Validation and Analysis of MOPITT CO Observations of the Amazon Basin

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Abstract.

We analyze satellite retrievals of carbon monoxide from the MOPITT instrument over the Amazon Basin, focusing on the MOPITT Version 6 "multispectral" retrieval product (exploiting both thermal-infrared and near-infrared channels). Validation results based on in-situ vertical profiles measured between 2010 and 2013 are presented for four sites in the Amazon Basin. Results indicate a sig-
5 nificant negative bias in retrieved lower-tropospheric CO concentrations. The possible influence of smoke aerosol as a source of retrieval bias is investigated using collocated AERONET AOD measurements at two sites, but does not appear to be significant. Finally, we exploit the MOPITT record to analyze both the mean annual cycle and the interannual variability of CO over the Amazon Basin
10 since 2002.

1 Introduction

Seasonal biomass burning in the Amazon Basin profoundly affects atmospheric composition, both regionally and globally. Amazonian emissions include a wide range of trace gases and aerosols (Andreae et al., 2001, 2012). With respect to climate change, future emissions from the Amazon
15 Basin could play a significant role in determining the trajectory of global temperatures (Gullison et al., 2007). Biomass burning emissions in Amazonia are the result of deforestation practices (Malhi et al., 2008) and understory fires, i.e., accidental fires that spread into intact rainforest (Cochrane, 2003). Accurately quantifying emissions from Amazonia from all sources will be increasingly important

as policies are enacted to reduce emissions of greenhouse gases associated with deforestation and
20   degradation (Miles and Kapos, 2008; Aragão and Shimabukuro, 2010).

Coupled with satellite observations of active fires, burned-area estimates are used as the basis
of published "bottom-up" biomass burning emissions inventories, such as the Global Fire Emissions
Database (GFED) (Giglio et al., 2013) and the Fire INventory from NCAR (FINN) (Wiedinmyer et al.,
2011). Such methods for estimating emissions of biomass burning products (i.e., surface fluxes of
25   individual species) are characterized by large uncertainties. Alternatively, emissions of some species
may be quantified using atmospheric observations in "top-down" approaches. To account for the
chemistry and dynamics that affect trace gas concentrations after their production, inverse model-
ing methods are exploited (Arellano et al., 2006; Fortems-Cheiney et al., 2011; Hooghiemstra et al.,
2012). Satellite observations are often employed in inverse modeling because of their unique abil-
30   ity to monitor atmospheric composition over large regions with relatively high (weekly or better)
sampling frequency and spatial density.

However, the optimal use of satellite retrievals for trace-gas concentrations in inverse modeling
systems requires that (1) the vertical sensitivity of the retrieved profiles and the inclusion of a priori
information be properly represented (Kopacz et al., 2010) and (2) any retrieval biases be compen-
35   sated (Hooghiemstra et al., 2012). The geographical variability of retrieval properties (i.e., vertical
sensitivity and retrieval biases) for some satellite products highlights the importance of thoroughly
analyzing retrieval performance prior to the use of such products in inverse modeling studies.

In the following, we analyze retrievals of carbon monoxide (CO) from the MOPITT ("Measure-
ments of Pollution in the Troposphere") satellite instrument (Drummond et al., 2010; Deeter et al.,
40   2014) to investigate the utility of MOPITT products for estimating emissions from the Amazon
Basin. We first characterize the vertical sensitivity of MOPITT CO retrieval products over Amazo-
nia through an analysis of the MOPITT retrieval averaging kernels. Next, we present new MOPITT
validation results by exploiting a set of in-situ CO profiles measured from aircraft at four sites in
Amazonia between 2010 and 2013. The influence of smoke aerosols on MOPITT retrievals is then
45   studied by comparing MOPITT retrieval biases with ground-based aerosol optical depth measure-
ments from two Amazonian sites in the AERONET network. Finally, we present and interpret the
recent history of CO concentrations over Amazonia (2002-2015) based on the MOPITT record.

2   Satellite Retrieval Properties

2.1   MOPITT Retrieval Products

50   MOPITT is a gas-filter correlation radiometer instrument on the NASA Terra polar-orbiting satellite.
MOPITT observations enable retrievals of tropospheric vertical profiles and total column amounts
of carbon monoxide (CO) based on simultaneous thermal-infrared (TIR) and near-infrared (NIR)
observations (Drummond et al., 2010). The MOPITT instrument has been operating nearly continu-

ously since 2000, providing the longest available satellite record for CO. MOPITT retrieval products

55 have progressively improved as the result of accumulated knowledge regarding the instrument, forward modeling methods, and geophysical variables (Worden et al., 2014). Results presented in this manuscript exploit the MOPITT Version 6 (V6) TIR-only and "multispectral" TIR-NIR products (Deeter et al., 2014). The TIR-NIR product offers the greatest vertical resolution, and particularly the greatest sensitivity to CO in the lower troposphere. However, the improved sensitivity of this

60 product is only achieved in daytime MOPITT observations over land. The TIR-only product offers the highest temporal stability and similar performance in variable observing situations (day and night, land and ocean).

Typically, TIR-based satellite retrieval products (including those from the AIRS, TES, and IASI instruments) exhibit relatively low sensitivity to CO concentrations near the earth's surface except

65 in situations with high thermal contrast (Deeter et al., 2007). Thus, CO molecules emitted at the surface may only become "visible" to TIR-based instruments after they ascend into the free troposphere through vertical mixing and may be advected large distances horizontally. The combined effects of instrumental vertical resolution and atmospheric dynamics thus limit both the spatial and temporal resolution of estimated CO emissions based on inverse modeling. Similar limitations affect the esti-

70 mation of CO_2 emissions using TIR satellite observations (Chevallier et al., 2005). To maximize the sensitivity to near-surface CO, the MOPITT instrument uniquely incorporates both TIR and NIR gas correlation radiometers. For daytime observations over land, the added information provided by the NIR channels can substantially enhance the sensitivity to CO near the surface compared to TIR-only retrievals (Worden et al., 2010). This enhancement is illustrated below in an analysis of the MOPITT

75 averaging kernels.

An example of the difference in TIR-only and TIR-NIR retrievals of surface-level CO concentrations for a single MOPITT overpass of the western Amazon Basin on September 17, 2010 is shown in Fig. 1. The selected date represents a day approximately in the middle of the dry season when fire emissions affect much of the southern Amazon Basin. Maps of the CO a priori, V6 TIR-only, and V6

80 TIR-NIR products are presented along with a map of gridded MODIS/Aqua fire counts for the eightday period from Sept. 15 to Sept 22, 2010. Fires during this period were observed in southwestern Brazil and north-central Bolivia. As described in the following section, the a priori represents the background (or default) CO concentration for the retrieval algorithm. The a priori map indicates the highest surface-level CO concentrations in northern Bolivia, with CO concentrations decreasing to-

85 wards the north, west, and south. V6 TIR-only surface-level retrievals exhibit a pattern very similar to the a priori, but with somewhat lower CO concentrations in Peru and southwestern Bolivia, and somewhat higher CO concentrations in Brazil. In contrast, the map of V6 TIR-NIR surface-level retrievals reveal a visibly different pattern compared to the a priori, with the region of highest CO concentrations including much of western Brazil.

90 2.2 Averaging Kernel Variability

The MOPITT retrieval algorithm relies on optimal estimation (Rodgers, 2000; Pan et al., 1998; Deeter et al., 2003, 2014). Retrieved CO volume mixing ratio (VMR) profiles are reported on a ten-level pressure grid; the retrieval for each level indicates the mean VMR in the layer immediately above that level (Deeter et al., 2013). For each retrieved profile resulting from a single MOPITT

95 observation, the retrieval averaging kernel matrix A is produced simultaneously and is provided as a diagnostic in the MOPITT Level 2 and Level 3 data files. A characterizes the sensitivity of the retrieved profile to both the true profile x_{true} and a priori profile x_a through the relation

$$x_{rtv} = x_a + \mathbf{A}(x_{true} - x_a) \tag{1}$$

In the V6 retrieval algorithm, x_a profiles vary seasonally and geographically according to a multi-year model-based CO climatology (Lamarque et al., 2012; Deeter et al., 2014). Each row of A cor-

100 responds to one level in the retrieved profile whereas each column of A corresponds to one level in the "true" CO profile. Each element of A describes the sensitivity of the retrieved CO concentration at one level (expressed as the logarithm of the volume mixing ratio) to the CO concentration at one level in the true profile. A depends on the weighting function matrix K, a priori covariance matrix C_a and observation error covariance matrix C_e according to the relation (Rodgers, 2000)

$$A = (K^T C_e^{-1} K + C_a^{-1})^{-1} K^T C_e^{-1} K \tag{2}$$

105 Elements of K (also known as the "Jacobian" matrix) describe the vertical sensitivity of the measured radiances to applied perturbations to individual levels in the CO vertical profile. As implied by Eq. 2, the features of K largely dictate the features of A. MOPITT averaging kernels are strongly sensitive to both instrumental variables and geophysical parameters (Worden et al., 2013; Deeter et al., 2015).

110 Analysis of the averaging kernels enables an understanding of the vertical resolution and information content of the MOPITT retrieved profiles. The width of each averaging kernel provides a measure of the vertical resolution associated with a specific level of the retrieved profile whereas the area under the averaging kernel (i.e., the sum of the elements) indicates the integrated sensitivity of the retrieval to the true profile (Rodgers, 2000). Averaging kernels characterized by a small

115 area (e.g., much less than one) indicate a retrieval level heavily weighted by the a priori. The trace of A (i.e., the sum of the diagonal elements) defines the "Degrees of Freedom for Signal" or DFS and is often interpreted as the number of independent pieces of information in the retrieval from the measurement. Properties of the averaging kernel for the retrieved surface-level CO concentration are particularly important since the primary sources of CO are located at the surface, and tropospheric

120 concentrations of CO often peak at or near the surface.

2.3 Averaging Kernel Comparisons

Mean MOPITT averaging kernels for a region of the western Amazon Basin between 10°S, 5°S, 70°W, and 65°W for a daytime overpass of the MOPITT instrument on September 17, 2010 are shown in Fig. 2. Mean averaging kernels for the TIR-only product and the TIR-NIR product are both presented. MOPITT averaging kernels for another daytime overpass of the same region on July 1, 2010 are presented in Fig. 3. Whereas the date of the first overpass represents a period in the middle of the Amazon dry season, characterized by high CO values across most of the Amazon Basin, the date of the second overpass corresponds to the beginning of the dry season when CO concentrations are near the minimum of the annual cycle (Edwards et al., 2006). For the selected region, mean retrieved surface-level CO concentrations on September 17 for the TIR-only and TIR-NIR products were 431 and 512 ppbv, respectively. Corresponding values for July 1 were 144 and 146 ppbv.

Mean averaging kernels for the TIR-only product depicted in the top panel of Fig. 2 are considered first. For the selected scene, the mean TIR-only surface-level averaging kernel peaks at 800 hPa and decreases sharply at the surface, revealing that the TIR-only surface-level retrievals exhibit poor sensitivity to CO at the surface. Also, the strong similarity in the shape of the surface-level and 800 hPa averaging kernels illustrates the lack of independent information in the retrieved profile at these two levels. In contrast, for the TIR-NIR averaging kernels shown in the bottom panel of Fig. 2, the mean surface-level averaging kernel peaks at the surface and rapidly decreases at higher levels. Based on the full-width half-maximum of this averaging kernel, the sensitivity of the TIR-NIR surface-level retrieval is dominated by the layer between the surface and about 800 hPa. Moreover, it is clear from the TIR-NIR averaging kernels that the retrieved concentrations at the surface and at 800 hPa contain independent information with respect to the CO profile. This enhancement in the retrieval information content is also consistent with the higher mean DFS value observed for the TIR-NIR retrievals (2.15) compared to the TIR-only retrievals (1.53).

Differences in the mean retrieval averaging kernels for July 1 and September 17 are expected as the result of the large difference in mean CO concentrations over the Amazon Basin for those two dates (Deeter et al., 2015). Increasing CO concentrations tend to strengthen the weighting functions for both the TIR and NIR channels, which affects characteristics of the averaging kernels. For the TIR-only product, this effect most severely impacts the 800 hPa retrieval level. Both the width and peak value of the 800 hPa averaging kernel are significantly degraded on July 1. Comparing Figures 2 and 3, the TIR-NIR surface-level averaging kernel on July 1 peaks at a higher altitude and is substantially broader than for September 17. For characterizing CO concentrations in the lower troposphere, these results demonstrate that the MOPITT TIR-NIR product yields more information as CO concentrations increase.

3   Validation

Retrieval validation involves the rigorous analysis of differences between retrieval products and independent measurements with well characterized uncertainties. MOPITT retrievals have been validated with a variety of in-situ datasets from which CO vertical profiles have been produced (Emmons et al., 2009; Deeter et al., 2014). For validating optimal estimation-based retrievals, the standard measure of retrieval error excludes the known smoothing effect of the retrieval process (as represented by the averaging kernel matrix) and the dependence on the a priori profile. Thus, rather than direct comparisons of retrieval products with in-situ data, validation involves comparisons of retrieved profiles with simulated profiles calculated according to Eq. 1. For such comparisons, A and x_a are extracted from the MOPITT Level 2 data files and then applied to the x_{true} profile which is based on the in-situ data.

Because of the variety of sources of retrieval error, and the geographical and temporal variability of such errors, validation of the MOPITT CO product is an ongoing activity. Below, we present validation results for MOPITT retrieval products for the Amazon Basin for the first time. At least two potential challenges for MOPITT retrievals occur in this region during the biomass burning season. First, extreme CO concentrations could conceivably exceed the valid range of CO concentrations for the MOPITT radiative transfer model. This was found to be an issue for the MOPITT Version 3 product which was addressed in the development of the Version 4 product (Deeter et al., 2010). Second, the effects of highly concentrated smoke aerosols could also degrade the quality of MOPITT retrievals. This topic is addressed in Section 3.3.

3.1   Review of Version 6 Validation Results

Validation results for the MOPITT V6 product were previously reported in Deeter et al. (2014). Those results were based on in-situ profiles acquired during the HIPPO ("HIAPER Pole to Pole Observations") field campaign (2009-2011) and through a long-term NOAA monitoring program mostly over North America. One significant finding from the HIPPO validation results was the apparent latitude dependence of observed retrieval biases in the V6 TIR-only product. For example, for the 800 hPa retrieval level, there appeared to be a significant negative bias in the Tropics which was absent in midlatitude and polar regions. The underlying cause of the observed latitude dependence of the bias has not been explained. Moreover, since nearly all HIPPO flights were over the open ocean, it is unclear if the observed latitude dependence would be expected over land scenes as well as ocean scenes.

3.2   Aircraft-based In-situ CO Measurements in the Amazon

Below, we exploit a set of in-situ CO vertical profiles derived from a trace-gas sampling program over the Amazon Basin that began in 2010 (Gatti et al., 2014). Profiles are based on air samples acquired

approximately bi-weekly from aircraft at four sites: Alta Floresta (ALF, 9°S, 57°W), Rio Branco
190 (RBA, 9°S, 68°W), Tabatinga (TAB, 6°S, 60°W), and Santarém (SAN, 3°S, 55°W). The primary
objective of the program is to observe and analyze the state, changes, and climate sensitivity of the
Amazon carbon pools. In addition to CO, concentrations of CO_2, CH_4, N_2O, and SF_6 are also mea-
sured for each air sample. Precision and accuracy of the in-situ CO dry-air mole fraction ("DMF")
measurements are reported to be less than 1 ppb (http://www.esrl.noaa.gov/gmd/ccgg/aircraft/qc.html).
195 The wide geographical distribution of the four sites permits the compositional analysis of airmasses
as they first enter the Amazon Basin and as they become progressively affected by regional biomass
burning emissions.

For most flights, the maximum sampling altitude attained by the aircraft flying over the Amazon
Basin was approximately 4.4 km, corresponding to a pressure of about 580 hPa. This altitude is
200 substantially less than the altitude required to produce a complete ten-level vertical profile (from
the surface to 100 hPa) suitable for MOPITT validation. Aircraft measurements of CO DMF were
transformed into MOPITT VMR "validation profiles" in a process requiring several steps and two
distinct pressure grids. First, in-situ data for each profile were interpolated onto the standard 35-level
fine grid used internally in the MOPITT retrieval algorithm (Edwards et al., 1999). Since this step is
205 limited to grid levels only up to 600 hPa, the interpolated DMF value at 600 hPa was extended ver-
tically up to 250 hPa. In-situ DMF values were then converted to moist-air VMR values using water
vapor mixing ratios included in the CAM-chem model-based climatology (Lamarque et al., 2012).
Maximum differences between CO DMF and VMR values were about 3%; differences generally
peak at the surface. For levels in the fine grid at and above 100 hPa (the approximate altitude of the
210 tropical tropopause), CO VMR values were taken from the CAM-chem climatology. VMR values
for grid between 250 and 100 hPa were obtained by interpolation. Finally, resulting fine-grid VMR
validation profiles were regridded into coarse ten-level validation profiles (for consistency with the
actual MOPITT retrieval grid) by averaging the fine-grid VMR values in the layers immediately
above the corresponding levels in the retrieval grid (Deeter et al., 2013).

215 The lack of in-situ data above 580 hPa precludes the use of these profiles for validating MOPITT
retrievals of upper-tropospheric CO concentrations. However, inspection of the MOPITT averaging
kernels in Figures 2 and 3 indicates that CO retrievals for the lower troposphere are only weakly
sensitive to the actual CO concentrations at 500 hPa and above. For example, for the TIR-only mean
averaging kernel for the 800 hPa retrieval level on September 17, 2010, we find that 80% of the
220 kernel area corresponds to levels within the in-situ measured altitude range from the surface to 600
hPa. For the 600 hPa retrieval level, 66% of the kernel area is associated with levels within the
measured altitude range. This diagnostic decreases to less than 50% for higher retrieval levels. Thus,
the lack of in-situ data above 4.4 km suggests that validation results will be most meaningful for
retrieval levels from the surface to 600 hPa.

3.2.1 V6 TIR-only

Comparisons of MOPITT TIR-only retrieved CO concentrations with simulated retrievals based on in-situ data from the four Amazonian sites are presented in Fig. 4. Results for the surface, 800 hPa, and 600 hPa retrieval levels are shown separately. Corresponding comparisons for the MOPITT TIR-NIR product are shown in Fig. 5. Each plotted point indicates (1) the mean MOPITT retrieved value for all daytime observations within 200 km of the site of the in-situ data and within 24 hours of the in-situ sampling flight and (2) the mean simulated value calculated after applying Eq. 1 to the in-situ profile. Vertical error bars indicate the variability (standard deviation) of the MOPITT data used to calculate each of the plotted mean values. In comparison, uncertainties in the in-situ DMF measurements (less than 1 ppb) are insignificant. Summary statistics (bias, standard deviation, and correlation coefficient) are listed on each panel and in Table 2.

Mean TIR-only biases at the surface, 800 hPa, and 600 hPa retrieval levels are -9.8%, -16%, and -13%, respectively, indicating that MOPITT TIR retrievals underestimate surface and lower-tropospheric CO concentrations. For the HIPPO validation results, observed mean TIR-only biases for measurements acquired between 30°S and the Equator for the surface, 800 hPa, and 600 hPa retrieval levels were -1%, -2%, and -2%, respectively. However, the HIPPO validation results were based on relatively few in-situ profiles in the same latitude range as the Amazonian profiles (i.e., between 20°S and the Equator), possibly because prevailing cloudiness near the ITCZ (Intertropical Convergence Zone) resulted in fewer MOPITT retrievals. Statistically, therefore, the HIPPO results for this latitude range might not be as robust as for other regions. In the latitude range between the Equator and 30°N, HIPPO mean biases for the surface, 800 hPa, and 600 hPa retrieval levels were -6%, -11%, and -12%, respectively. Thus, relative to the HIPPO results, observed mean TIR-only retrieval biases for the Amazon appear to be more consistent with the observed biases in the northern Tropics than the southern Tropics.

As shown in Fig. 4, surface-level in-situ CO concentrations (smoothed with the TIR-only averaging kernels) vary from about 100 ppbv to over 500 ppbv. In previous MOPITT validation papers (e.g., (Deeter et al., 2014)), the highest CO concentrations included in the in-situ profiles were less than 400 ppbv. Inspection of Fig. 4 indicates that the retrieval biases do not visibly increase at the upper range of CO concentrations. For example, in the results for the surface and 800 hPa, the best-fit line appears to fit the data equally well at low and high CO concentrations. Thus, for the range of CO concentrations presented by the Amazonian profiles, the quality of MOPITT TIR-only retrievals appears to be unaffected by high CO concentrations.

The correlation coefficients for the three retrieval levels decrease with increasing altitude, from 0.94 at the surface to 0.82 at 600 hPa. This trend likely reflects (1) the increasing role of errors associated with the lack of in-situ data above 4.4 km and (2) the stronger a priori weighting for the surface-level retrieval, as indicated by the listed mean kernel area values.

3.2.2 V6 TIR-NIR

As indicated in Fig. 5, mean TIR-NIR biases at the surface, 800 hPa, and 600 hPa retrieval levels are -4.6%, -27%, and -25%, respectively. Thus, compared to the TIR-only results, the negative bias at the surface is smaller while the negative biases at 800 and 600 hPa are substantially larger. Like the TIR-only results, the least-squares best fit lines appear to fit the data equally well at low and high CO concentrations. Thus, the quality of MOPITT TIR-NIR retrievals also appears to be unaffected by high CO concentrations.

As indicated by both the residual standard deviation values and correlation coefficients listed in Figures 4 and 5 (and in Table 1), errors for the TIR-NIR product generally appear to be larger than for the TIR-only product. This effect is most evident at the surface, where the standard deviation is 17% for the TIR-NIR product compared to 8% for the TIR-only product. To some extent, larger biases and variable retrieval errors are evident in the TIR-NIR product due to the use of a "gain enhancement factor" to amplify the influence of the NIR radiances in the TIR-NIR product (Deeter et al., 2011). This strategy magnifies retrieval errors due to errors in both the TIR and NIR radiances. However, because of the greater CO sensitivity of the TIR-NIR retrievals (as indicated by the larger mean kernel area A_Σ), larger standard deviations would also be expected due to potential CO horizontal gradients within the 200 km radius used to match MOPITT retrievals with the in-situ profiles. Therefore, the larger standard deviation associated with the TIR-NIR product does not by itself necessarily indicate lower retrieval quality.

3.2.3 Effects of limited aircraft altitude range

The set of aircraft in-situ profiles used for MOPITT validation include a small subset for which the CO VMR in the extrapolated section of the validation profiles (between 600 and 250 hPa) was at least 120 ppbv. While even higher CO concentrations have recently been observed in the Amazonian upper troposphere during the dry season (Wendisch et al., 2016), it is plausible that the occurrence of biomass burning plumes at altitudes near 580 hPa (i.e., the altitude of the highest in-situ sample) might yield some validation profiles containing overestimated CO concentrations in the upper troposphere. A simple sensitivity experiment was therefore performed to recalculate the validation statistics after discarding all profiles where the extrapolated mixing ratio was greater than 120 ppbv. Overall, this eliminated about one-sixth of the profiles. For both the TIR-only and TIR-NIR products, the maximum resulting change in the overall retrieval bias was 3%. Thus, the MOPITT validation results for the surface, 800 hPa, and 600 hPa are not strongly affected by the inclusion of profiles with extrapolated VMR values above 120 ppbv. This finding is consistent with the properties of the MOPITT averaging kernels and provides further evidence that the lack of in-situ data above 4.4 km does not severely affect the validation of the MOPITT retrieval levels in the lower troposphere.

3.3 Comparisons with AERONET AOD measurements

The radiative transfer model on which the MOPITT retrieval algorithm is based neglects the potential effects of aerosols (Edwards et al., 1999). While aerosols often produce significant radiative effects in the ultraviolet and visible spectral regions, aerosol optical depth (AOD) diminishes rapidly with increasing wavelength (Eck et al., 1999). (MOPITT's NIR channels operate in the 2.3 μm band, while MOPITT's TIR channels operate in the 4.7 μm band.) Thus, in most atmospheric conditions, aerosols probably have a negligible effect on MOPITT retrieval error. However, MOPITT validation results have never previously been reported for regions strongly affected by biomass burning emissions, where AODs of one or greater are often observed (Eck et al., 1999).

For MOPITT CO retrievals, dense smoke aerosols produced by biomass burning could conceivably cause retrieval errors through at least two mechanisms. For MOPITT's NIR channels, scattering from aerosol particles could potentially alter the effective optical path length through the atmosphere. This effect is explicitly represented in retrieval algorithms for carbon dioxide and methane using satellite observations in spectral bands near 1.6 and 2.0 μm (Butz et al., 2011; Yoshida et al., 2013). Alternatively, extreme aerosol concentrations might locally affect meteorological conditions, either by reducing insolation at the surface or through atmospheric heating caused by absorbing aerosols such as black carbon (Ramanathan and Carmichael, 2008). This effect could indirectly result in retrieval error for both the TIR-only and TIR-NIR retrievals if it caused a significant error in the assumed atmospheric temperature profile.

Two of the four Amazonian sites for which aircraft CO profiles were exploited for MOPITT validation in Section 3.2 also host AERONET stations (Holben et al., 2001). This coincidence permits the analysis of the dependence of MOPITT retrieval biases on AOD. Possible MOPITT aerosol-dependent retrieval biases were studied by comparing MOPITT retrieval biases at 800 hPa from Section 3.2 with ground-based AOD measurements at 500 nm from the Alta Floresta and Rio Branco AERONET sites. The 800 hPa retrieval level was selected for this comparison because (1) the averaging kernel area for this level typically indicates a weak dependence on the a priori for both the TIR-only and TIR-NIR products and (2) calculated retrieval biases at this level are not strongly affected by the lack of in-situ data for the upper troposphere (as described above). AOD values at 500 nm were extracted from AERONET daily-mean data files for the same dates on which the aircraft in-situ data were acquired.

The dependence of MOPITT retrieval bias at 800 hPa on AOD for both the TIR-only and TIR-NIR products is presented in Fig. 6. Each plotted point indicates the mean MOPITT retrieval bias and daily-mean AOD for a single daytime MOPITT overpass of one of the two AERONET sites. While there are relatively few data points indicating high AOD values, the plotted data do not appear to demonstrate a clear AOD dependence for the retrieval bias for either type of MOPITT product. To analyze the relationship further, mean and standard deviation retrieval bias values were calculated separately for data subsets where AOD < 0.5 (a total of 28 overpasses) and AOD >= 0.5 (nine

overpasses). Presumably, if aerosols did produce a systematic retrieval bias, it would be revealed by comparing the means and standard deviations for these two subsets. For the TIR-only data, the overall bias for the low-AOD subset was -15.9 ± 11.5% and for the high-AOD subset was -18.4

335 ± 25.0%. For the TIR-NIR data, the overall bias for the low-AOD subset was -28.7 ± 18.2% and for the high-AOD subset was -26.4 ± 26.1%. Thus, a systematic retrieval bias related to aerosols is not indicated in either the MOPITT TIR-only or TIR-NIR products. The greater standard deviations observed for the AOD >= 0.5 subsets might be explained by stronger CO geographical and temporal variability during the Amazonian dry season, however this is only a hypothesis.

340 ## 4   Observed CO Variability over the Amazon Basin

Significant interannual variability in CO concentrations over the Amazon Basin primarily results from two sources: deforestation and understory fires. Annual deforestation rates in the "Legal Amazon" region estimated by Brazil's PRODES (Program to Calculate Deforestation in the Amazon) satellite-based monitoring system (www.obt.inpe.br/prodes/index.php) plummeted from 27,772 km^2/yr

345 in 2004 to 4,571 km^2/yr in 2012. Suggested causes of falling deforestation rates in Brazil include (1) the expansion of protected areas in Brazil, (2) increased monitoring and law enforcement and (3) market forces including declining soy prices and economic disincentives for landholders implicated in deforestation (Nepstad et al., 2009; Macedo et al., 2012). While much of the biomass burning activity in Amazonia relates directly to land use practices, human-initiated fires often escape from

350 deforested areas into neighboring standing forests (Cochrane, 2003; Morton et al., 2013). These fires typically spread slowly in the forest understory, mainly consuming leaf litter, yet cause substantial long-term damage to the tropical forest ecosystem (Peres, 1999; Cochrane, 1999). Regionally, areas burned in understory fires may be comparable to or larger than clear-cut areas (Morton et al., 2013; Alencar et al., 2006; Aragão et al., 2007). Areas burned in understory fires are particularly extensive

355 during droughts, indicating some degree of climate influence (Alencar et al., 2006).

The MOPITT data record enables the analysis of evolving CO concentrations over the Amazon Basin. In the following, we examine the CO record based on the MOPITT V6 TIR-NIR Level 3 monthly-mean product. MOPITT Level 3 products are gridded at one-degree resolution (latitude and longitude) and are gridded separately for daytime and nighttime overpasses; the following analysis

360 only exploits daytime Level 3 data. For analyzing CO variability over a relatively large region and over multiple years, analyzing Level 3 monthly-mean products is much more efficient than for Level 2 data. MOPITT records of basin-averaged CO total column are analyzed in addition to retrieved CO concentrations at the surface and at 400 hPa. While no new validation results are presented in this manuscript for 400 hPa, TIR-NIR averaging kernels for this level (shown in Figs. 2 and 3)

365 demonstrate its usefulness as a retrieval of upper-tropospheric CO concentrations with very weak sensitivity to CO in the lower troposphere.

In the following analysis, no attempt was made to correct the MOPITT data for retrieval bias or bias drift. For interpreting the mean CO annual cycle, validation results presented above imply that TIR-NIR long-term monthly means shown in Section 4.1 likely underestimate both true CO

370   concentrations at the surface and CO total column values. For the upper troposphere, the retrieval bias over the Amazon Basin is not well characterized, although HIPPO TIR-NIR validation results for the Tropics indicate biases at 400 hPa between -10 and 0%. Temporally fixed retrieval biases would not necessarily affect the interpretation of interannual variability presented in Section 4.2, although the bias long-term trend (or "bias drift") should be considered in that case. Previous TIR-

375   NIR validation results based on CO vertical profiles measured over North America (Deeter et al., 2014) indicated a bias drift for the 400 hPa level of 1.08%/yr, a bias drift for the surface of -0.48%/yr, but only a very weak bias drift for CO total column (0.003×10^{18} mol/cm^2/yr). However, the geographical variability of bias drift has not yet been investigated.

4.1 The Annual Cycle: Long-term Monthly Means

380   Long-term monthly means were calculated by separately averaging Level 3 (gridded) MOPITT CO data for each month over the period from 2002 to 2015 and are presented in Fig. 7. (MOPITT data from 2000, 2001 and 2009 were excluded from these long-term averages because of instrumental anomalies that resulted in incomplete years of observations.) Geographically, CO data were averaged for all grid cells within the digitized boundaries of the Amazon Basin as defined for the "Large-Scale

385   Biosphere-Atmosphere Experiment in Amazonia" field campaign (https://daac.ornl.gov/LBA/lba.shtml). Grid cells with no data (due to persistent cloudiness) were excluded from the long-term monthly-mean basin averages. Thus, during the Amazonian wet season from January to May, the presented monthly-mean basin averages may reflect CO concentrations only over some fraction of the entire Amazon Basin. Separate panels in Fig. 7 indicate MOPITT long-term monthly means for CO to-

390   tal column, 400 hPa CO, and surface-level CO concentrations. The interannual variability (standard deviation) of the monthly-means used to calculate the long-term monthly means is indicated by the red shaded area. Mean a priori values were also calculated from the Level 3 data files and are shown as dashed lines. V6 a priori CO concentrations are derived from a model-based climatology for the years 2000-2009.

395   In all three panels, long-term mean CO concentrations appear generally stable from January to June. As the dry season develops, mean CO concentrations then increase along with the CO interannual variability (as indicated by the width of the red shaded area). At the surface, mean CO concentrations begin to increase in June, followed by a larger increase in July. Mean surface-level CO concentrations peak in September, for which the long-term monthly mean is approximately three

400   times the mean for the wet season months. Mean CO concentrations at the surface follow the annual cycle indicated by the the a priori, but are generally smaller by 10-20%. (This difference would be somewhat smaller if we accounted for the observed retrieval bias of -4.6% found in Section 3.2.) At 400

hPa, increased CO concentrations are generally not observed until August and mean concentrations peak in October. The delayed peak in CO concentrations at 400 hPa (compared to the surface) likely reflects the characteristic time required for CO produced at the surface to ascend into the upper troposphere. Observed mean CO concentrations at 400 hPa exhibit a stronger annual cycle compared to the a priori, and are about 20% smaller during the wet season months.

4.2 Interannual Variability

CO monthly anomalies for the Amazon Basin were calculated by subtracting the long-term monthly means (shown in Fig. 7) from the monthly means for each individual year of MOPITT observations. The resulting anomaly timeseries for CO total column, CO VMR at 400 hPa and surface-level VMR are shown in Fig. 8. Monthly-mean anomalies during the period from January to June exhibit little interannual variability for CO total column and at 400 hPa, but exhibit significant interannual variability for surface-level CO. However, for this period, the apparent interannual variability at the surface is qualitatively consistent with the expected result of bias drift as described above; i.e., monthly-mean anomalies exhibit an overall decreasing trend. Thus, the apparent interannual variability of surface-level CO for the months between January and June might not be significant.

The three years indicating the largest positive anomalies in Fig. 8 are 2005, 2007, and 2010. These years correspond to years when exceptionally large areas burned in Amazonian understory fires (Morton et al., 2013). Extensive droughts were recorded in the Amazon Basin in both 2005 and 2010 (Lewis et al., 2011). Factors which contributed to the relatively large CO emissions in 2007 were recently analyzed in Bloom et al. (2015). The apparent association of large CO anomalies with understory fires illustrates the importance of climate-driven fires to the Amazonian carbon budget. Efforts to reduce deforestation in the Amazon Basin may not affect future emissions from understory fires (Morton et al., 2013).

Significant negative dry-season monthly anomalies are apparent for 2011, 2013, and 2014. Excluding the years 2005, 2007, and 2010, the history of monthly anomalies presented in Fig. 8 generally indicates an overall long-term trend toward decreasing CO emissions from the Amazon Basin since 2002. This trend might be the result of falling deforestation rates (Nepstad et al., 2009) or may be related to climate. However, this trend does not seem to hold for the most recent year of the analysis. While monthly-mean anomalies for 2015 are relatively weak in the early dry season months, exceptionally strong positive anomalies for 2015 are evident in November and December.

5 Conclusions

Biomass burning emissions inventories such as GFED and FINN are widely used in modeling simulations, despite their large uncertainties. Satellite observations of CO might be useful for refining such inventories. This work demonstrates the utility of MOPITT products for analyzing CO emis-

sions from the Amazon Basin. Comparisons of retrieval averaging kernels and values of Degrees of
Freedom for Signal for MOPITT TIR-only and TIR-NIR products indicate enhanced sensitivity to
CO in the lower troposphere for the TIR-NIR product, particularly during the Amazonian dry sea-

440   son. Validation results based on routine sampling flights at four sites in the Amazon Basin exhibit a
negative bias in MOPITT V6 retrievals of CO in the lower troposphere, but do not indicate degraded
performance in high-CO scenes. Likewise, comparisons with ground-based AOD measurements do
not reveal retrieval biases associated with dense smoke from biomass burning. Over the MOPITT
mission, observed long-term monthly mean surface-level CO concentrations averaged over the Ama-

445   zon Basin are generally similar to the CO climatology used as the retrieval a priori, whereas observed
long-term mean CO concentrations at 400 hPa exhibit a much stronger annual cycle. A brief anal-
ysis of the history of monthly-mean anomalies for the Amazon Basin indicates that the largest CO
emissions occurred in years when drought-driven understory fires burned exceptionally large areas.

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data used for this research are available at the NASA Atmospheric Sciences Data Center (ASDC) datapool
at https://eosweb.larc.nasa.gov/HPDOCS/datapool/ . We thank Louis Giglio for making the MODIS fire count

455   data available. MODIS Collection 5 fire count data were obtained from http://modis-fire.umd.edu.

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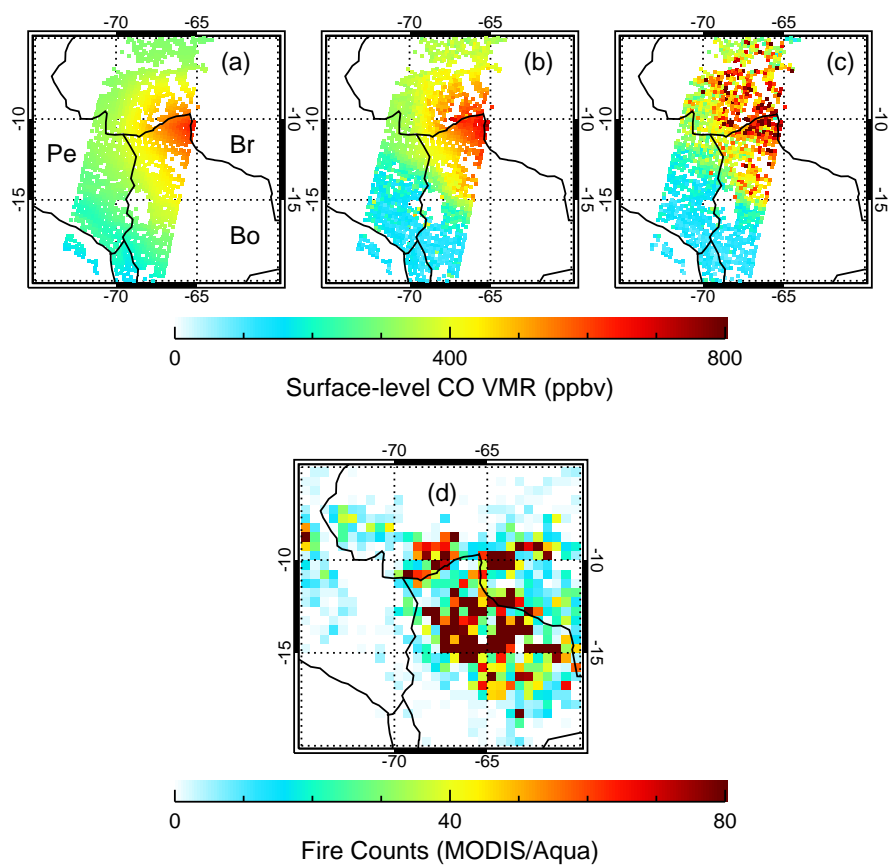
Figure 1. Comparison of (a) climatological MOPITT surface-level CO a priori concentrations, (b) V6 TIR-only retrieved surface-level CO, and (c) V6 TIR-NIR retrieved surface-level CO for a daytime MOPITT overpass of the western Amazon Basin on September 17, 2010. Country abbreviations in panel (a) include 'Pe' for Peru, 'Br' for Brazil and 'Bo' for Bolivia. Gridded fire counts based on MODIS/Aqua observations for the 8-day period from Sept. 15 to 22, 2010, are shown in panel (d). The map of V6 TIR-NIR surface-level retrievals reveal a region of high CO concentrations over much of western Brazil, whereas the V6 TIR-only retrievals tend more towards the a priori.
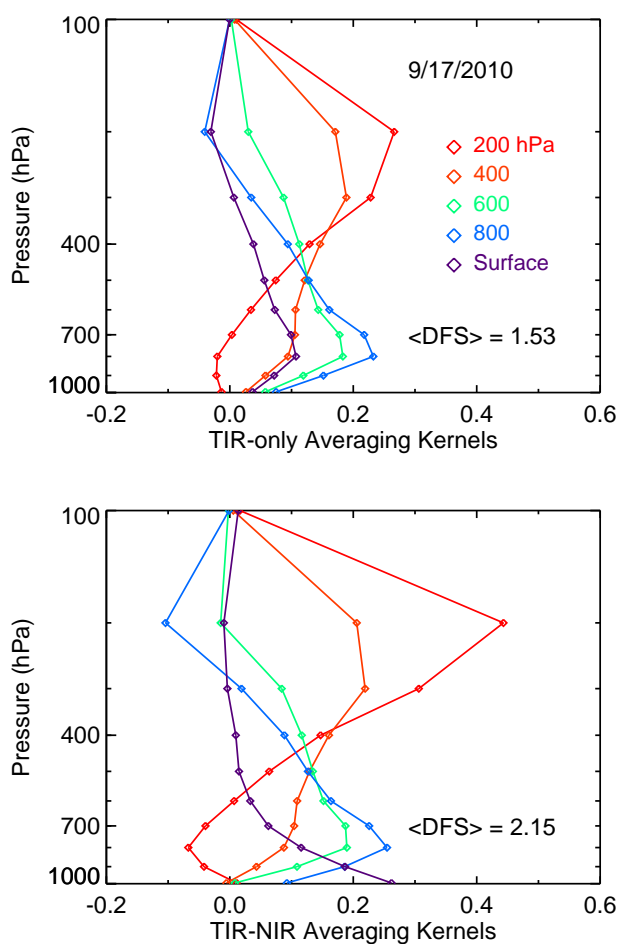
Figure 2. Comparison of mean retrieval averaging kernels for the MOPITT Version 6 TIR-only (top panel) and
TIR-NIR (bottom panel) products for the western Amazon Basin (between 10°S, 5°S, 70°W, and 65°W) on
September 17, 2010. For clarity, only the averaging kernels for alternating levels in the MOPITT retrieval grid
(i.e., surface, 800 hPa, 600 hPa, etc.) are actually plotted.
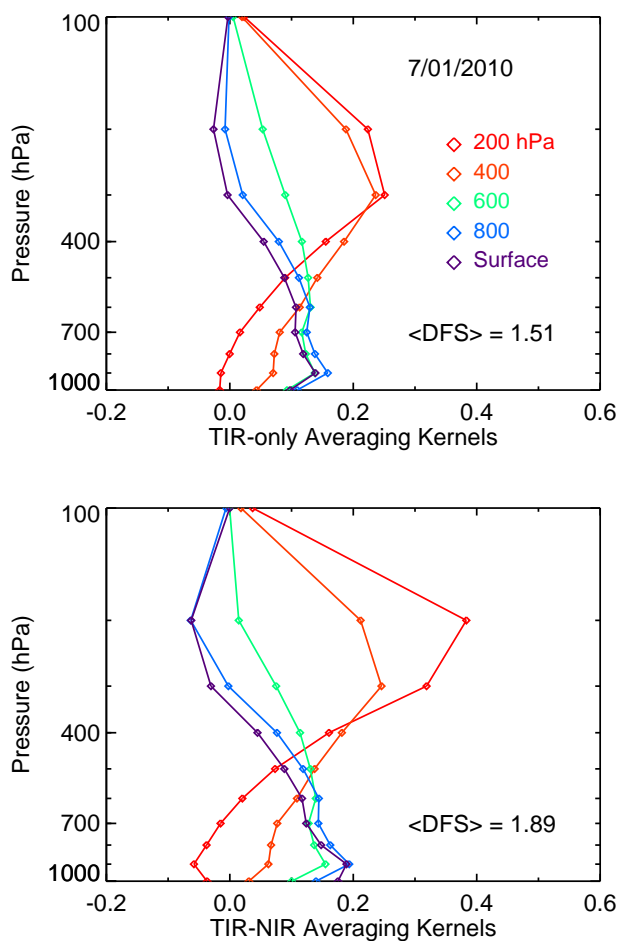
Figure 3. Comparison of mean retrieval averaging kernels for the MOPITT Version 6 TIR-only (top panel) and TIR-NIR (bottom panel) products for the western Amazon Basin on July 1, 2010.
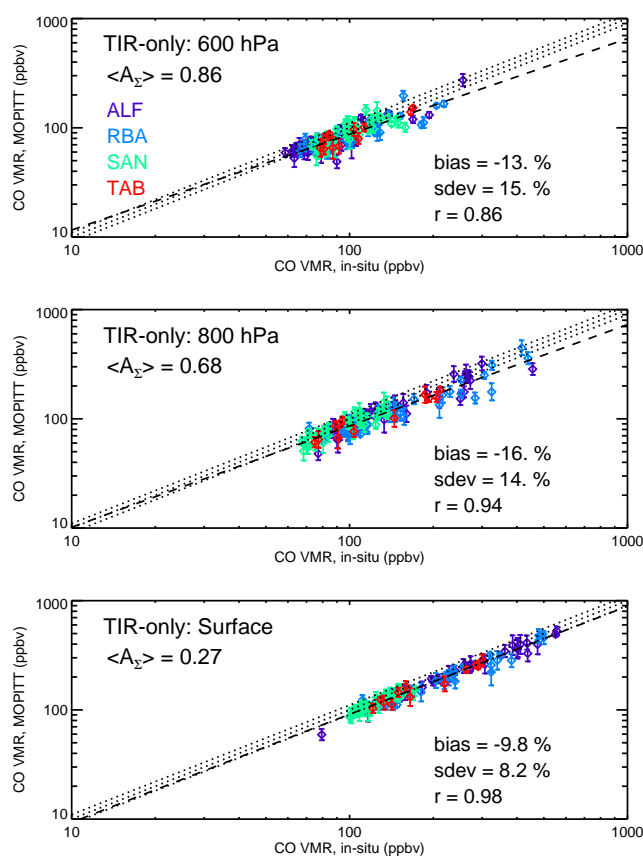
Figure 4. MOPITT Version 6 TIR-only validation results for MOPITT mid- and lower-tropospheric CO re-
trievals based on aircraft profiles acquired at four Amazonian sites: Alta Floresta (ALF), Rio Branco (RBA),
Tabatinga (TAB) and Santarém (SAN). Vertical error bars indicate the variability (standard deviation) of the
MOPITT data used to calculate each of the plotted mean values. Each panel shows the least-squares best-fit
line (dashed) as well as boundaries around the ideal one-to-one line corresponding to $\pm$ 10% errors (indicated
by the dotted lines). Summary statistics (bias, standard deviation, and correlation coefficient) are listed on each
panel in addition to the mean kernel area (A_{Σ}); this diagnostic provides a useful index for a priori dependence.
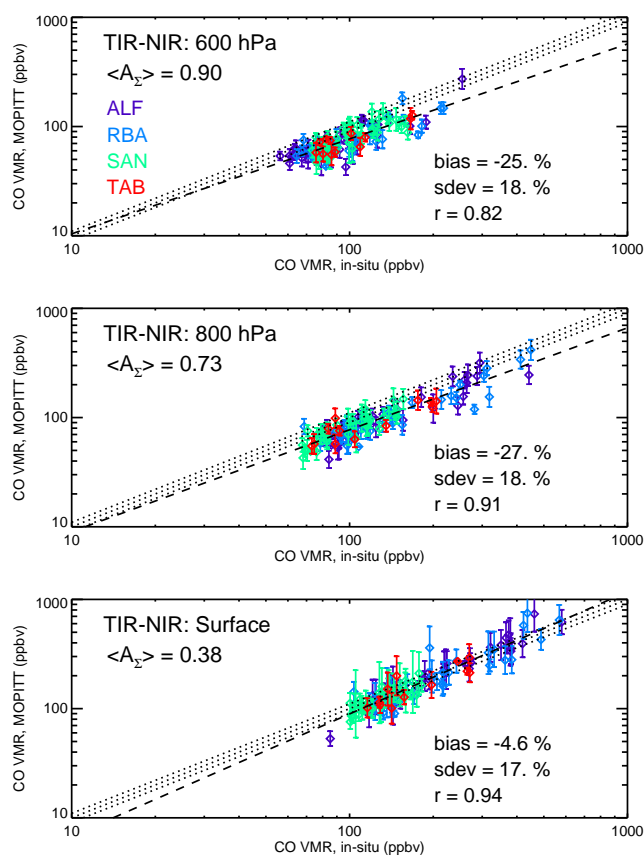
Figure 5. MOPITT Version 6 TIR-NIR validation results for MOPITT mid- and lower-tropospheric CO retrievals based on aircraft profiles acquired at four Amazonian sites. See caption to Fig. 4.
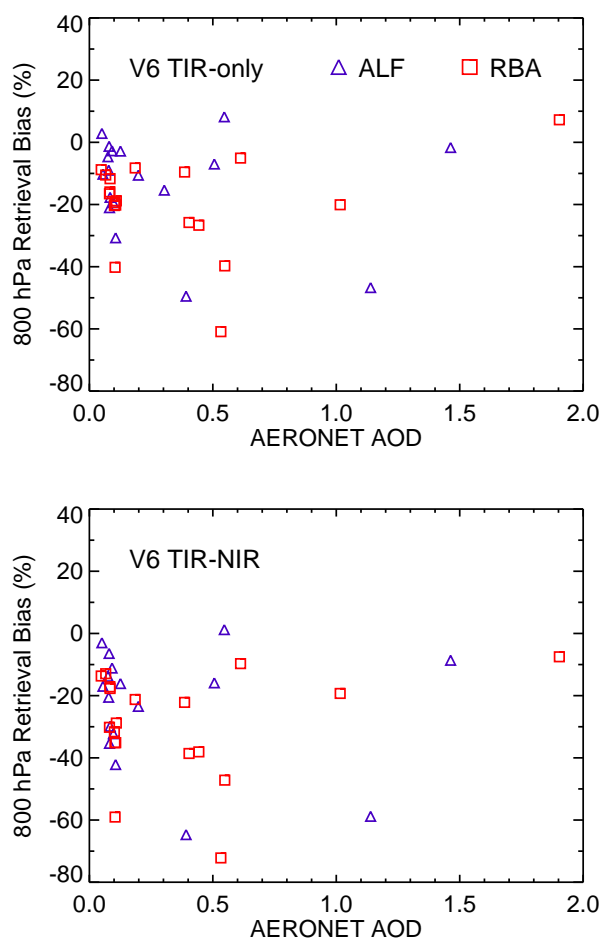
Figure 6. Comparison of observed MOPITT TIR-only and TIR-NIR retrieval biases at 800 hPa at two Amazonian sites with ground-based measurements of aerosol optical depth at 500 nm from the AERONET network. Each plotted point indicates the mean MOPITT retrieval bias and daily-mean AOD for a single daytime MOPITT overpass of one of the two AERONET sites. While there are relatively few data points at high AOD values, the plotted data do not indicate a clear AOD dependence for the retrieval bias for either type of MOPITT product.
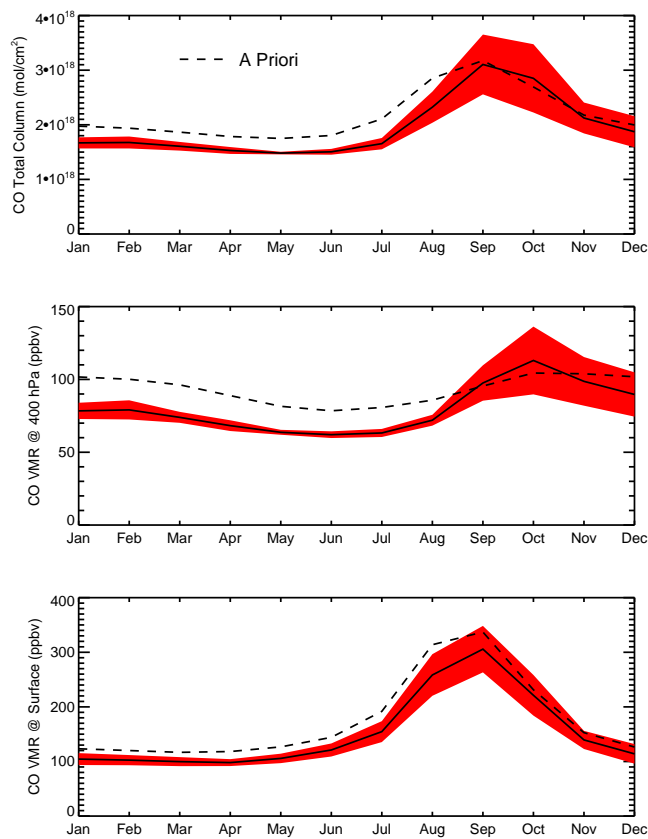
Figure 7. MOPITT TIR-NIR long-term monthly-means of retrieved CO total column and CO concentrations for the Amazon Basin, based on observations from 2002 to 2015. Shaded red area indicates variability (standard deviation) of monthly means. Monthly-mean a priori values are also indicated for comparison.
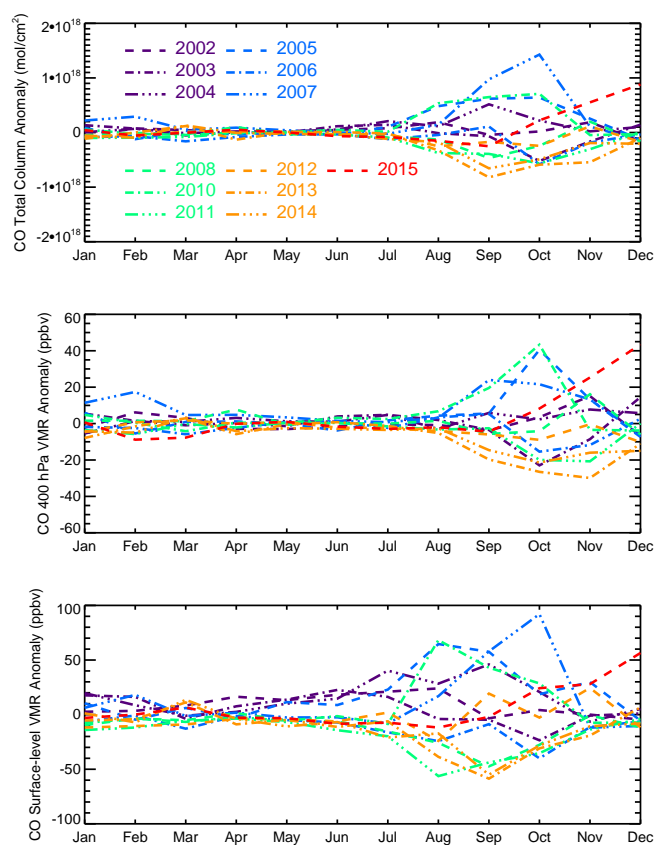
Figure 8. Monthly-mean anomalies of MOPITT TIR-NIR CO products for each of the years used to generate the long-term monthly means shown in Fig. 7.

Table 1. MOPITT validation results based on in-situ CO vertical profiles acquired at four sites in the Amazon Basin from 2010 to 2013.

		Surface	800 hPa	600 hPa
V6T	bias (%)	-9.8	-16.	-13.
	sdev (%)	8.2	14.	15.
	r	0.98	0.94	0.86
V6J	bias (%)	-4.6	-27.	-25.
	sdev (%)	17.	18.	18.
	r	0.94	0.91	0.82