIAMACHY [*Buchwitz et al.*, 2006, 2007]. It is generally assumed that all lower-tropospheric MOPITT retrievals have little sensi-

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tivity. This perception is due, in part, to the fact that the primary application of MOPITT has been in the context of intercontinental transport of pollution [e.g., *Edwards et al.*, 2003, 2006; *Heald et al.*, 2003; *Li et al.*, 2005], focusing, for example, on transpacific or transatlantic transport, where the lower-tropospheric retrievals do indeed have little sensitivity because of the cold ocean. However, *Deeter et al.* [2007] recently pointed out that interpretation of the vertical sensitivity of MOPITT retrievals, as reflected in the averaging kernels, has been influenced by the nonuniform thickness of the layers of the retrieval grid. They found that after normalizing for these grid effects, the averaging kernels indicate useful sensitivity to CO in the lower troposphere, particularly over dayside tropical regions with sparse vegetation [*Deeter et al.*, 2007]. In this paper we explore this issue of the sensitivity of the lower-tropospheric MOPITT retrievals with emphasis over the Indian subcontinent, where the thermal contrasts are expected to be high.

[4] We use lower-tropospheric MOPITT retrievals along with measurements of tropospheric NO_2 from the SCIAMACHY instrument to assess the sensitivity of the MOPITT retrievals to the surface sources of CO. We focus on India because the high population density and recent accelerated industrial activity imply strong sources of CO in the region. In addition, while a number of aerosol studies over India have been carried out in recent years with emphasis on the Indo Gangetic (IG) basin [e.g., *Singh et al.*, 2004; *Jethva et al.*, 2005; *Tripathi et al.*, 2006; *Nair et al.*, 2007], few studies have been published on the distribution and evolution of the gas species, particularly using the remotely sensed measurements of the various tropospheric species that are now available. We examine here the spatial structures in the distribution of CO in lower-tropospheric MOPITT retrievals and assess their consistency with our a priori knowledge of the CO sources in India, as reflected in the emission inventory in the GEOS-Chem model and in the model simulation of the horizontal and vertical structure of CO. We show that the low-altitude MOPITT retrievals do indeed capture some of the source information over this region.

2. Data

2.1. MOPITT

[5] The MOPITT instrument is onboard the Terra spacecraft, which is flying in a polar sun synchronous orbit at an altitude of 705 km. Upwelling infra red radiation near 4.7 μm is measured using gas correlation radiometry which is then used to retrieve the CO mixing ratios. The CO mixing ratio profile is reported at 7 levels (surface, 850, 700, 500, 350, 250 and 150 hPa levels) although these measurements are not independent of one another [*Deeter et al.*, 2004]. The MOPITT retrieval ($\hat{\mathbf{x}}$) can be described as a linear estimate of the true atmospheric state ($\mathbf{x}^{\text{true}}$)

$$\hat{\mathbf{x}} = \mathbf{x}_a + \mathbf{A}(\mathbf{x}^{\text{true}} - \mathbf{x}_a) + \varepsilon \quad (1)$$

where $\mathbf{x}_a$ is the MOPITT a priori CO profile, $\mathbf{A}$ is the MOPITT averaging kernel matrix, and ε is the retrieval error. The averaging kernel represents the sensitivity of the MOPITT retrieval to the true state of the atmosphere and provides a measure of the vertical resolution of the retrieval. The trace of the averaging kernel matrix indicates the number of independent pieces of information (degrees of freedom for signal) retrieved in the profile. MOPITT profiles of CO mixing ratios typically have between 1.4–1.8 degrees of freedom mostly in the tropics and fall off sharply at higher latitudes [*Deeter et al.*, 2004].

[6] The MOPITT version 3 retrievals that are used in this analysis have been validated using in situ aircraft measurements and the results show good agreements with the average bias being less than 20 ppbv at all levels [*Emmons et al.*, 2004]. We have used only the dayside retrievals from MOPITT in this work as these retrievals have the maximum information content [*Deeter et al.*, 2004].

2.2. SCIAMACHY

[7] The SCIAMACHY instrument was launched on the European Envisat spacecraft on 1st March 2002 in a sun-synchronous orbit with an inclination of 98.55° at an altitude of 800 km [*Bovensmann et al.*, 1999]. The instrument measures backscattered solar radiation in the ultraviolet, visible, and near-infrared regions of the spectrum, from 0.24–2.38 μm, in the nadir and limb modes. The instrument also makes limb observations using solar and lunar occultation as well as scattered light. In the nadir mode the horizontal resolution is 30 km × 60 km, and the instrument achieves global observational coverage within 6 days. The column abundances of a broad range of trace gases have been retrieved from SCIAMACHY radiances. We use retrievals of the tropospheric column abundance of NO_2 using DOAS algorithm [*Richter et al.*, 2005].

3. Model Description

[8] The GEOS-Chem model is a global three-dimensional chemical transport model, driven by assimilated meteorology from the Goddard Earth Observing System (GEOS-4). This model has been widely used for various studies of tropospheric chemistry and transport [e.g., *Bey et al.*, 2001; *Fiore et al.*, 2003; *Heald et al.*, 2004; *Liu et al.*, 2006; *Zhang et al.*, 2006]. It has been used previously with MOPITT data for the study of long-range transport of pollution [e.g., *Heald et al.*, 2003; *Li et al.*, 2005] and for inverse modeling of atmospheric CO [e.g., *Heald et al.*, 2004; *Arellano et al.*, 2004, 2006; *Kopacz et al.*, 2007]. The version of GEOS-Chem used here is v7-02-04 (http://www-as.harvard.edu/chemistry/trop/geos/index.html) with a horizontal resolution of 2° × 2.5° and with 30 levels in the vertical from the surface to 0.01 hPa. We simulate the tropospheric CO distribution for the years 2000–2004 for comparison with MOPITT using a linearized version of the CO chemistry in which monthly mean abundances of OH, the main sink for CO, are specified from a model simulation with full nonlinear tropospheric chemistry [*Evans and Jacob*, 2005].

[9] The inventory of CO emissions employed in the model is described in *Duncan et al.* [2007] with emissions for the base year of 1985 in the inventory scaled to 1998. For the period of interest here, May through December, the total CO emissions in India are 27 Tg CO, with fossil fuel combustion, biomass burning, and biofuel combustion accounting for 15%, 22%, and 63% of the total, respectively. The spatial distribution of these emissions is plotted in

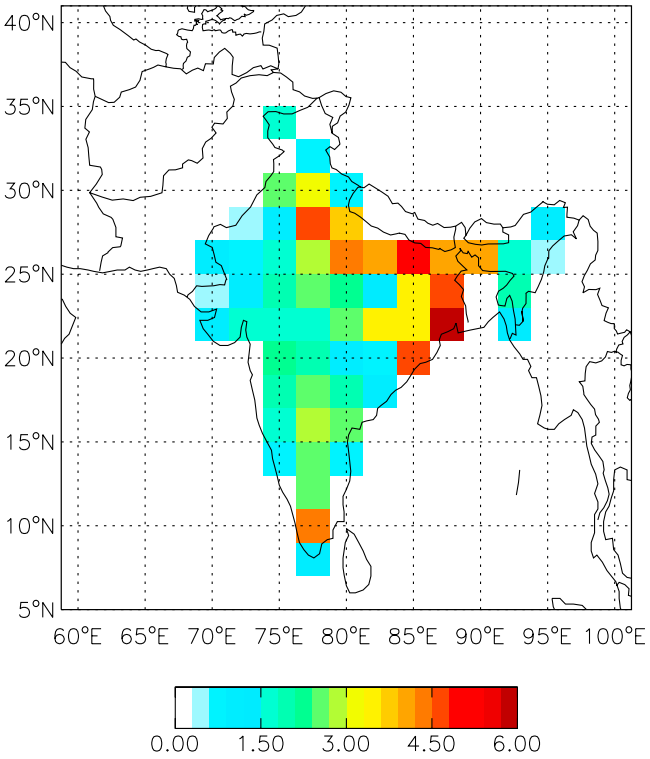

Figure 1. Averaged May–December emissions of CO used in the GEOS-Chem simulation. Values shown include the direct emissions of CO from fossil fuel combustion, biomass burning, biofuel combustion, and the secondary source of CO from the oxidation of hydrocarbons emitted by these processes. The units are 10^{18} molecules cm^{-2} $month^{-1}$.

Figure 1 and shows that the emissions are highest in northern India, along the IG basin and over the Bihar and West Bengal region. The studies of *Heald et al.* [2004] and *Kopacz et al.* [2007] conducted inversion analyses for Asian emissions during February through April 2001, during the biomass burning season, and found that the model significantly overestimated Indian emissions; they found that biomass burning accounts for 79% of emissions in February through April. As our focus is on MOPITT data for May–December, when biomass burning provides a significantly smaller contribution to the total Indian emissions, the analysis presented here will not be influenced by this potential overestimate of biomass burning in the model.

4. Results

4.1. CO Over Indo Gangetic Basin in Spring

[10] Figure 2 shows the distribution of MOPITT CO mixing ratios at 850 hPa as well as the tropospheric column NO_2 from the SCIAMACHY instrument for May 2004 over the Indian subcontinent. The data drop outs in the CO distribution over the southern parts of the country are due to clouds that can also be seen in the outgoing long-wave radiation (OLR) maps from NCEP as well as from MODIS cloud fraction maps during May 2004 (not shown). The reason why data drop outs are not seen in the corresponding NO_2 distribution could be related to the comparatively weaker cloud screening employed in the SCIAMACHY retrievals. Despite these data drop outs, strong spatial gradients can be seen in the CO 850 hPa distribution. In particular, there are several areas of localized enhancements in the CO 850 hPa map, which are also seen in the corresponding NO_2 distribution. Note the enhanced CO and NO_2 values over the IG basin (~24–28°N, 75–85°E) as well as over West Bengal and Bangladesh (~22–25°N, 85–90°E), essentially following the very high population density in this region. This is quite similar to the recent GIS based CO Indian national inventory (1° × 1°) as well as higher resolution (0.5° × 0.5°) regional emission inventory in Asia (REAS) version 1.1 [*Dalvi et al.*, 2006, their Figure 6; *Ohara et al.*, 2007]. The MOPITT CO distribution is also consistent with high anthropogenic pollution levels along the IG basin observed in the satellite measurements of aerosol and tropospheric column ozone distributions [*Jethva et al.*, 2005; *Fishman et al.*, 2003]. Note that there are some differences in detail between the CO and NO_2 distributions, along the south eastern coast and over the north-eastern states (20–28°N, 92–96°E). A direct one-to-one correlation

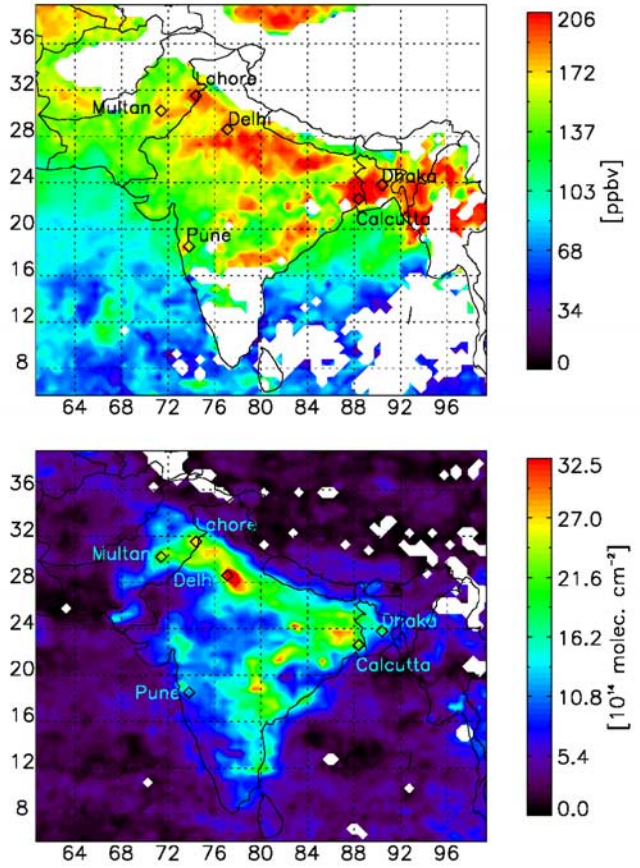

Figure 2. Distribution of MOPITT CO mixing ratios (ppbv) at 850 hPa (top) and SCIAMACHY tropospheric column NO_2 (10^{14} cm^{-2}; bottom). Data have been binned in 0.5° in latitude and longitude. Both distributions are for May 2004. Only daytime data from MOPITT measurements have been used. The white areas indicate data dropouts due to clouds or topography. Locations of some of the cities with large population are marked out.

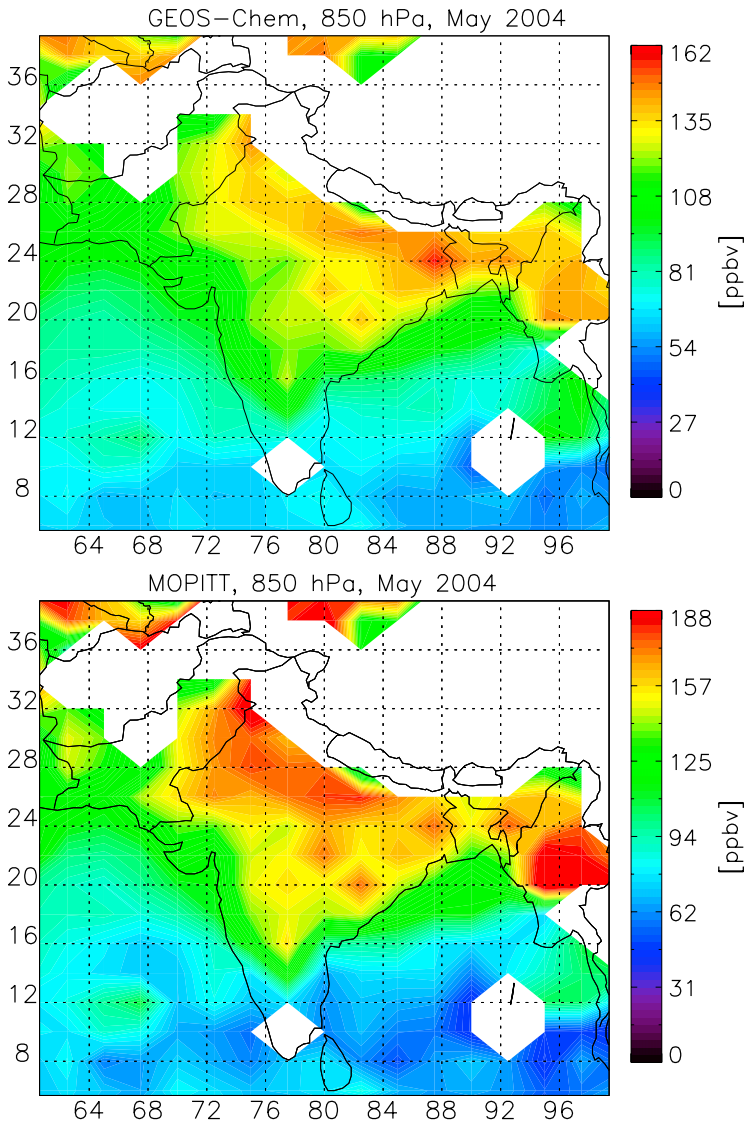

Figure 3. CO mixing ratio at 850 hPa (ppbv) over the Indian subcontinent in May 2004, as simulated by GEOS-Chem model after applying the MOPITT averaging kernels (top) and the corresponding MOPITT CO at 850 hPa (bottom) at the model resolution ($2^\circ \times 2.5^\circ$). Note the different scales in the two cases.

between CO and NO_2 may not always be expected because of differences in the sources and their lifetimes [*Buchwitz et al.*, 2007]. Over India, domestic biofuel burning dominates the CO emissions [~50%, *Dalvi et al.*, 2006], while the NOx emissions are dominated by fossil fuel burning with power and transport sectors contributing about 30% each to the NOx emissions [*Garg et al.*, 2001; *Kunhikrishnan et al.*, 2006]. Further, nonuniform temporal coverage by the two instruments and differences in retrieval characteristics associated with aerosol interference may also contribute to some of the differences between the CO and NO_2 observations.

[11] In contrast to CO, NO_2 is a short-lived species and its distribution is more indicative of local surface pollution sources [*Richter et al.*, 2005]. Furthermore, SCIAMACHY measurements are sensitive to NO_2 in the boundary layer and can clearly localize emissions to individual cities [e.g., *Buchwitz et al.*, 2007]. This can also be seen in Figure 2 (bottom) with isolated plumes of NO_2 near the locations of some cities in the subcontinent with large populations and associated high pollution levels. Interestingly, signatures of these cities can also be seen in the CO 850 hPa map. Note the strong isolated plume of CO over Pune, which was also observed in the CO total column measurements by SCIAMACHY [*Buchwitz et al.*, 2007]. However, the NO_2 plume is located somewhat to the north of the CO plume, toward Bombay. There are also similar but somewhat extended plumes of CO around other cities like Delhi, Lahore and Multan. The CO mixing ratios over Calcutta and Dhaka are very high and the individual plumes cannot be distinguished clearly in this color scale. Nonetheless, it is reasonably clear that the low-altitude MOPITT retrievals are capturing some of the source information in this case, which was hitherto thought to be improbable.

[12] The CO distribution at 850 hPa level in May 2004 simulated by the GEOS-Chem model is shown in Figure 3 (top). For comparison, Figure 3 (bottom) shows the

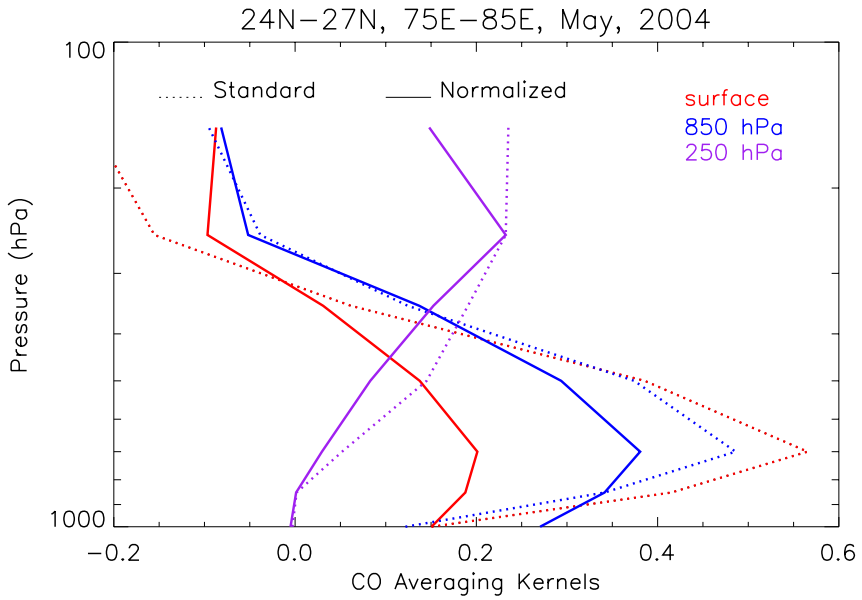

Figure 4a. MOPITT mean standard (dashed lines) and grid-normalized averaging kernels (solid lines) for the month of May 2004 over 24–27°N and 75–85°E, which includes the Indo Gangetic basin.

MOPITT mixing ratios at 850 hPa gridded to the GEOS-Chem model resolution of 2° × 2.5°. MOPITT averaging kernels and the a priori have been applied to the model runs shown in the figure. The two distributions are qualitatively quite similar, although the model underestimates the CO mixing ratios. The underestimate is likely due to the CO emission inventory used in the model. In particular, the model bias is largest in northwestern India, where the specified emissions in the model are low (Figure 1). *Beig and Brasseur* [2006], for example, have estimated an increase of 10–20% in CO mixing ratios near the IG plains between 1991 and 2001 for the month of July. However, the modeled CO spatial distribution is quite similar to that simulated for May 2001 by *Beig and Ali* [2006] using the MOZART-2 chemical transport model with the recent Indian national inventory for CO emissions [*Dalvi et al.*, 2006]. As our focus here is primarily to demonstrate the sensitivity of MOPITT retrievals to CO in the lower troposphere, we do not attempt to optimize the model emissions based on the MOPITT observations. However, the consistency between spatial distributions of CO and NO_2 suggests that a coupled, multispecies inversion analysis of CO and NO_2, accounting for the different emission factors for these species, from biomass burning and fuel combustion, may provide valuable information to help independently quantify the CO emissions from these different source types.

[13] The MOPITT retrievals are performed with a uniform a priori profile, which has a maximum concentration of CO of 120 ppbv at the surface. The significant departure of the retrievals shown in Figure 2 from the a priori values indicate significant sensitivity to lower atmospheric CO in the retrievals for this region. The sensitivity of the MOPITT retrievals to CO in the lower troposphere can be assessed by examination of the MOPITT averaging kernels. However, as pointed out by *Deeter et al.* [2007], the nonuniform spacing between the retrieval levels (thus layers with nonuniform thickness) affects physical interpretation of the averaging kernels, which are proportionately larger for levels in the true profile associated with thicker layers. Grid-normalized averaging kernels take into account the thickness of the layers (thus the absorber amounts) and these should be used for a proper interpretation of the averaging kernels [see *Deeter et al.*, 2007 for details]. Figure 4a shows the standard mean MOPITT averaging kernels for May 2004 over 24–27°N and 75–85°E roughly corresponding to the IG basin area, as well as the grid-normalized kernels for the surface, the 850 hPa and the 250 hPa levels. Firstly note that the averaging kernels for both the surface and 850 hPa are nonzero at the surface thus indicating sensitivity to the surface sources of CO. Secondly, the normalized kernel

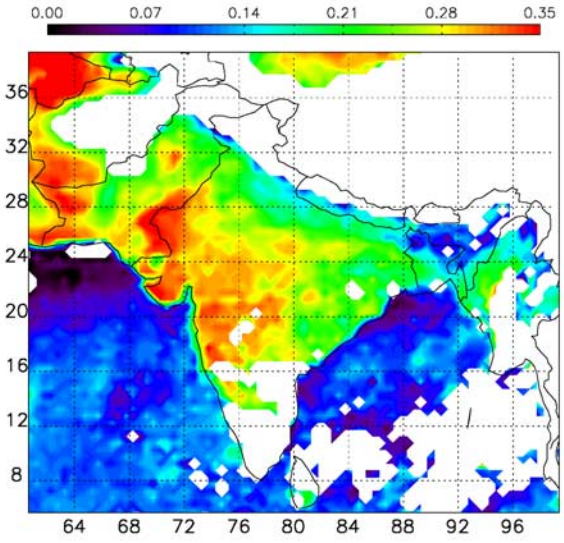

Figure 4b. Distribution of the surface value of MOPITT grid-normalized averaging kernel for 850 hPa for the month of May 2004. Data binned in 0.5° in latitude and longitude and white areas indicate missing data.

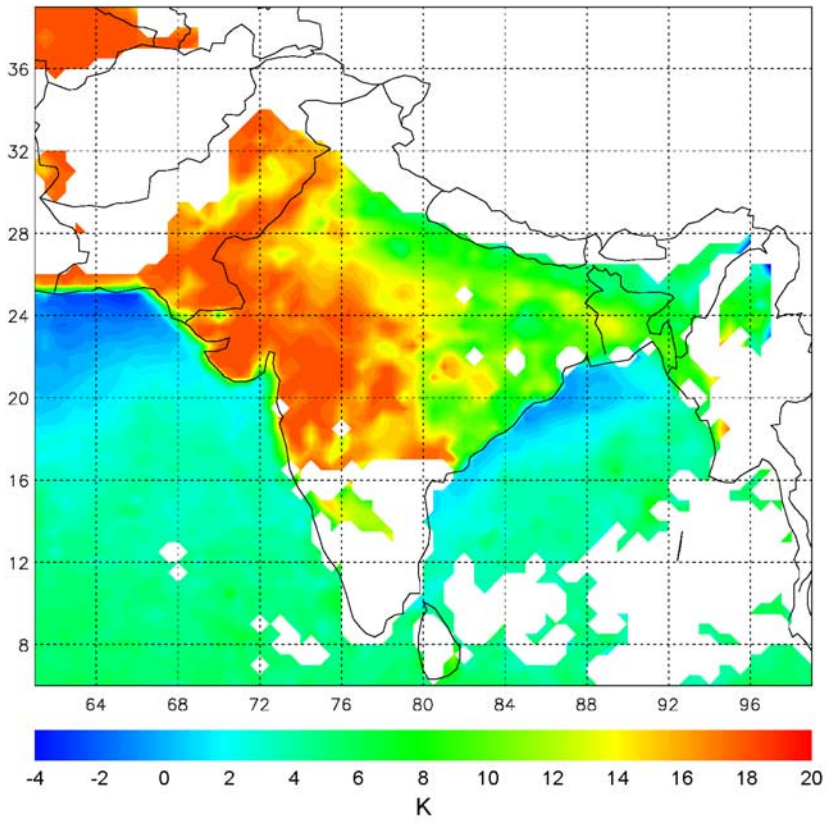

Figure 5. Thermal contrast, defined as the difference between the surface skin temperature (retrieved by MOPITT) and air temperature at 925 hPa from NCEP reanalyses data for May 2004. Data binned in 0.5° in latitude and longitude and white areas indicate missing data.

for the surface shows nearly uniform sensitivity to layers from the surface to ~500 hPa, in contrast to the standard kernel, which shows far higher sensitivity to 700 hPa level as compared to the surface. Similarly the normalized averaging kernel for 850 hPa also shows enhanced sensitivity to the surface as compared to the standard kernel.

[14] The distribution of the grid-normalized 850 hPa averaging kernel evaluated at the surface over the Indian subcontinent in May 2004 is shown in Figure 4b. This quantity indicates the sensitivity of the retrieved amount at the 850 hPa level to perturbations of the absorber amount of the surface layer in the true profile. Significant sensitivity to the boundary layer pollution can be seen over most parts of the country with the highest sensitivity occurring over the deserts of Rajasthan (~24–30°N, 68–75°E) in the west. Interestingly a pronounced minimum sensitivity can be seen over the north-eastern states between 90–96°E, which could be due to the presence of significant forested areas there. *Deeter et al.* [2007] found similar low sensitivity over the Congo and Amazon basins and attributed this to evaporation and evapotranspiration processes.

[15] The sensitivity of the MOPITT retrievals to lower-tropospheric CO should be related to the thermal contrast between the surface and the lowest layers of the atmosphere. Higher thermal contrast yields higher sensitivity to lower-tropospheric CO. Figure 5 shows the difference between the surface skin temperature, as retrieved by MOPITT and the atmospheric temperature at 925 hPa level, as obtained from the NCEP reanalyses for May 2004. In general, the temperature contrast is quite significant over most of the region with maximum values of ~20 K over the western parts of the subcontinent. Also, the thermal contrast map is generally consistent with the grid-normalized 850 hPa averaging kernel map (Figure 4b), as might be expected. Although the thermal contrast should be estimated between the surface and a layer immediately higher up, we have used the NCEP temperature at 925 hPa to avoid data uncertainties associated with topographical variations at lower levels. Further, the retrieval errors and the percent a priori diagnostic at 850 hPa level also show similar distributions over the country with lower values in the western parts with larger thermal contrasts (not shown).

[16] The likelihood of directly sensing the surface sources of CO in May is further increased by the large planetary boundary layer (PBL) heights over India at this time. The PBL heights obtained from the European Center for Medium-range Weather Forecasts (ECMWF) data for 6 UT corresponding to ~11:30 am local time which is quite close to the MOPITT overpass time (10:30 am) indicate PBL heights in excess of 2 km over much of the central part of the country and reaching 3 km or more over the IG basin (not shown). It should be noted that the CO mixing ratio retrieved by MOPITT would depend upon both the sensitivity as well as the strength of the source itself. In particular, over the IG basin, the sensitivity of MOPITT retrievals to emissions in the surface layer is lower than over the western regions and

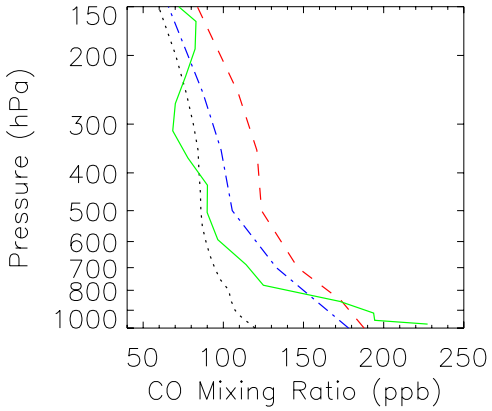

Figure 6. Comparison of MOPITT and GEOS-Chem profiles of CO on 29 May 2004 at 87°E, 24°N. The MOPITT profiles have been averaged on the GEOS-Chem 2° × 2.5° grid. The retrieved MOPITT profile and the a priori profile used in the retrieval are shown as the red dashed and black dotted lines, respectively. The GEOS-Chem profile of CO is shown as the green solid line, whereas the modeled profile transformed by the MOPITT averaging kernels and a priori profile is indicated by the blue dash-dotted line.

yet the retrievals provide information about the sources because of the high source strength over the basin.

[17] The vertical structure of the MOPITT and modeled profiles over eastern India (24°N, 87°E) on 29 May 2004 is shown in Figure 6. Because of the strong surface source of CO in the region, the modeled profile decreases from a maximum CO abundance of about 230 ppbv at the surface to background values at about 600 hPa. Above 600 hPa, the modeled CO profile is similar to the MOPITT a priori profile. Transforming the model profile with the MOPITT averaging kernels and a priori profile smoothes the modeled profile. However, the smoothed model profile still shows significantly higher abundances of CO in the lower troposphere than in the middle and upper troposphere, due to the sensitivity to lower troposphere in the retrievals as reflected in the averaging kernels.

[18] Figure 7a shows the distribution of CO mixing ratios at 850 hPa averaged over the month of May for the years 2000, 2002, 2003, 2005, 2006, and 2007. Data for May 2001 are not available because of instrument issues [*Emmons et al.*, 2004]. The strong enhancement over the IG basin is very clearly observed for each year with significant interannual variations. This indicates the persistent nature of this pollution feature and shows that MOPITT is consistently capturing source information. However, the signature of the various cities in the form of isolated plumes

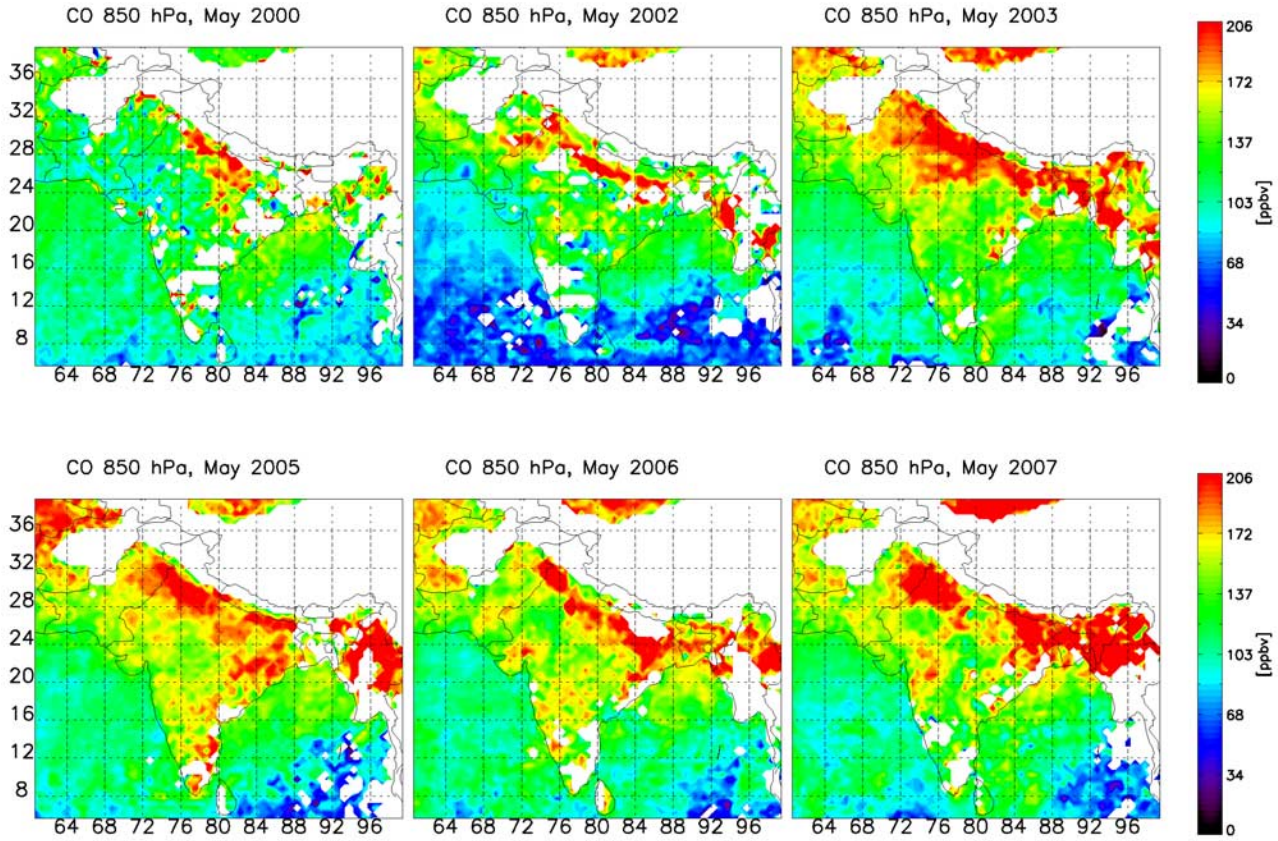

Figure 7a. CO mixing ratio (ppbv) retrieved at 850 hPa level by MOPITT over the Indian subcontinent for May 2000, 2002, 2003, 2005, 2006, and 2007. Data binned in 0.5° in latitude and longitude, and white areas indicate missing data. Only daytime data have been used. Data for May 2001 are not available.

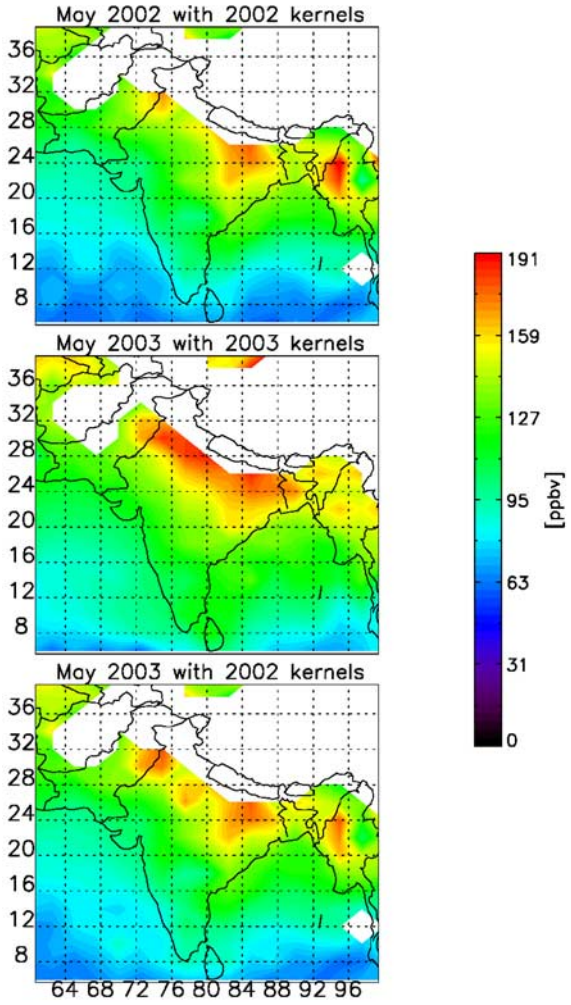

Figure 7b. Modeled CO mixing ratios for May 2002 convolved with the corresponding MOPITT averaging kernels (top), for May 2003 convolved with the corresponding MOPITT kernels (middle), and for May 2003, but convolved with the MOPITT kernels for May 2002 (bottom).

is not seen uniformly from year to year for the same month, with the plumes associated with specific cities showing up strongly in some years and not in others. This might be related to the local meteorology and data coverage issues and needs to be further explored.

[19] The inter annual variation is strongest in the early part of the decade with distinct lower values in 2000 and 2002 as compared to 2003–2007, the latter years showing much less variability. We have investigated the possible reasons for this variability by focusing on the two months of May 2002 and May 2003; MOPITT observations in 2003 show significantly more CO than in 2002. We compared the GEOS-Chem simulation of CO for May 2002 and 2003, using the same emissions in the model for both years. As shown in Figure 7b, the modeled CO abundance, after smoothing with the MOPITT averaging kernels was significantly greater in May 2003 than in May 2002. To assess the potential impact of differences in the MOPITT observational coverage and vertical sensitivity on the modeled CO for 2002 and 2003, we sampled the May 2003 modeled distribution based on the observational coverage for May 2002 and applied the 2002 averaging kernels to smooth the fields. This resulted in lower abundances of CO (Figure 7b, bottom) than that obtained with the 2003 averaging kernels (Figure 7b, middle), but more than that produced with the modeled data and averaging kernel for 2002 (Figure 7b, top). The results in Figure 7b show that the differences between May 2002 and 2003 reflect a combination of differences in the vertical sensitivity and spatiotemporal sampling of the MOPITT retrievals and in atmospheric transport of CO due to the different meteorological conditions. Properly quantifying the relative contributions of these different factors to the year to year variability shown in Figure 7a will require a more detailed modeling analysis, which is beyond the scope of this paper.

[20] A noticeable difference between the GEOS-Chem meteorological fields for May 2002 and 2003 is that the mean PBL height, averaged over the area defined by 20–30°N and 68–88°E, was about 400 m higher in May 2003 (with a height of 2.25 km) as compared to May 2002 (with a height of 1.84 km). An increased PBL height should mix the boundary layer pollution higher up where MOPITT has better sensitivity and may explain the larger variation than simulated in the model, with constant emission. Further, the cloud fraction also showed a significant decrease from 0.43 in May 2002 to 0.27 in May 2003. This can also be seen in the enhanced MOPITT data dropouts in May 2002 (Figure 7a). This non-uniform sampling may also contribute to the differences.

4.2. "Bihar Pollution Pool" in CO and NO_2 in Winter

[21] *Di Girolamo et al.* [2004] had earlier observed high aerosol optical depths over the eastern states of Bihar and West Bengal in winter (the so-called "Bihar pollution pool") from the data obtained by the MISR instrument, also on Terra. As they pointed out, strong subsidence and light winds over this area in winter prevent venting of the boundary layer pollution to higher altitudes, accumulating and limiting it close to the source locations. Therefore it would be interesting to explore the sensitivity of MOPITT to low-altitude pollution in this area in winter.

[22] Figure 8 (top) shows the MOPITT CO mixing ratios retrieved at 850 hPa level for the months of October, November, and December 2004. The corresponding tropospheric columns of NO_2 retrieved from SCIAMACHY are shown (bottom). In October, relatively enhanced CO emissions can still be seen along the IG basin, although at a much lower level as compared to the month of May. The high CO mixing ratios seen below 20°N and between 80–100°E are likely the emissions from Southeast Asia and China which are transported to the Indian ocean. Note that there are no MOPITT data over much of the IG basin region for November and December. This has been observed in the MOPITT data consistently for all years for the months of November–January. The reason for this is not clear at this time but could be related to the intense haze and foggy conditions that develop each winter over this area [*Tripathi et al.*, 2006; *Gautam et al.*, 2007] that might be affecting the CO retrievals, although thick fog layers should also affect NO_2 retrievals. However, the evolution of CO in the lower troposphere in the rest of the subcontinent can still be studied. In particular note the large plume of CO that

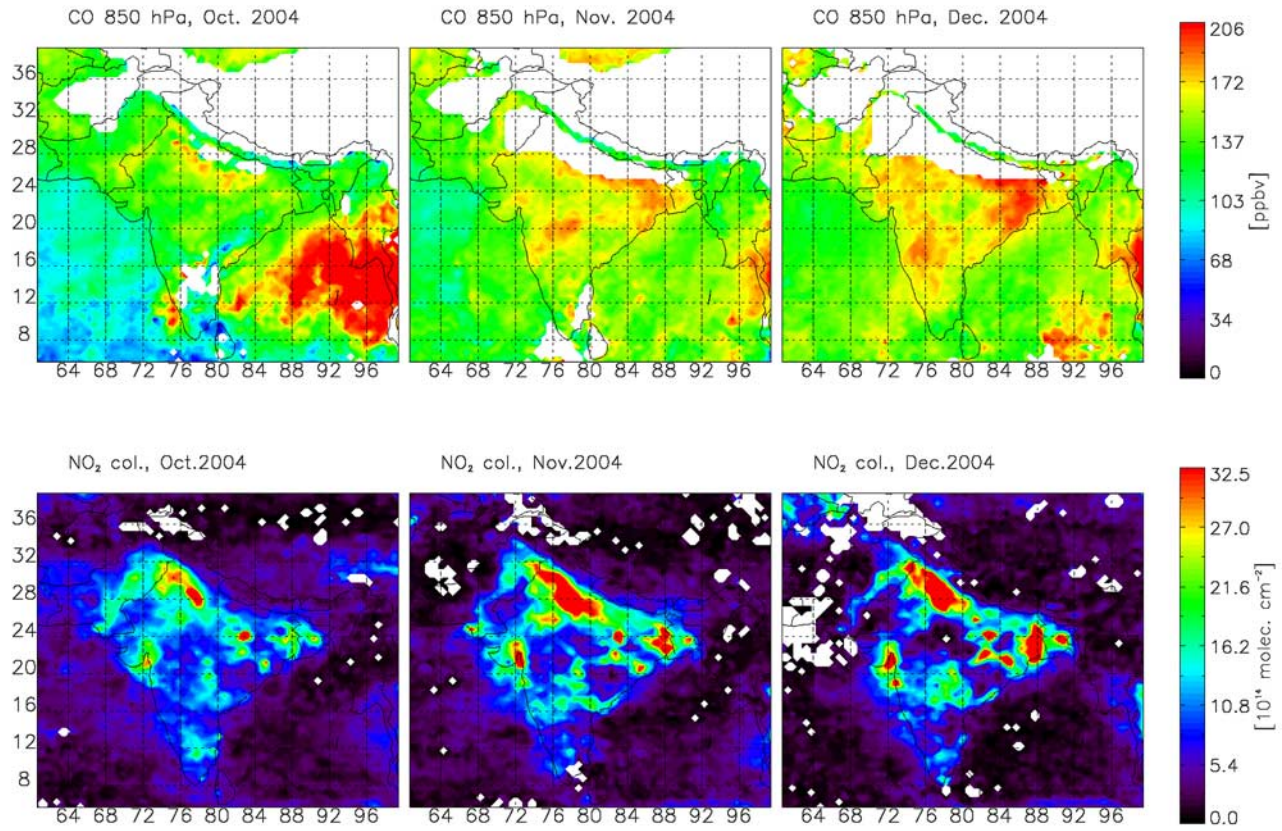

Figure 8. MOPITT CO mixing ratios at 850 hPa (ppbv, top) and SCIAMACHY tropospheric NO_2 columns (10^{14} cm^{-2}, bottom) for the months of October–December 2004. Data binned in 0.5° in latitude and longitude and white areas indicate missing data.

develops over the states of Bihar and West Bengal (~20–25°N, 84–88°E) in November and further increases in December. This can be seen in the corresponding NO_2 distribution as well. There is no biomass burning in this region in winter [*Di Girolamo et al.*, 2004]. Therefore this CO is likely to be the locally generated pollution from mostly fuel combustion. Thus both the MOPITT CO data at 850 hPa as well as the NO_2 tropospheric columns confirm the Bihar pollution plume during winter over this area in the gas species, which is remarkably similar to the aerosol climatology of *Di Girolamo et al.* [2004] (their Figure 3), clearly indicating the concentration of pollution which should have important implications for atmospheric chemistry and climate in this region.

[23] Figure 9 shows the corresponding GEOS-Chem simulations for October–December 2004 at the 850 hPa level after incorporating the MOPITT averaging kernels (top). To facilitate comparison between the model and the data we have replotted the MOPITT 850 hPa mixing ratios for the 3 months at the GEOS-Chem resolution grid (bottom). The evolution of the CO distribution in the lower troposphere as seen in MOPITT retrievals is quite similar to the model results with the plume over the eastern states of Bihar and West Bengal reaching a maximum in December. Analysis of the GEOS-Chem model simulations in which we tagged the CO emissions from different geographical regions show that the CO observed over the Bay of Bengal in the model is from emissions in East Asia and Southeast Asia (not shown). The modeled CO mixing ratios are lower than the observed values across India and could be due to an underestimate of Indian emissions.

[24] The subsidence in the region is reflected in the vertical profiles of CO simulated by the GEOS-Chem model, shown in Figure 10. The profiles show high abundances of CO in the boundary layer, exceeding 250 ppbv, and low concentrations in the free troposphere, less than about 80 ppbv, which is characteristic of background concentrations of CO in the northern hemisphere. The midtropospheric concentrations of CO in the model in this region are in fact less than the MOPITTa priori. For the scene shown in Figure 10a, the boundary layer is capped at about 800 hPa, whereas in Figure 10b the boundary layer is capped at about 700 hPa. Transforming the modeled profiles using the MOPITT averaging kernel and the a priori profile significantly smoothes the profiles because of the coarse vertical resolution of the retrievals. However, the significant departure of the smoothed model profile from the a priori in the lower troposphere indicates sensitivity to lower-tropospheric CO, as reflected in the MOPITT averaging kernels.

[25] The vertical structure of CO in the model depicted in Figure 10, with low concentrations of CO in the free troposphere, is indicative of the distribution of CO across the region. This suggests that the structures in the CO distribution that are present in the modeled fields at the 850 hPa level after application of the MOPITT averaging kernel and a priori profile are not due to the influence of CO

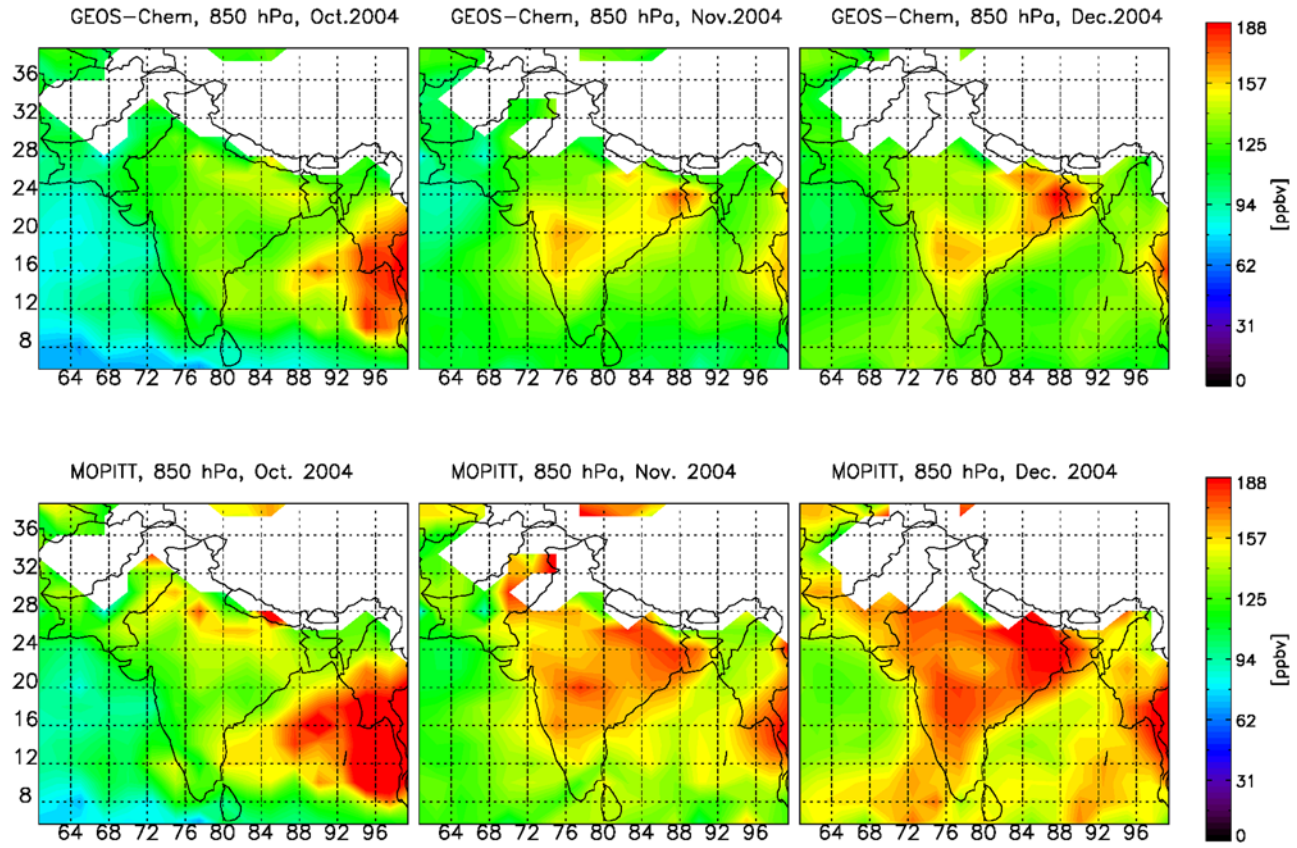

Figure 9. CO mixing ratios at 850 hPa (ppbv) from October to December 2004 as simulated by GEOS-Chem after application of the MOPITT averaging kernels for each month (top). The corresponding MOPITT data at the model resolution grid (2° × 2.5°) are shown (bottom). Missing data are shown in white.

in the middle and upper troposphere. Comparison of the smoothed model and MOPITT profiles in Figure 10 indicate that the model does provide an underestimate of the middle and upper tropospheric CO. The bias in the model is larger in the upper troposphere than in the lower troposphere. Indeed, for the observations in Figure 10b, the model is biased slightly high relative to MOPITT in the lower troposphere, whereas it is biased low in the upper troposphere. Because the averaging kernels are broad (not shown), with the greatest sensitivity in the free troposphere, the presence of a plume of CO in the middle and upper troposphere will influence the retrieved CO in the lower troposphere. However, to produce the large departures of the retrieved profile from the a priori in the lower troposphere, as shown in Figure 10, requires the presence of a large source of CO in the lower troposphere. We use the model here as a proxy for the vertical structure of the true atmosphere, and although the model simulation does provide an underestimate of the CO abundance in the middle and upper troposphere, and therefore an underestimate of the influence of CO at these levels to the lower-tropospheric retrievals, the agreement in the vertical structure of the smoothed model profiles and the retrievals does demonstrate that the MOPITT retrievals do have sensitivity to lower-tropospheric CO.

[26] The distribution of the grid-normalized 850 hPa averaging kernels evaluated at the surface for the month of December 2004 is shown in Figure 11. While the sensitivity to the lower-tropospheric pollution has decreased somewhat in the western region as compared to May, it is still quite significant over most of the country, particularly over the Bihar pollution pool region. The latter, coupled with the strong and trapped emission makes it possible for MOPITT to detect the Bihar pollution pool in winter. The Bihar pollution pool was observed in MOPITT data for years other than 2004 as well, as can be seen in Figure 12, showing the CO distribution at 850 hPa for 2000, 2001, 2002, 2003, 2005, and 2006, averaged for the month of December. Once again strong interannual variability can be seen in the December maps. Tagged CO simulation runs indicate contributions from the South East Asia and East Asian source regions during December, variations of which tend to control the interannual variability of CO seen in the south-central regions of the country.

[27] Note that in the December CO maps (Figures 8 and 12), apart from the Bihar pollution plume, the spatial gradients are not as strong as in May. In particular, the cities are not easily distinguishable in the CO 850 hPa map as such. This could be due to the increased lifetime of CO in winter making it more susceptible to transport effects. In fact, pollutants from the southern areas are transported out to the Arabian sea by moderately high easterly winds [*Di Girolamo et al.*, 2004] as indeed can be seen in the December CO maps for various years. Similar plumes of

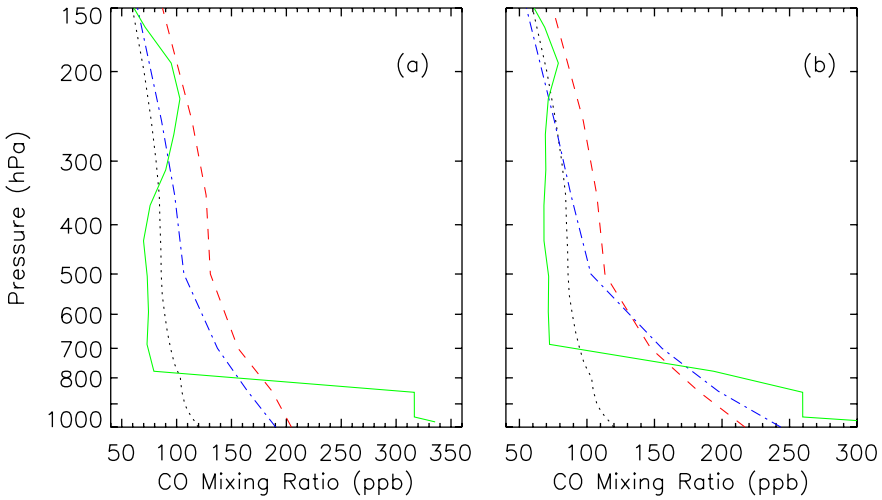

Figure 10. Comparison of MOPITT and GEOS-Chem profiles of CO on 14 December 2004 at (a) 84°E, 20°N and (b) 88°E, 24°N. The MOPITT profiles have been averaged on the GEOS-Chem 2° × 2.5° grid. The retrieved MOPITT profile and the a priori profile used in the retrieval are shown as the red dashed and black dotted lines, respectively. The GEOS-Chem profile of CO is shown as the green solid line, whereas the modeled profile transformed by the MOPITT averaging kernels and a priori profile is indicated by the blue dash-dotted line.

advected CO can also be seen over the Bay of Bengal, particularly for the years 2003, 2005, and 2006.

[28] Similar strong and localized enhancements in CO mixing ratios at 850 hPa were seen for other months as well (not shown), including signature of strong biomass burning emissions in March as also for all the years with available data. It is clear that the MOPITT retrievals of low-altitude CO are able to capture at least part of the lower-tropospheric pollution particularly where the sources are the strongest. This is consistent with the work of *Deeter et al.* [2007], even though the total CO columns might give quantitatively lower estimates over these areas as compared to SCIAMACHY, the latter having better sensitivity to the boundary layer [*Buchwitz et al.*, 2006, 2007].

5. Conclusions

[29] We have presented evidence that MOPITT dayside retrievals in the lower troposphere over continental areas with strong thermal contrast are able to provide useful information on surface emissions of CO. In particular over the Indian subcontinent, the low-altitude retrievals can effectively delineate the strong surface source regions over the IG basin in spring as well as signatures of some large cities. In winter, the evolution of the so-called Bihar pollution pool was also clearly visible in the MOPITT 850 hPa retrievals. The large scale pollution features seen in the CO distribution were also seen in tropospheric column NO_2 data from SCIAMACHY, thus indicating that MOPITT low-altitude retrievals were indeed capturing the source regions. These pollution features were shown to be consistent with a priori knowledge of the spatial distribution of CO emissions in India as reflected in the GEOS-Chem simulation of CO over this region, although the model provided an underestimate of the observed CO abundance.

[30] The sensitivity of the MOPITT retrievals in this region to lower-tropospheric CO was further demonstrated using vertical profiles of CO from the GEOS-Chem simulation in December when the CO emissions in the model are trapped in the lower troposphere as a result of atmospheric subsidence. We showed that the enhanced CO in the lower troposphere is still present in the modeled fields after smoothing with the MOPITT averaging kernels. This study indicates that, MOPITT retrievals may provide useful information on CO abundances in the lower troposphere, and together with complementary satellite data, could be used

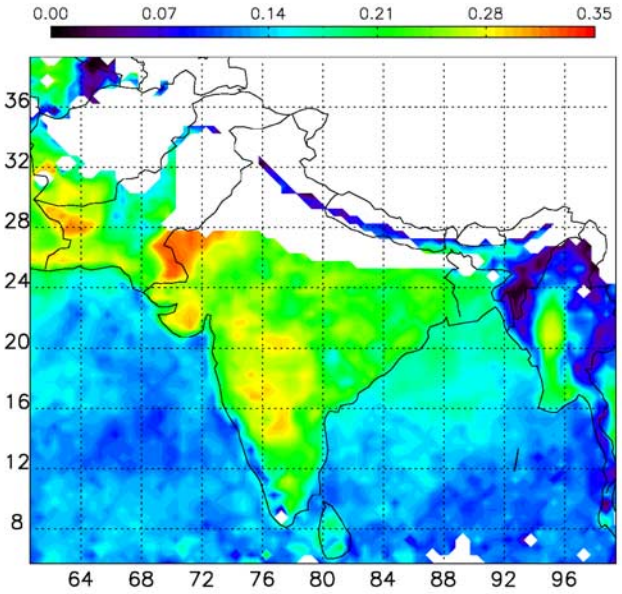

Figure 11. Surface value of the 850-hPa grid normalized averaging kernel for December 2004. Data binned in 0.5° in latitude and longitude and white areas indicate missing data.

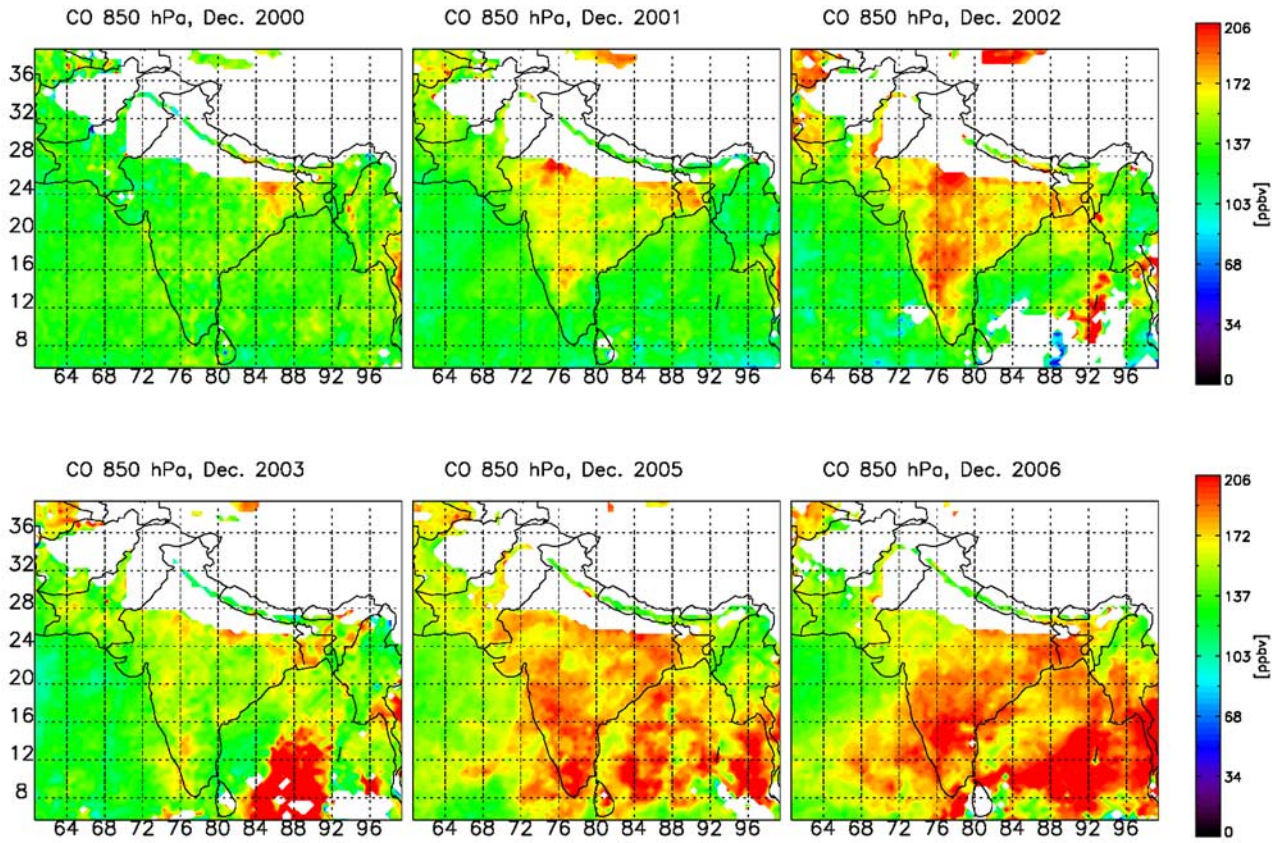

Figure 12. CO mixing ratio (ppbv) retrieved at 850 hPa level by MOPITT over the Indian subcontinent for December 2000, 2001, 2002, 2003, 2005, and 2006. Only daytime data have been used. Data binned in 0.5° in latitude and longitude and white areas indicate missing data.

for air quality studies. In particular, the evolution of regional pollution features like the Bihar pollution pool can be studied and monitored, since the temporal coverage of MOPITT now extends over 7 years. Work is needed to extend this analysis to other high-pollution regions in other parts of the globe.

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