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Validation of Measurements of Pollution in the Troposphere (MOPITT) CO retrievals with aircraft in situ profiles

L. K. Emmons,[1] M. N. Deeter,[1] J. C. Gille,[1] D. P. Edwards,[1] J.-L. Attié,[2] J. Warner,[1]
D. Ziskin,[1] G. Francis,[1] B. Khattatov,[1] V. Yudin,[1] J.-F. Lamarque,[1] S.-P. Ho,[1] D. Mao,[1]
J. S. Chen,[1] J. Drummond,[3] P. Novelli,[4] G. Sachse,[5] M. T. Coffey,[1] J. W. Hannigan,[1]
C. Gerbig,[6] S. Kawakami,[7] Y. Kondo,[8] N. Takegawa,[8] H. Schlager,[9] J. Baehr,[9]
and H. Ziereis[9]

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[1] Validation of the Measurements of Pollution in the Troposphere (MOPITT) retrievals of carbon monoxide (CO) has been performed with a varied set of correlative data. These include in situ observations from a regular program of aircraft observations at five sites ranging from the Arctic to the tropical South Pacific Ocean. Additional in situ profiles are available from several short-term research campaigns situated over North and South America, Africa, and the North and South Pacific Oceans. These correlative measurements are a crucial component of the validation of the retrieved CO profiles and columns from MOPITT. The current validation results indicate good quantitative agreement between MOPITT and in situ profiles, with an average bias less than 20 ppbv at all levels. Comparisons with measurements that were timed to sample profiles coincident with MOPITT overpasses show much less variability in the biases than those made by various groups as part of research field experiments. The validation results vary somewhat with location, as well as a change in the bias between the Phase 1 and Phase 2 retrievals (before and after a change in the instrument configuration due to a cooler failure). During Phase 1, a positive bias is found in the lower troposphere at cleaner locations, such as over the Pacific Ocean, with smaller biases at continental sites. However, the Phase 2 CO retrievals show a negative bias at the Pacific Ocean sites. These validation comparisons provide critical assessments of the retrievals and will be used, in conjunction with ongoing improvements to the retrieval algorithms, to further reduce the retrieval biases in future data versions. *INDEX TERMS:* 0394 Atmospheric Composition and Structure: Instruments and techniques; 3360 Meteorology and Atmospheric Dynamics: Remote sensing; *KEYWORDS:* carbon monoxide, MOPITT, validation

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1. Introduction

[2] The Measurements of Pollution in the Troposphere (MOPITT) instrument has been making observations of carbon monoxide (CO) from the NASA EOS Terra satellite since March 2000. This gas filter radiometer uses both pressure-modulated and length-modulated gas correlation cells to measure infrared radiation upwelling from the Earth's surface and atmosphere. CO mixing ratio profiles and total column amounts are retrieved from the radiances. MOPITT views the Earth over all latitudes with a pixel size of 22 km by 22 km and a cross-track swath that measures a near-global distribution of CO every 3 days, providing the first continuous global measurements of CO in the troposphere.

[3] Validation of these products is critical to understanding their value for further scientific analyses. Presented in this paper is the validation of the retrieved CO mixing ratio profiles and column amounts, using coincident in situ aircraft observations. A companion paper [*Deeter et al.*, 2004] discusses the validation of the radiance measurements (Level 1 data).

[4] The MOPITT CO data provide the first opportunity to study CO as a function of latitude, longitude, altitude, and time. The results will greatly improve our understanding of

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[1] Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, Colorado, USA.
[2] Laboratoire d'Aerologie, Toulouse, France.
[3] University of Toronto, Ontario, Canada.
[4] Climate Monitoring and Diagnostics Laboratory, National Oceanic and Atmospheric Administration, Boulder, Colorado, USA.
[5] NASA Langley Research Center, Hampton, Virginia, USA.
[6] Harvard University, Cambridge, Massachusetts, USA.
[7] Japan Aerospace Exploration Agency, Tsukuba, Japan.
[8] University of Tokyo, Tokyo, Japan.
[9] Deutsches Zentrum Für Luft- und Raumfahrt (DLR), Germany.

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the effect of natural and anthropogenic emissions on the global atmosphere. In this paper we summarize the retrieval technique and the approach used to interpret the retrievals and properly compare them to in situ measurements. The aircraft data used in this work are described, and the bias (the difference between MOPITT and the in situ measurements) is presented. In order to quantify the uncertainty in the biases, the results of several sensitivity tests are also discussed.

2. MOPITT Retrievals of CO

[5] MOPITT measures infrared radiation in down-looking view using gas-correlation radiometry [*Drummond and Mand*, 1996]. The instrument's eight channels provide information allowing for the retrieval of the vertical distribution of carbon monoxide. All results discussed here are from the version 3 MOPITT CO retrieval algorithm (V3, released beginning September 2002). The MOPITT instrument includes four channels measuring radiances in the thermal infrared (IR) (near 4.7 μm) and four near-IR channels using reflected solar radiation (near 2.3 μm). The near-IR channels are not used in this version of the retrievals due to low observed signal-to-noise levels. The gas correlation technique results in two signals being measured in each channel: a "difference" (D) signal, which is sensitive to the retrieved target gas, and an "average" (A) signal, which provides information about the background radiance that depends on the surface characteristics and contaminating gas signals. In May 2001, one of the two instrument coolers failed, after which only four of the eight channels were operational, requiring a change to the retrieval algorithm [*Deeter et al.*, 2003]. The data before and after the cooler failure are therefore fundamentally different and have been termed Phase 1, covering March 2000 to May 2001, and Phase 2, starting from August 2001. For the first year of measurements, the retrievals use the D signals from channels 1, 3, and 7 and the A signal from channel 7. After the cooler failure (which disabled channels 1–4), channel 7 was reconfigured to improve the signal-to-noise ratio (see *Deeter et al.* [2004] for more details). This new configuration, using channels 5A, 5D, and 7D, results in the retrievals having a vertical resolution very similar to those of Phase 1, despite the use of different signals in the retrievals. Due to these changes in the instrument and the retrievals, Phase 1 and Phase 2 data are validated separately.

[6] The Level 2 data product includes retrievals of CO mixing ratios for seven levels (at the surface, 850, 700, 500, 350, 250, and 150 hPa), as well as total column amounts. These data products do not represent eight independent pieces of information but are provided so as to give users a consistent vertical profile and total column. The vertical resolution and correlations between retrieval levels (as illustrated by the averaging kernels) are discussed below. A future paper will quantify the number of pieces of information contained in the measurements through additional analysis. The retrieved error covariance matrix is also included in the data file for each retrieval. Further details of the data products are available at the NCAR MOPITT website (http://www.eos.ucar.edu/mopitt/) and the data is available from the NASA Langley Distributed Active Archive Center (DAAC).

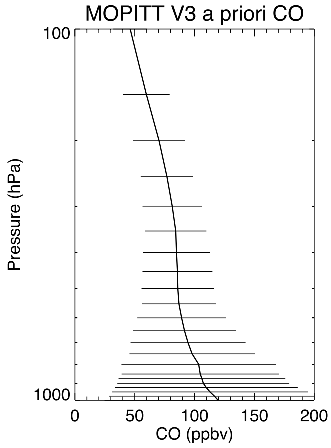

Figure 1. MOPITT a priori CO profile and uncertainty (the square root of the diagonal elements of the covariance matrix).

[7] The retrievals are performed using an optimal estimation technique, the maximum a posteriori solution (MAP) [*Rodgers*, 2000; *Pan et al.*, 1998]. The specific implementation of the retrievals is described by *Deeter et al.* [2003]. The radiances measured by MOPITT are not sufficient to uniquely determine the atmospheric vertical distributions, therefore the inversions must be constrained with a priori information about the concentrations and variability in the atmosphere. A single a priori profile and covariance matrix are used for all locations and seasons in the MOPITT retrievals and were compiled from numerous aircraft observations, representing our best estimate of a global average mixing ratio profile and the correlations between altitudes (see *Deeter et al.* [2003] for details). The a priori CO profile is shown in Figure 1, along with the standard deviation of the mean profile, which is the square root of the diagonal elements of the covariance matrix. A constant a priori was chosen for this version of the retrievals to be sure that the observed variation in the retrievals comes only from the measurements and not from the a priori information. For each MOPITT pixel, the fraction of the retrieved CO that is contributed by the a priori is determined and saved in the Level 2 data product. We are currently exploring the use of a variable a priori for future MOPITT data versions.

[8] The CO retrievals depend on the surface temperature, temperature profile, and surface emissivity. Sufficient information is contained in the thermal band radiances to allow for the retrieval of surface temperature and emissivity along with the CO profile, while temperature profiles are determined from National Centers for Environmental Prediction (NCEP) analyses [see *Deeter et al.*, 2003]. Retrievals are performed on only cloud-free pixels. The occurrence of clouds is determined by comparison of the retrieved surface temperature to NCEP analyzed surface temperatures, along with the Moderate Resolution Imaging Spectroradiometer (MODIS) cloud mask product [*Warner et al.*, 2001].

[9] The new generation of tropospheric satellite measurements of chemical composition do not have the inherent vertical sensitivity that is provided by limb-sounding geometry but are more like many operational nadir temperature sounders in having low vertical resolution. Therefore a

priori information plays a greater role in the retrievals and must be considered explicitly. An understanding of the retrieval technique and its dependence on a priori information is necessary, not only for validation of the retrievals but also for model evaluation and scientific analyses with the MOPITT CO data. This dependence for the vertical profiles and column amounts is described below.

2.1. Mixing Ratio Profiles

[10] The CO profiles retrieved using optimal estimation ($\hat{\mathbf{x}}$) can be expressed approximately as a linear combination of the true profile ($\mathbf{x}$) and the a priori profile ($\mathbf{x_a}$):

$$\hat{\mathbf{x}} \approx \mathbf{A}\mathbf{x} + (\mathbf{I} - \mathbf{A})\mathbf{x_a}, \tag{1}$$

where **A** is the averaging kernel matrix and **I** is the identity matrix [*Rodgers*, 2000]. The averaging kernels provide the relative weighting between the true and a priori profiles and indicate the sensitivity of the retrieval to the measurement. The averaging kernels are very sensitive to the surface temperature and will be different for each point on the globe. Averaging kernels are calculated from the retrieval covariance matrix ($\mathbf{C_{\hat{x}}}$), determined for each retrieval, and the a priori covariance matrix ($\mathbf{C_a}$): $\mathbf{A} = \mathbf{I} - \mathbf{C_{\hat{x}}}\mathbf{C_a}^{-1}$, where **I** is the identity matrix. The retrieval covariance matrix can be related to the weighting function matrix **K**, which describes the model-calculated sensitivity of the measurements to the retrieved quantities, in the following manner:

$$\mathbf{C_{\hat{x}}} = \left(\mathbf{C_a^{-1}} + \mathbf{K^T}\mathbf{C_e^{-1}}\mathbf{K}\right)^{-1}, \tag{2}$$

where $\mathbf{C_e}$ is the radiance error matrix [cf. *Deeter et al.*, 2003].

[11] When comparing the MOPITT retrievals with in situ data, it is necessary to take into account the sensitivity of the retrievals to the true profiles. In order to perform the most meaningful and accurate validation of the retrievals, the in situ correlative data must be transformed using the averaging kernels and a priori profile. A "retrieved" comparison profile, $\hat{\mathbf{x}}_{\mathbf{comp}}$ is calculated by using the in situ profile, $\mathbf{x_{comp}}$, as the "true" profile in equation (1). Then, $\hat{\mathbf{x}}_{\mathbf{comp}}$ is the appropriate quantity to compare with the MOPITT retrievals.

2.2. Total Columns

[12] CO total column amounts are retrieved from the MOPITT observations in addition to the profile retrievals but do not contain any additional information than the profile retrievals and are provided for the convenience of users. The retrieved CO total column is related to the retrieved profile $\hat{\mathbf{x}}$:

$$\hat{c} = \mathbf{t}^T\hat{\mathbf{x}}, \tag{3}$$

where **t** is the total column operator and T indicates the transpose operation. The CO total column averaging kernel can be calculated from the profile averaging kernels by $\mathbf{a} = \mathbf{t}^T\mathbf{A}$. The column operator simply converts the mixing ratio for each retrieval level to a partial column amount. Using the hydrostatic relation, the operator is found to be $\mathbf{t} = 2.120 \times 10^{13}\boldsymbol{\Delta}\mathbf{p}$ (in molecules/cm^2/ppbv), with $\boldsymbol{\Delta}\mathbf{p}$ being the vector of the thicknesses of the retrieval pressure levels (in hPa). The interfaces of the retrieval layers are set at the surface, top of the atmosphere, and the midpoints between the standard seven retrieval levels. Since the CO concentration decreases with altitude above 150 hPa, an effective layer thickness for the 150 hPa retrieval level was determined empirically (by comparing the V3 profile and column retrievals) to be 159 hPa. Therefore for a surface pressure of 1010 hPa, the layer thicknesses would be 80, 155, 175, 175, 125, 100, and 159 hPa (starting at the surface). Column amounts are calculated from the in situ profiles according to equation (3) to validate the CO total column retrievals.

2.3. Averaging Kernels

[13] The shape and magnitude of the averaging kernels depend on the contrast between the surface temperature and air temperature, and on the surface emissivity, and therefore show significant variation with location (as illustrated below in Figure 2). This strong temperature sensitivity also results in a change in the shape of the averaging kernels between day and night over land, as well as between land and ocean locations, as illustrated by *Deeter et al.* [2003]. The change in averaging kernels (reflecting a change in retrieval sensitivity) can therefore result in a change in the retrieved CO profile or column when there is no change in the true CO concentration.

[14] The retrieval uncertainties (the square root of the diagonal elements of the retrieval covariance matrix) depend on the smoothing error, model parameter error, forward model error, and error due to instrument noise [*Rodgers*, 1990]. The off-diagonal elements of the retrieval covariance matrix provide information about the correlations between altitudes in the retrievals, which are evident in the averaging kernels. The shape of the averaging kernels therefore provides one measure of the vertical resolution of the retrievals.

[15] Since ocean surface temperatures generally do not change between day and night, the shapes of the averaging kernels, as well as the retrieved CO values, do not change on average for a uniform CO distribution. However, greater variability in CO retrievals has been found during day than during night (e.g., J. Crawford et al., Exploring the relationship between MOPITT and in situ observations of CO based on a large-scale feature sampled during TRACE-P, submitted to *Journal of Geophysical Research*, 2004). It is likely that this difference is a result of a difference in the uncertainties in the retrieved surface emissivity between day and night. The thermal band radiances are more sensitive to surface emissivity at night than during the day; therefore the nighttime retrievals are less constrained by the a priori emissivity values. Since the surface emissivity and CO profiles are retrieved simultaneously, the CO retrievals consequently will have a greater reliance on the a priori CO. This results in the CO retrievals showing less variability at night than during the day where other conditions (CO, temperature) are the same. While this effect does not impact significantly the validation results presented here, it should be considered in the interpretation of MOPITT CO retrievals.

3. Validation Data

[16] Data from several sources have been used for the validation of the MOPITT CO retrievals, including measurements planned for coincidence with MOPITT overpasses, and other campaigns that happened to overlap with MOPITT observations. Table 1 summarizes the data

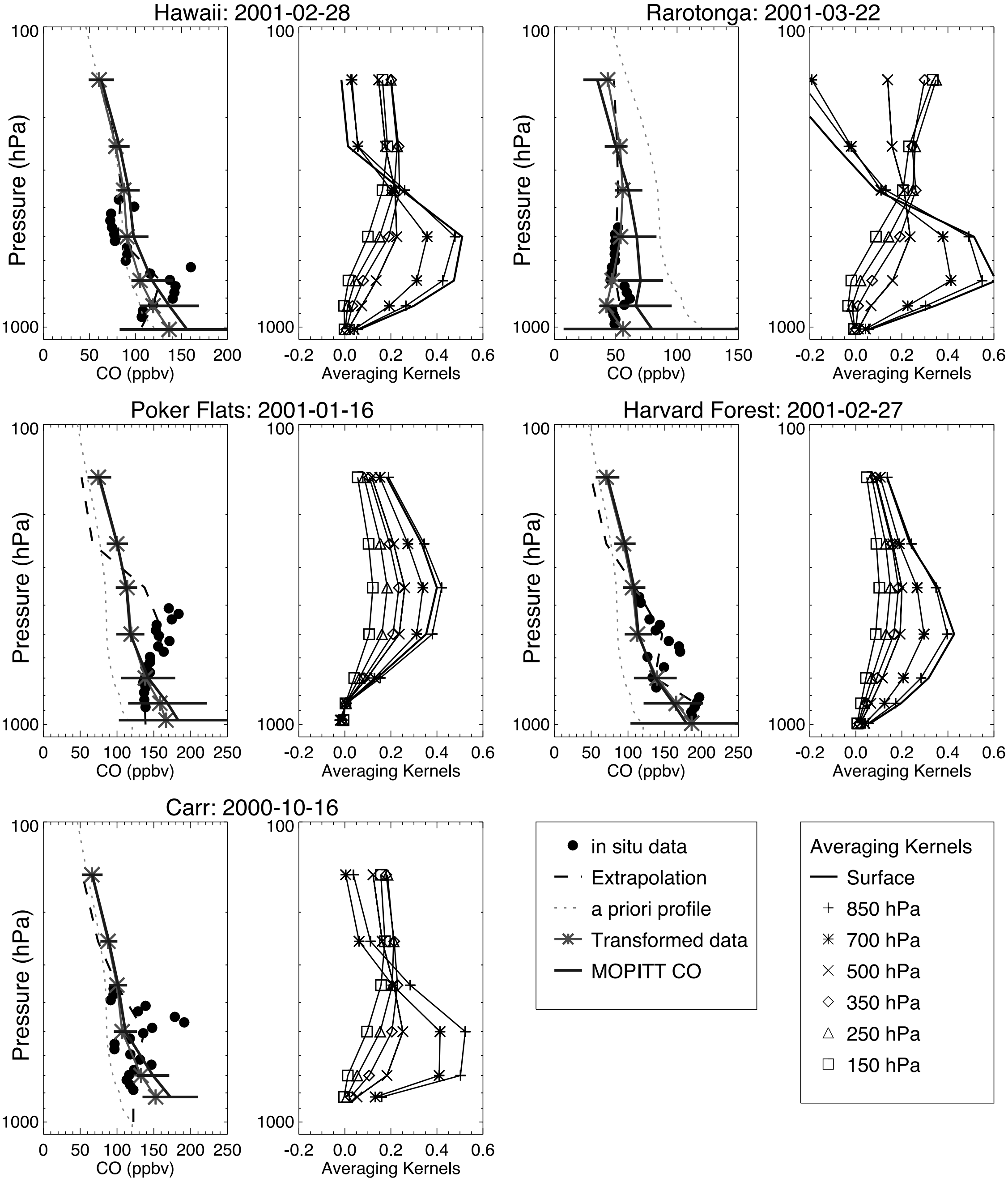

Figure 2. Examples of profile comparisons for each CMDL site, with the corresponding averaging kernels. The left panel of each pair shows the original in situ data, the extrapolated profile, the a priori profile, the "retrieved" in situ profile, and the MOPITT retrieved CO profile with uncertainties. The right panel shows the averaging kernels for each retrieval level. Note for Carr, the surface pressure is less than 850 hPa so there are only six kernels. See color version of this figure at back of this issue.

sets used, with their dates and locations, while additional details are given below.

[17] As part of the MOPITT validation program, a number of ground-based spectroscopic measurements were made from numerous locations, many as part of the Network for the Detection of Stratospheric Change (NDSC). These measurements have not been included in the analysis here because of the difficulty of comparing the retrievals from two different remote-sensing instruments [*Rodgers and Connor*, 2003]. The ground-based instruments have very different averaging kernels from the MOPITT averaging kernels, and without knowing the true vertical profile, there is not sufficient information to make an accurate comparison. The ground-based measurements, however, do provide a valuable record of the seasonal cycle and will be used for a qualitative comparison with MOPITT in a future paper.

3.1. CMDL Standard Sites

[18] As part of the NASA MOPITT validation program, in situ observations of CO and CH_4 were obtained on

Table 1. Profile Validation Sites and Campaigns, With Number of Usable Coincidences, Location of Measurement, and Maximum Altitude (Minimum Pressure) of the Aircraft

Site	Code	Dates	Number	Location	Max. Alt.
		Phase 1			
Hawaii[a]	HAA	Mar 2000–May 2001	6	21.2 N, 158.9 W	375 hPa
Rarotonga[a]	RTA	Mar 2000–May 2001	4	21.2 S, 159.8 W	500 hPa
Poker Flats[a]	PFA	Mar 2000–May 2001	6	65.1 N, 147.3 W	405 hPa
Harvard Forest[a]	HFM	Mar 2000–May 2001	2	42.5 N, 72.2 W	375 hPa
Carr, CO[a]	CAR	Mar 2000–May 2001	6	40.9 N, 104.8 W	370 hPa
MOVE[a]	CIT	Oct 2000	3	Colo., Calif.	195 hPa
SAFARI-2000[a]	S2K	Aug–Sep 2000	8	South Africa	355 hPa
TRACE-P[b]	TRP	Feb–Apr 2001	4	N. Pacific	240 hPa
TOPSE[c]	TOP	Apr–May 2000	6	N. America	400 hPa
BIBLE-C[d]	BBC	Nov–Dec 2000	3	W. Pacific	200 hPa
COBRA[e]	COB	Aug 2000	15	N. America	335 hPa
INCA[f]	INC	Mar–Apr 2000	5	Chile-Germany	200 hPa
		Sep–Oct 2000	1	Scotland	200 hPa
		Phase 2			
Hawaii[a]	HAA	Aug 2001–Dec 2002	20	21.2 N, 158.9 W	375 hPa
Rarotonga[a]	RTA	Aug 2001–Dec 2002	24	21.2 S, 159.8 W	375 hPa
Poker Flats[a]	PFA	Aug 2001–Dec 2002	6	65.1 N, 147.3 W	395 hPa
Harvard Forest[a]	HFM	Aug 2001–Dec 2002	15	42.5 N, 72.2 W	380 hPa
Carr, CO[a]	CAR	Aug 2001–Dec 2002	8	40.9 N, 104.8 W	360 hPa

[a] P. Novelli, CMDL.
[b] G. Sachse, NASA LaRC.
[c] M. Coffey, NCAR.
[d] Y. Kondo, Univ. of Tokyo.
[e] C. Gerbig, Harvard Univ.
[f] H. Schlager, DLR.

biweekly to monthly aircraft flights at five sites operated by the NOAA Climate Monitoring and Diagnostics Laboratory (CMDL) Carbon Cycle Group (see Table 1). The measurements at Carr, Colorado were part of a NOAA program and were not specifically coordinated with Terra overpasses but were usable for validation in numerous cases. Measurements at the other four sites were scheduled to coincide with MOPITT observations when the sky was clear to partly cloudy. Air samples were collected using turboprop aircraft with maximum altitude limits of 300–350 hPa. Individual flights required about 1.5 hours to complete and were timed to be halfway through the profile during the overpass.

[19] Measurements were made by collecting samples of air (approximately 1 liter volume at 2 atmospheres pressure) in glass containers. Seventeen to twenty flasks are held in a suitcase-sized container, and collection of air in a single flask at a unique altitude allows a sampling vertical resolution of 400 m. After each flight the flask packages are shipped to the NOAA laboratory in Boulder, Colorado for trace gas analysis. The mole fractions of CO are determined by gas chromatography, followed by HgO reduction detection [*Novelli et al.*, 1998]. All measurements are referenced to the CMDL/WMO 2000 CO scale [*Novelli et al.*, 2003].

3.2. Intensive Field Campaigns

[20] Several aircraft campaigns have taken place since MOPITT was launched. In several cases aircraft profiles were coordinated with overpasses of MOPITT so as to provide coincident data for validation. In other campaigns, although no coordination was made with MOPITT overpasses, there are a number of cases where the aircraft data coincide with MOPITT observations.

[21] Profiles were measured by NOAA/CMDL as part of the field campaign SAFARI-2000 (The Southern African Regional Science Initiative), and the MOPITT Validation Experiment (MOVE), using the same portable sampling equipment as used for the standard sites. The SAFARI-2000 campaign took place in southern Africa during August and September, when biomass burning is prevalent in the region [*Swap et al.*, 2003]. Air sampling was made from near the surface to 350 hPa using an Aerocommander operated by the South African Weather Service. The MOVE campaign included measurements from a Citation aircraft, which reached approximately 200 hPa during flights over Colorado and California. The vertical sampling was somewhat coarser than at the standard sites, however, allowing for the possibility that fine structure plumes may have been missed by the measurements.

[22] The NASA Global Tropospheric Experiment (GTE) Transport and Chemical Evolution over the Pacific (TRACE-P) consisted of measurements from two aircraft over the North Pacific, during February–April 2001 [*Jacob et al.*, 2003]. On seven flights the DC-8 aircraft collected data in vertical profiles (0–12 km) coincident with MOPITT overpasses, with four of these cases in clear-sky conditions suitable for validation comparisons. The in situ CO measurements were made by the fast response tunable diode laser (TDL) instrument DACOM (Differential Absorption CO Measurement) [*Sachse et al.*, 1987]. The time response of the measurements is 1 s with a precision of 1% or 1 ppbv, whichever is greater. Measurement accuracy is closely tied to the accuracy of the reference gases obtained from NOAA/CMDL.

[23] The Tropospheric Ozone Production about the Spring Equinox (TOPSE) campaign, organized by NCAR, was composed of seven series of aircraft flights between Colorado and Greenland during February–May 2000. Several flights in April and May that happened to coincide

with MOPITT overpasses are used for validation. CO was measured from the NCAR C-130 using a tunable diode laser instrument by M. Coffey and J. Hannigan (NCAR), as well as from canister samples later analyzed by gas chromatography by D. Blake (University of California, Irvine). The two sets of measurements agreed well, so the higher temporal resolution TDL data are used here. TDL observations are made every second with a precision of 1–2 ppbv. Calibration is achieved by the introduction of stored mixtures with concentrations of CO which are traced back to NIST standards. The flights included numerous gradual ramps generally covering from near the surface to an altitude of 7 km but over significant horizontal distance (100–200 km). The CO distribution appeared to be fairly uniform (from the aircraft and MOPITT data), so the ramps were treated as vertical profiles at the midpoint of their horizontal extent.

[24] The Biomass Burning and Lightning Experiment (BIBLE-C) campaign, organized by the Earth Observation Research Center of National Space Development Agency of Japan, took place in November and December 2000, between Japan and Australia. Measurements of CO were made from a Gulfstream II jet aircraft, using the vacuum ultraviolet (VUV) resonance fluorescence technique [*Takegawa et al.*, 2001]. At CO mixing ratios of 100 ppbv, the measurement precision is 1–2 ppbv (at 1 s sampling) and the accuracy is 5%. The calibration standard used agreed with the NOAA CMDL scale to within 2%. Vertical profiles were extracted from the data sets where ascents and descents covered less than 100 km.

[25] The CO_2 Budget and Rectification Airborne study (COBRA) made measurements from the University of North Dakota Citation over the northern United States in August 2000. CO was measured using the Vacuum-Ultraviolet (VUV) fluorescence technique, with a precision of 2 ppbv and accuracy of 3 ppbv [*Gerbig et al.*, 1996, 1999]. Sampling was at 1 Hz, calibrations were made with gas standards traceable to NOAA CMDL. There were extensive fires in Idaho and Colorado during the time of sampling [*Gerbig et al.*, 2003], resulting in highly variable CO distributions over the region.

[26] The INCA (Interhemispheric Differences in Cirrus Properties from Anthropogenic Emissions) campaign included measurements of CO from the DLR Falcon aircraft [*Baehr et al.*, 2003]. The first component of this campaign included flights concentrated near Punta Arenas, Chile between 23 March and 13 April 2000, with transits from Germany. The second set of flights were from Prestwick, Scotland from 27 September to 12 October 2000. CO was measured by VUV Fluorescence [*Gerbig et al.*, 1996] with an accuracy of 5% and precision of 6 ppbv. Vertical profiles were created from ascents and descents in the flight tracks, as for the other campaigns.

3.3. In Situ Profile Extension

[27] Most of the in situ profiles did not reach up to 150 hPa, so in order to apply the averaging kernels for validation, the profiles needed to be extended. Results from the NCAR global chemical transport model MOZART-2 [*Horowitz et al.*, 2003] were used. Monthly means from a simulation driven with analyzed meteorology for 2000 from NCEP have been used. The data and model results are interpolated to the 35 level vertical grid used in MOPITT Forward Model calculations (0.2–1060 hPa) [*Edwards et al.*, 1999]. Since there is often disagreement between the model results and the aircraft profiles, an interpolation scheme was used to result in smooth profiles between the top of the in situ data and the model results in the stratosphere. If the aircraft profile does not reach the tropopause (as determined by the MOZART temperature profile), the measured profile is extended by using the value at the highest altitude up to two levels below the tropopause. The profile is completed by a linear interpolation between that point and the model values above the tropopause. Only profiles with measurements reaching to an altitude above 500 hPa are used for the validation comparisons. This restriction was not applied to the data from Rarotonga during Phase 1 since there are so few profiles and the mixing ratios are fairly constant throughout the troposphere there. The uncertainty introduced into the validation by extending the profiles is estimated below.

4. Profile Comparisons

[28] For each aircraft profile that coincides with a MOPITT overpass, validation comparisons are performed using all cloud-free MOPITT pixels within a 200 km radius of the aircraft profile (generally about 100 pixels). For each pixel the corresponding averaging kernel is applied to the aircraft profile and this transformed profile is compared with the MOPITT retrieved CO profile. The bias (MOPITT minus aircraft) for each aircraft profile is determined as the median of the differences for each pixel. A comparison is made if there are more than 10 MOPITT retrievals within 4 hours of the time the in situ profile was measured. The CMDL and TRACE-P measurements, which were timed to coincide with MOPITT overpasses, generally have less than 1 hour time difference, while the other campaigns and Carr measurements had on average a larger difference. The comparisons are also filtered to use only those profiles where the fraction of the retrieval based on the a priori is less than 60%. The a priori fraction is not constant with altitude and is generally significantly higher at the surface and the highest altitudes, where the MOPITT weighting functions are smallest. The radius of 200 km around the aircraft profile was chosen as a compromise between a larger radius for the purpose of obtaining better statistics and a smaller radius that would reduce errors introduced by variability in the CO distribution. The overall results are not changed by using a smaller radius.

[29] Examples of comparisons of an aircraft profile with an individual MOPITT pixel for each of the CMDL sites are shown in Figure 2. The original in situ profile, the model-extended profile, and the “retrieved” in situ profile are shown with the MOPITT retrieved CO profile. The application of the averaging kernels to the in situ profile generally results in significant smoothing, as well as a shift in mixing ratio when the a priori profile is significantly different from the in situ profile. As discussed above, the shape and magnitude of the averaging kernels show significant variation with location. In the examples of Figure 2, the profiles over the ocean (HAA, RTA) and warm land (CAR) have averaging kernels that show the retrievals distinguish the upper troposphere from the lower tropo-

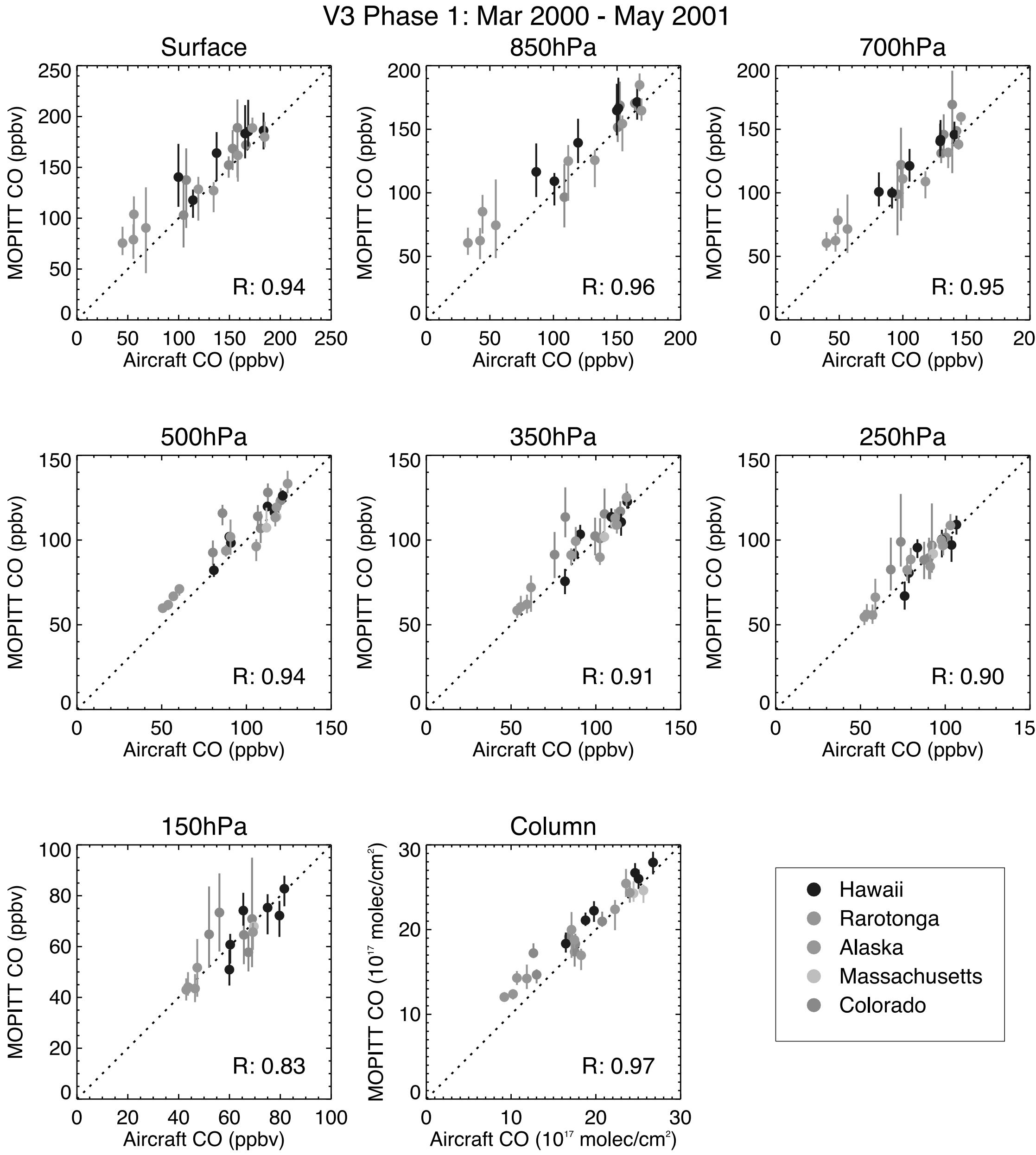

Figure 3. Scatter plot of MOPITT vs aircraft for each retrieval level and column, for Phase 1 data from the five CMDL sites (March 2000–May 2001). The error bars indicate the inter-quartile range for each MOPITT overpass. The dashed line is the 1:1 line and the Pearson correlation coefficient (R) is given. See color version of this figure at back of this issue.

sphere. For the cold continental sites (PFA, HFM), where there is little contrast between the air and surface temperatures, all of the retrieval levels show a broad sensitivity to the middle troposphere and therefore have little vertical resolution.

[30] The examples in Figure 2 illustrate that comparison of MOPITT retrievals to in situ observations without taking into account their averaging kernels would not accurately represent the validation results. The error bars on the MOPITT CO profiles represent the estimated retrieval uncertainties (discussed above). At 500 hPa, the retrieval uncertainties are approximately 20% in the tropics and at middle latitudes and 30–40% at high latitudes, with the largest contribution expected to be due to smoothing error. As seen in Figure 2 the uncertainty in the lower troposphere can be quite large (e.g., 50% at Rarotonga).

4.1. Phase 1

[31] Figure 3 shows the correlations between MOPITT CO retrievals and the in situ CO (after applying the averaging kernels) at the five CMDL sites for each retrieval level and the columns. Each symbol represents the comparison of a single aircraft profile with an ensemble of pixels from a MOPITT overpass. The median and interquartile (i.e., where 50% of the values lie) range of the MOPITT CO retrievals within 200 km of the aircraft profile are shown. Uncertainties in the in situ measurements are believed to be on the order of a few percent (hence are not shown on the

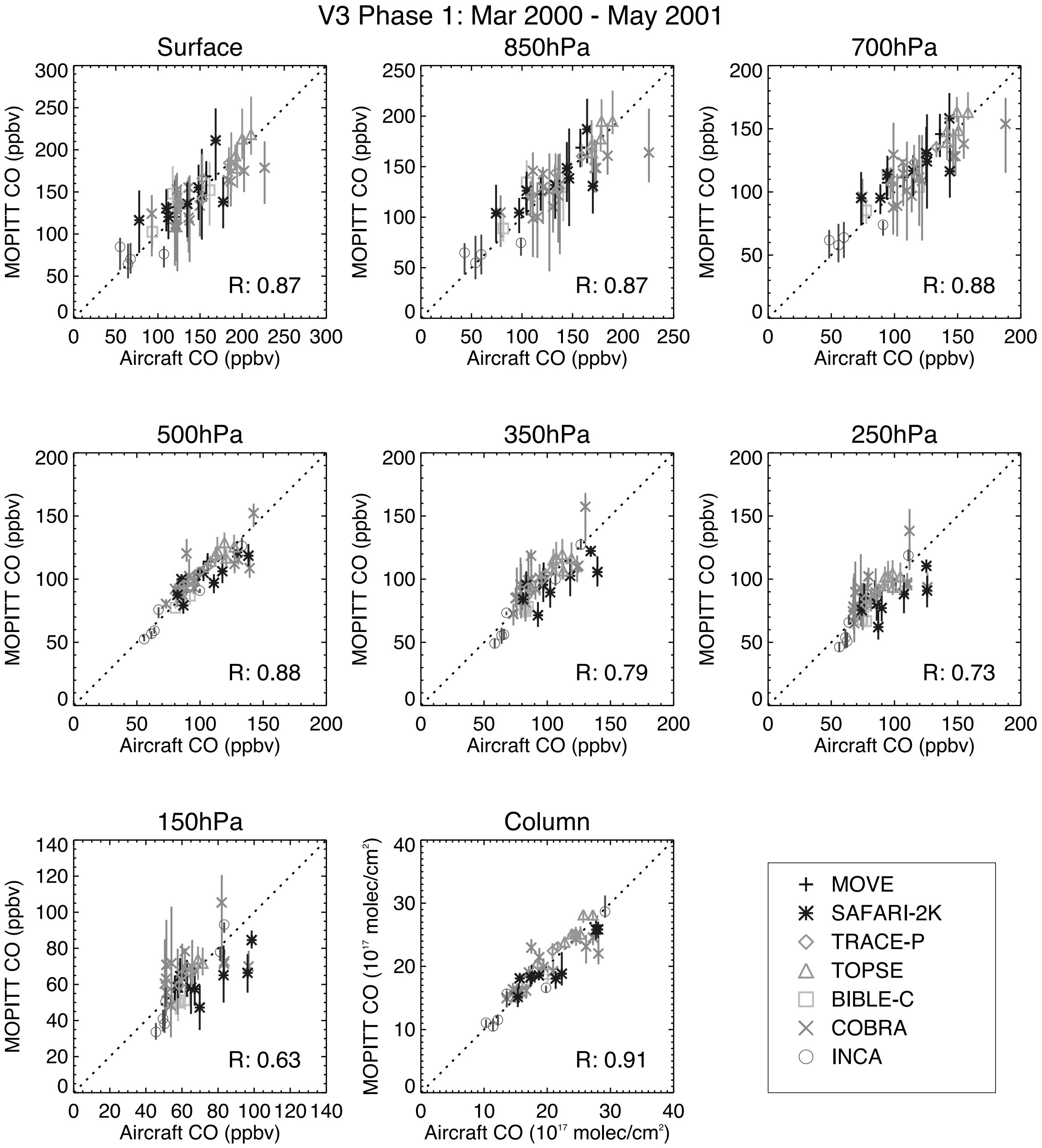

Figure 4. As Figure 3 for the campaign data during March 2000–May 2001. See color version of this figure at back of this issue.

graph for clarity). Examples of the estimated MOPITT uncertainties are shown in Figure 2. In general, very good correlation is seen. When examining these scatter plots it is useful to keep in mind the shapes of the averaging kernels (shown in Figure 2). For example, the 500 hPa retrieval level has significantly less scatter than the other levels as a result of the generally broad and weak averaging kernel, indicating a significant fraction of the information used in the retrieval is from the a priori profile. The mixing ratios at the higher altitudes are much less than in the lower troposphere and have a smaller range.

[32] The results from the field campaigns are shown separately in Figure 4. The correlation coefficients are significantly less for these comparisons than for the CMDL station data, and in general the campaign data show much greater variability. One notable exception is the TRACE-P campaign, for which the four comparisons lie very close to the 1:1 line. There are several factors that probably contribute to such good agreement, including that the aircraft flights were made coincident with MOPITT overpasses in a tight spiral up to 10–12 km altitude, which is higher than almost all of the other profiles. The measurements were also made over the Pacific Ocean away from source regions, so the profiles are fairly representative of the 200 km radius used for averaging the MOPITT retrievals. The measurements as part of MOVE and SAFARI-2K were also made coincident with MOPITT overpasses. The measurements from the Citation aircraft during MOVE show generally good agreement. The comparison with the SAFARI measurements is somewhat more variable, however. This poorer agreement is likely a result of the difficulty of making representative measurements from the aircraft in the com-

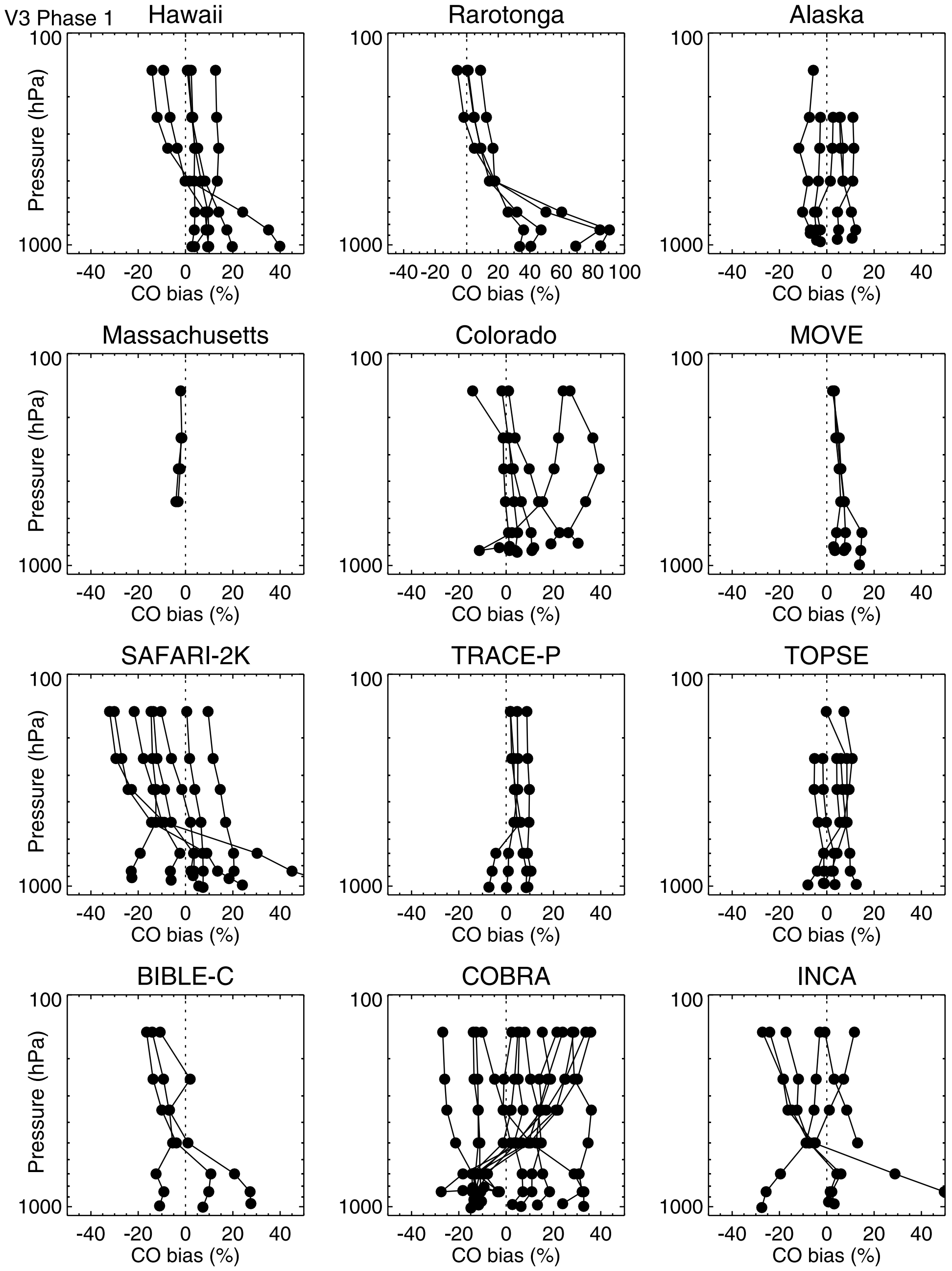

Figure 5. Bias (MOPITT minus aircraft, in percent) for each MOPITT overpass at each CMDL site for Phase 1. Lines connect medians of the bias at each level for each overpass (note scale change for Rarotonga).

plex environment of South Africa in the presence of intense biomass burning plumes that may not be representative of the CO distribution of a larger region. While biomass burning plumes generally contain high concentrations of particles, there is no indication that MOPITT CO retrievals are affected by aerosols. The comparisons for the profiles from TOPSE, BIBLE-C, and COBRA also show very large variability, which is likely due to the poorer coincidence in time between the aircraft and satellite observations. The measurements from these campaigns were also made over larger horizontal distances than TRACE-P and SAFARI so may have sampled a variety of airmasses.

[33] The bias between the Phase 1 MOPITT retrievals and the in situ measurements has been summarized for each location and campaign in Figure 5. The median value of the bias for each aircraft profile is shown. Some profiles have limited altitude range due to the filtering by a priori fraction (as mentioned above). There are some differences in the bias depending on location. The relatively clean ocean sites, Hawaii and Rarotonga, show a larger positive bias in the lower troposphere than at other sites. The reason for MOPITT being biased high for low mixing ratios at low altitudes may be because the off-diagonal elements of the a priori covariance matrix constrain the retrievals too much.

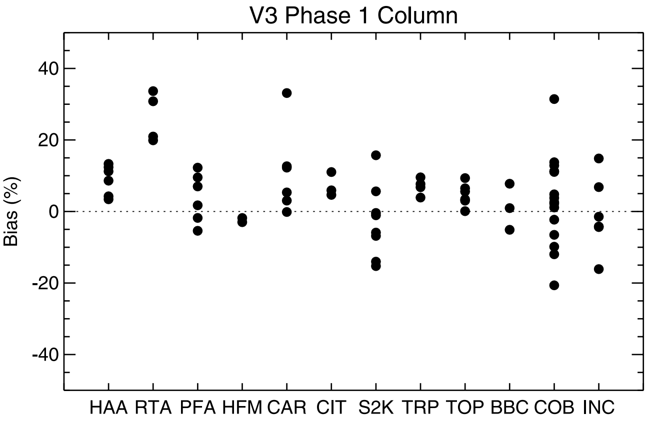

Figure 6. Bias in the column retrievals for Phase 1. Site codes are given in Table 1.

The results from Rarotonga, and several profiles from INCA, are the only ones available for the remote Southern Hemisphere but are expected to be representative of clean ocean environments. The wide range of values seen at several sites (e.g., Carr, SAFARI, and COBRA) are indicative of the difficulty in making in situ observations that are representative on the scale that the satellite observations are made, particularly in the vicinity of emission sources. Figure 6 shows the bias in the column retrievals for each site and is consistent with the biases in the profiles, on average. An analysis of all the validation results as a function of time did not indicate that the bias varied during this first year of data.

[34] Table 2 summarizes the bias for each altitude, averaging over all of the comparisons made for the Phase 1 data. The bias is shown both as the absolute difference in mixing ratio and as percent. In the lower troposphere MOPITT is high by 4–5 ppbv, or 6–8%, whereas the bias is somewhat less in the upper troposphere. The column retrievals have an average bias of about 5%. The standard deviations of the biases are significantly larger than the biases themselves at all altitudes and are an indication of the uncertainty in our determination of the bias. However, there are other sources of uncertainty in the retrieval bias that are not easily quantified. Several sensitivity tests have been performed to examine these issues and are discussed below. Some of the variability may also be due to uncertainties in the retrievals of surface temperature and surface emissivity.

4.2. Phase 2

[35] Similar validation comparisons have been made for the Phase 2 data, covering August 2001 through December 2002, using the five standard CMDL stations (see Table 1). The correlations of MOPITT to aircraft data are shown in Figure 7. While there is considerable variation, in general we find that at the surface, 850 and 700 hPa retrieval levels the lowest mixing ratios are biased low (MOPITT is lower than the correlative measurements), while at higher CO mixing ratios the MOPITT retrievals are greater than the in situ measurements. These are somewhat different results than those found for Phase 1 comparisons, where the very lowest values were biased high in MOPITT (see Figure 3).

[36] Figure 8 shows the biases for each profile sorted by site, and Figure 9 shows the bias in the column retrievals. The ocean sites, Hawaii and Rarotonga, show a negative bias of up to 40% in the lower troposphere (opposite to Phase 1), while the continental sites have a positive bias on average. In the upper troposphere the bias is less than ±20% on average, as seen in the Phase 1 data.

[37] The bias averaged over all of the sites for each retrieval level for Phase 2 is also given in Table 2. The results are somewhat different from the Phase 1 results, with the average bias less than ±1 ppbv (−3% to 2%) at all altitudes. While these average values are quite low, the wide range of biases seen between validation sites indicate that at a given location the retrieval error could be somewhat larger.

[38] These results are consistent with the radiance biases found in the Level 1 validation discussed in the accompanying paper [*Deeter et al.*, 2004]. For example, the negative biases in the radiances for Phase 1 and Hawaii and Rarotonga correspond to positive biases in the retrievals. In addition, during Phase 2 the radiance bias at Hawaii was positive, translating to a negative bias in the retrievals. The causes of the differences in the retrieval results between Phase 1 and Phase 2 are not completely understood at present and are continuing to be investigated.

5. Bias Uncertainty

5.1. In Situ Profile Extension With Model

[39] As indicated in Table 1, most of the aircraft profiles did not reach the upper troposphere, so a significant portion of the profile used for validation comes from model results (as described above). We evaluated the uncertainty introduced by this extrapolation of the profiles by modifying the in situ profiles at the upper retrieval levels and examining the effect on the validation comparisons. In particular, the model-extended profiles were decreased by 20 ppbv at the 150 and 250 hPa retrieval levels and then transformed with the averaging kernels and a priori profiles, as described above. These were then compared to the MOPITT retrievals. The resulting biases differed from the originals at all levels. The change in bias was computed for each validation profile,

Table 2. Absolute and Percentage Biases (Mean and Standard Deviation) for Each Retrieval Level and Column, Averaged Over All Comparisons

Level	Absolute Bias[a]	Percentage Bias
	Phase 1	
Surface	5.7 ± 20.6	8.1 ± 21.5
850 hPa	4.1 ± 18.8	8.1 ± 22.2
700 hPa	4.2 ± 14.5	6.5 ± 16.1
500 hPa	2.7 ± 9.8	3.8 ± 10.1
350 hPa	1.7 ± 11.9	2.6 ± 12.3
250 hPa	0.7 ± 11.5	1.7 ± 13.0
150 hPa	−0.8 ± 10.5	−0.2 ± 15.8
Column	0.7 ± 1.9	4.9 ± 10.8
	Phase 2	
Surface	0.7 ± 24.6	−1.5 ± 21.2
850 hPa	−0.6 ± 20.9	−2.4 ± 20.5
700 hPa	0.9 ± 16.1	−0.2 ± 16.3
500 hPa	0.5 ± 9.6	0.9 ± 10.4
350 hPa	0.7 ± 8.9	1.6 ± 10.1
250 hPa	0.6 ± 7.8	1.6 ± 9.9
150 hPa	−0.8 ± 6.8	−0.2 ± 10.8
Column	−0.2 ± 2.2	−0.5 ± 12.1

[a] Measured in ppbv for profiles and 10^{17} molec/cm^2 for columns.

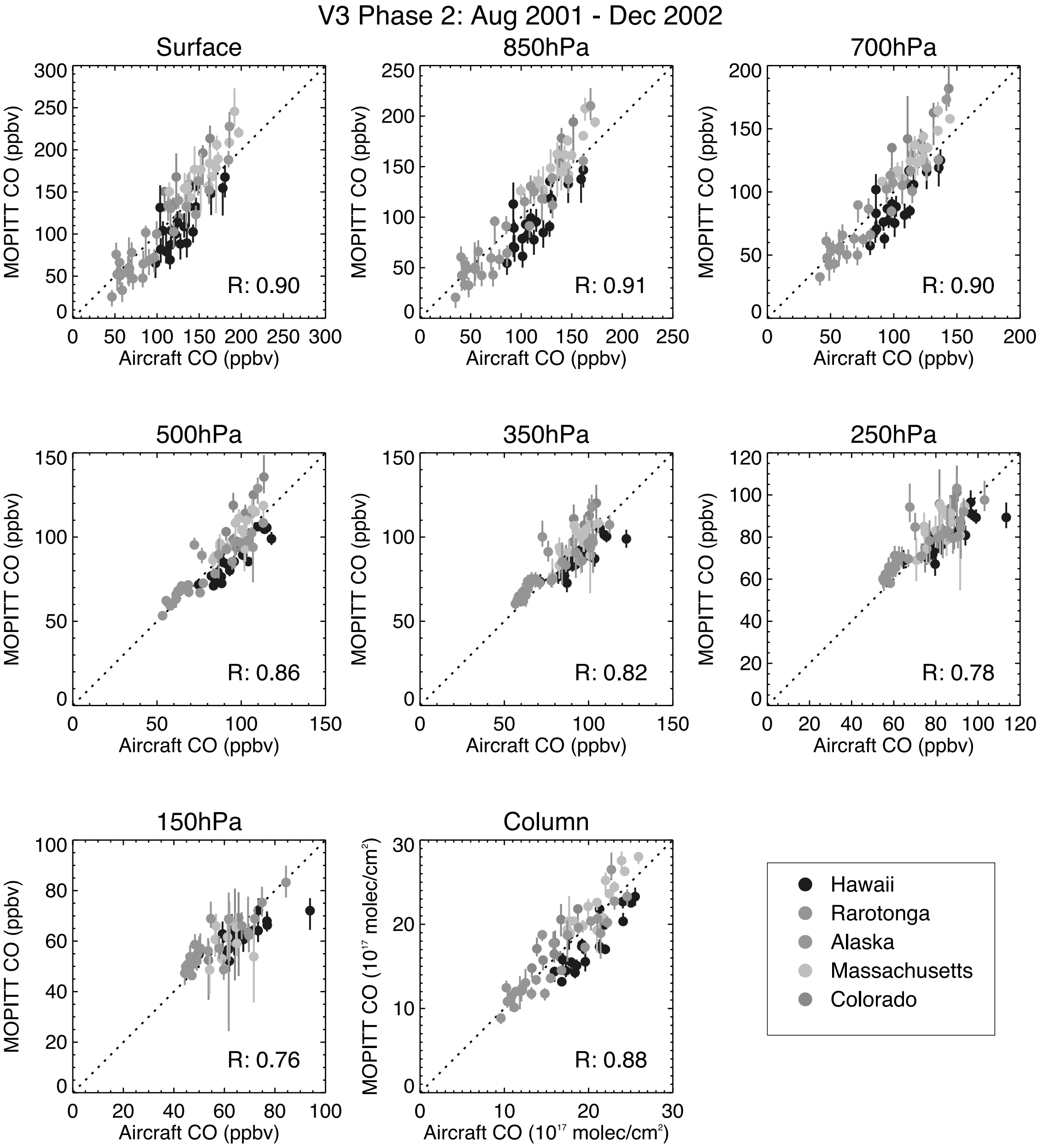

Figure 7. As Figure 3, for Phase 2 (August 2001–December 2002). See color version of this figure at back of this issue.

and the average and standard deviation over all of the profiles is given in Table 3 for each retrieval level. Due to the broad shape of the averaging kernels the validation profiles were changed significantly at the lower altitudes. The 20 ppbv change at the top two levels of the in situ profiles results in a change of about 4 ppbv in the lower troposphere and about 8 ppbv in the upper troposphere. Our knowledge of the CO concentrations in the upper troposphere and lower stratosphere is limited, particularly at the times that the in situ profiles were measured, so 20 ppbv is a reasonable estimate of the uncertainty in these profiles at those altitudes.

5.2. In Situ Measurement Uncertainty

[40] Another source of uncertainty in the bias is due to uncertainties in the in situ measurements. In general, these errors are much smaller than errors introduced by the retrievals and other sources. However, it is important to consider their contribution. A large fraction of the measurements used for the validation presented here are made by CMDL using reference gases tied to the CMDL CO scale. The uncertainty in the CMDL calibration scale (≈1–4%) is greater at lower mixing ratios [*Novelli et al.*, 2003]. This results in slightly higher uncertainty in the retrieval biases at the sites where low CO is measured (such as Rarotonga and Hawaii in summer).

[41] The measurements used here that were made by groups other than CMDL used different observing techniques, which could introduce some additional uncertainty. While no direct comparison has been made of all the instruments used in this validation study, a measurement intercomparison was made between a TDL and two VUV

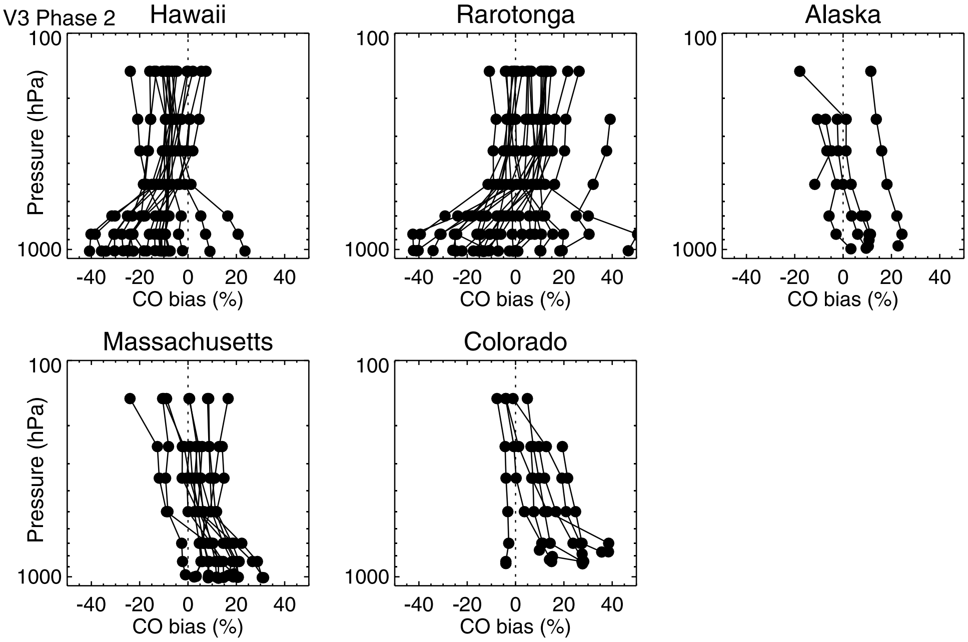

Figure 8. As Figure 5, for Phase 2.

CO instruments during the 1997 North Atlantic Regional Experiment [*Holloway et al.*, 2000]. The instruments in that comparison are quite similar to those used here, so the results of their agreement can be considered comparable to this case. The intercomparison showed that the three instruments agreed to within 11% and had systematic offsets of less than 1 ppbv.

6. Conclusions

[42] To properly use and interpret tropospheric satellite measurements, such as those from MOPITT, it is necessary to understand what the retrievals represent. Since the inversion of the measured radiances is an ill-posed problem, meaning there is not a unique solution, it is necessary to constrain the retrievals with a priori information. Limited vertical resolution of the retrievals is also inherent in the retrievals. The averaging kernels for the retrievals provide much of the information needed to interpret the MOPITT observations, including the profile smoothing and vertical resolution and the a priori content. In order to accurately validate the CO retrievals, as well as use the retrievals for model evaluation and other studies, the averaging kernels and a priori information must be taken into account. Tropospheric measurements are now being made, or soon will be, by other satellite instruments, such as SCIAMACHY on the ESA satellite ENVISAT, AIRS on EOS/Aqua, and TES on EOS/Aura, and use of these data will similarly require careful consideration of measurement sensitivity in the interpretation of their retrievals.

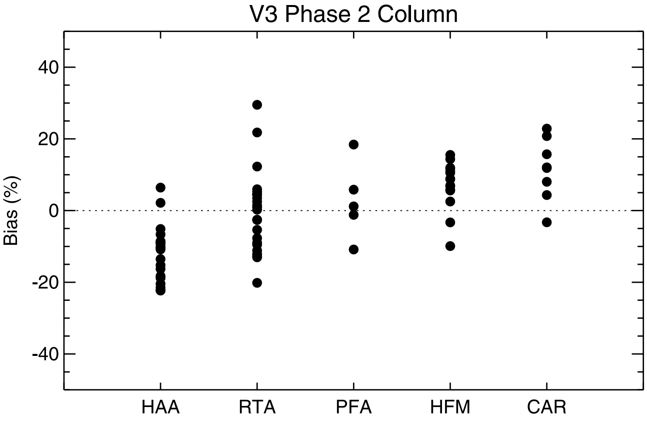

Figure 9. Bias in the column retrievals for Phase 2.

[43] The MOPITT CO retrievals have been validated using in situ CO measurements from aircraft. As part of a regular sampling program coordinated by NOAA CMDL, as well as coincident research experiments, 69 profiles were sampled during the Phase 1 (March 2000–May 2001) of MOPITT measurements, and 73 profiles were sampled during Phase 2 (August 2001–December 2002). The MOPITT CO profiles were compared with the in situ profiles after transforming them with the averaging kernels and the a priori CO profile, and the resulting bias was determined for each retrieval level as well as the total column. The overall agreement between the MOPITT retrievals and in situ measurements is very good for both Phase 1 and Phase 2, although there is a slight difference in the bias amounts between the two phases. For Phase 1, on average the bias is positive throughout the troposphere with

Table 3. Change in Bias Due to Reducing the In Situ Profile at 150 and 250 hPa by 20 ppbv

Level	Change in Bias, ppbv	Std Dev, ppbv
Surface	−1.9	8.1
850 mb	−4.6	6.7
700 mb	−3.7	4.8
500 mb	−6.7	1.3
350 mb	−8.8	1.4
250 mb	−8.4	1.9
150 mb	−6.7	1.9

slightly larger differences in the lower troposphere than the upper levels: at 700 hPa the average bias is about 4 ppbv (7%) and at 350 hPa it is about 2 ppbv (3%). The Phase 2 retrievals show a small average bias at all altitudes: 1 ppbv (−0.2%) at 700 hPa and 0.7 ppbv (1.6%) at 350 hPa. However, the standard deviation of these mean biases is large, particularly in the lower troposphere (20 ppbv, 20%), indicative of the large variability seen in the biases. During both Phase 1 and Phase 2, larger biases are seen in clean environments, such as the South Pacific. The validation profiles represent a wide variety of locations and environments, including oceanic sites in both the Northern and Southern Hemispheres and both clean and polluted atmospheric conditions in the Northern Hemisphere. This large number of comparisons for diverse conditions provides confidence in the accuracy of version 3 of the MOPITT CO retrievals.

[44] Future versions of the retrieval algorithms will be validated using the data and procedure described here. Further work is being done to quantify the pieces of independent information contained in the profile retrievals. In addition, the MOPITT CO measurements will continue to be validated against available in situ data throughout the life of the instrument to detect any long-term changes in the satellite observations.

[45] **Acknowledgments.** Valuable comments on this manuscript were provided by W. Randel and A. Lambert. This work was funded by the NASA EOS MOPITT program. The NOAA/CMDL measurements at Alaska, Massachusetts, Hawaii, and Rarotonga were supported by the NASA EOS Validation program. NCAR is sponsored by the National Science Foundation and operated by the University Corporation for Atmospheric Research.

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J.-L. Attié, Laboratoire d'Aerologie, Observatoire Midi Pyrenees, 14 av. E. Belin, 31400 Toulouse, France. (attjl@aero.obs-mip.fr)

J. Baehr, H. Schlager, and H. Ziereis, Deutsches Zentrum Für Luft- und Raumfahrt (DLR), Institut fur Physik der Atmosphäre, Oberpfaffenhofen, D-82234 Wessling, Germany. (janine.baehr@dlr.de; hans.schlager@dlr.de; ziereis@dlr.de)

J. S. Chen, M. T. Coffey, M. N. Deeter, D. P. Edwards, L. K. Emmons, G. Francis, J. Gille, J. W. Hannigan, S.-P. Ho, B. Khattatov, J.-F. Lamarque, D. Mao, J. Warner, V. Yudin, and D. Ziskin, Atmospheric Chemistry Division, National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO 80307, USA. (jschen@ucar.edu; coffey@ucar.edu; mnd@ucar.edu; edwards@ucar.edu; emmons@ucar.edu; gfrancis@ucar.edu; gille@ucar.edu; jamesw@ucar.edu; spho@ucar.edu; boris@ucar.edu; lamar@ucar.edu; dmao@ucar.edu; juying@ucar.edu; vyudin@ucar.edu; ziskin@ucar. edu)

J. Drummond, Department of Physics, University of Toronto, Toronto, Ontario, M5S 1A7, Canada. (james.drummond@utoronto.ca)

C. Gerbig, Department of Earth and Planetary Sciences, Harvard University, 20 Oxford Street, Cambridge, MA 02138, USA. (chg@io.harvard.edu)

S. Kawakami, Office of Space Applications, Japan Aerospace Exploration Agency, Satellite Program Office, 2-1-1 Sengen, Tsukuba-city, Ibaraki, Japan 305-8505. (kawakami.shuji@jaxa.jp)

Y. Kondo and N. Takegawa, Research Center for Advanced Science and Technology, University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo 153-8904, Japan. (kondo@atmos.rcast.u-tokyo.ac.jp; takegawa@atmos.rcast.u-tokyo.ac.jp)

P. Novelli, Climate Monitoring and Diagnostics Laboratory, National Oceanic and Atmospheric Administration, 325 Broadway, Boulder, CO 80305, USA. (pnovelli@cmdl.noaa.gov)

G. Sachse, NASA Langley Research Center, Hampton, VA 23681-2199, USA. (g.w.sachse@larc.nasa.gov)

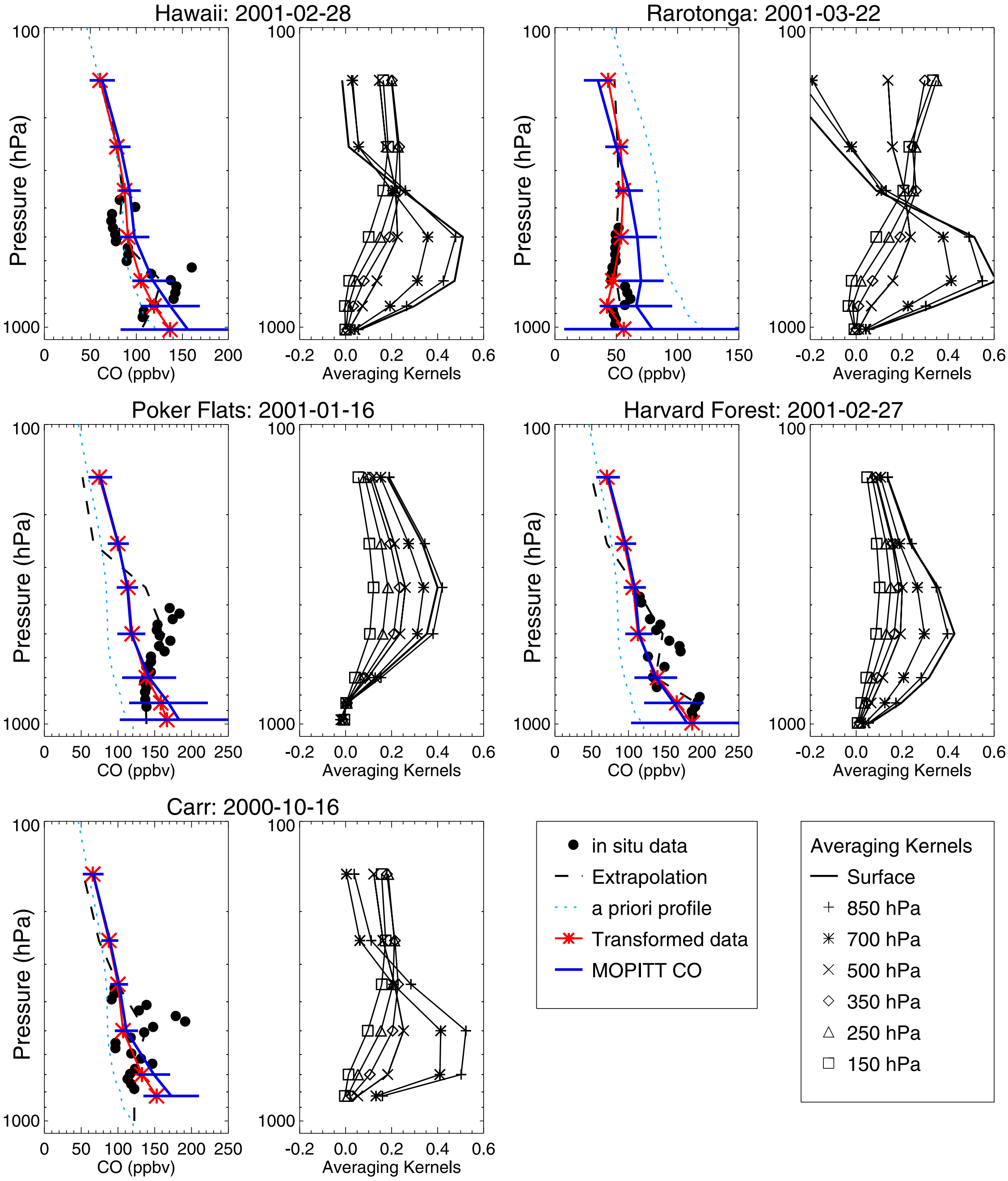

Figure 2. Examples of profile comparisons for each CMDL site, with the corresponding averaging kernels. The left panel of each pair shows the original in situ data, the extrapolated profile, the a priori profile, the "retrieved" in situ profile, and the MOPITT retrieved CO profile with uncertainties. The right panel shows the averaging kernels for each retrieval level. Note for Carr, the surface pressure is less than 850 hPa so there are only six kernels.

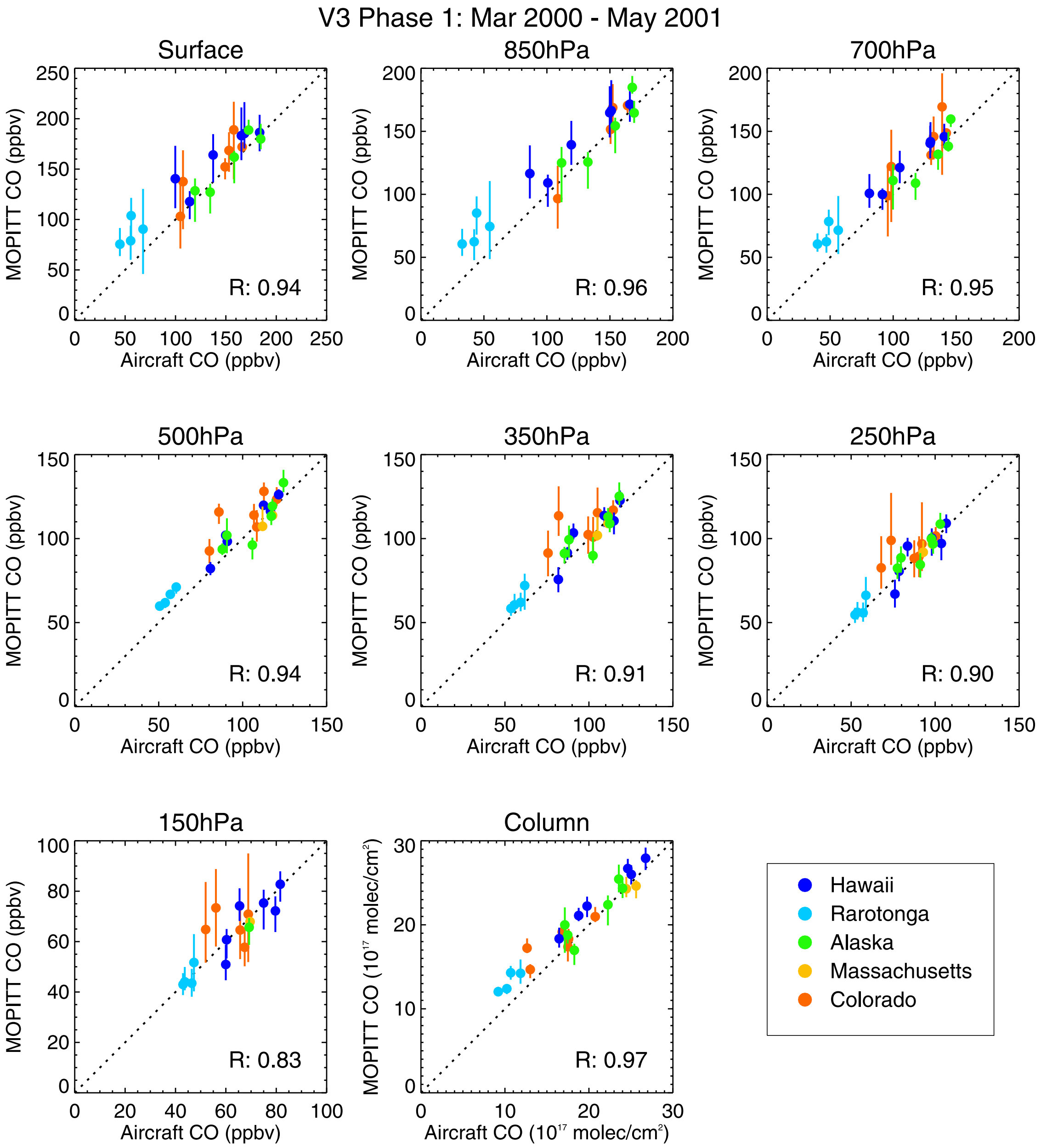

Figure 3. Scatter plot of MOPITT vs aircraft for each retrieval level and column, for Phase 1 data from the five CMDL sites (March 2000–May 2001). The error bars indicate the inter-quartile range for each MOPITT overpass. The dashed line is the 1:1 line and the Pearson correlation coefficient (R) is given.

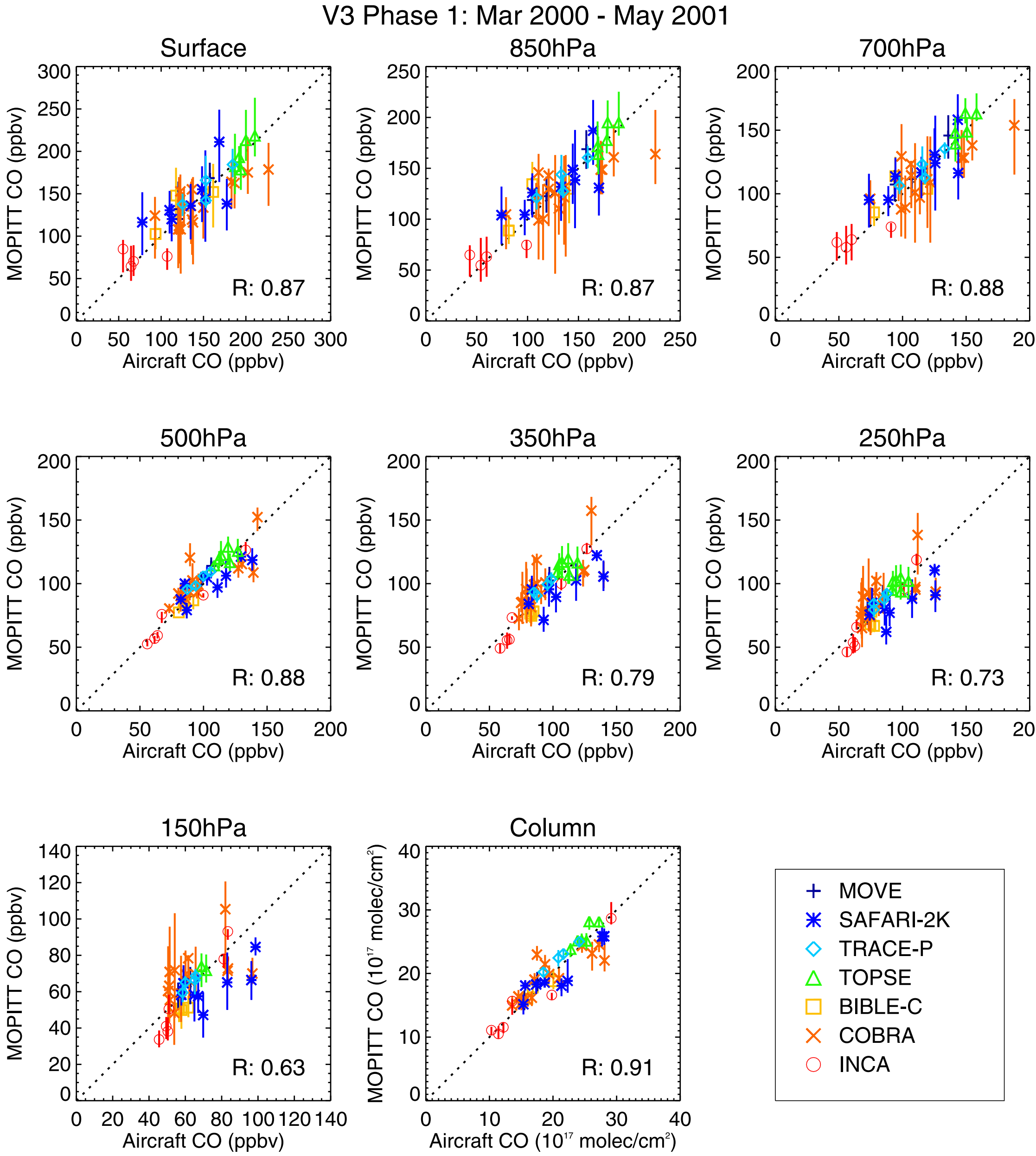

Figure 4. As Figure 3 for the campaign data during March 2000–May 2001.

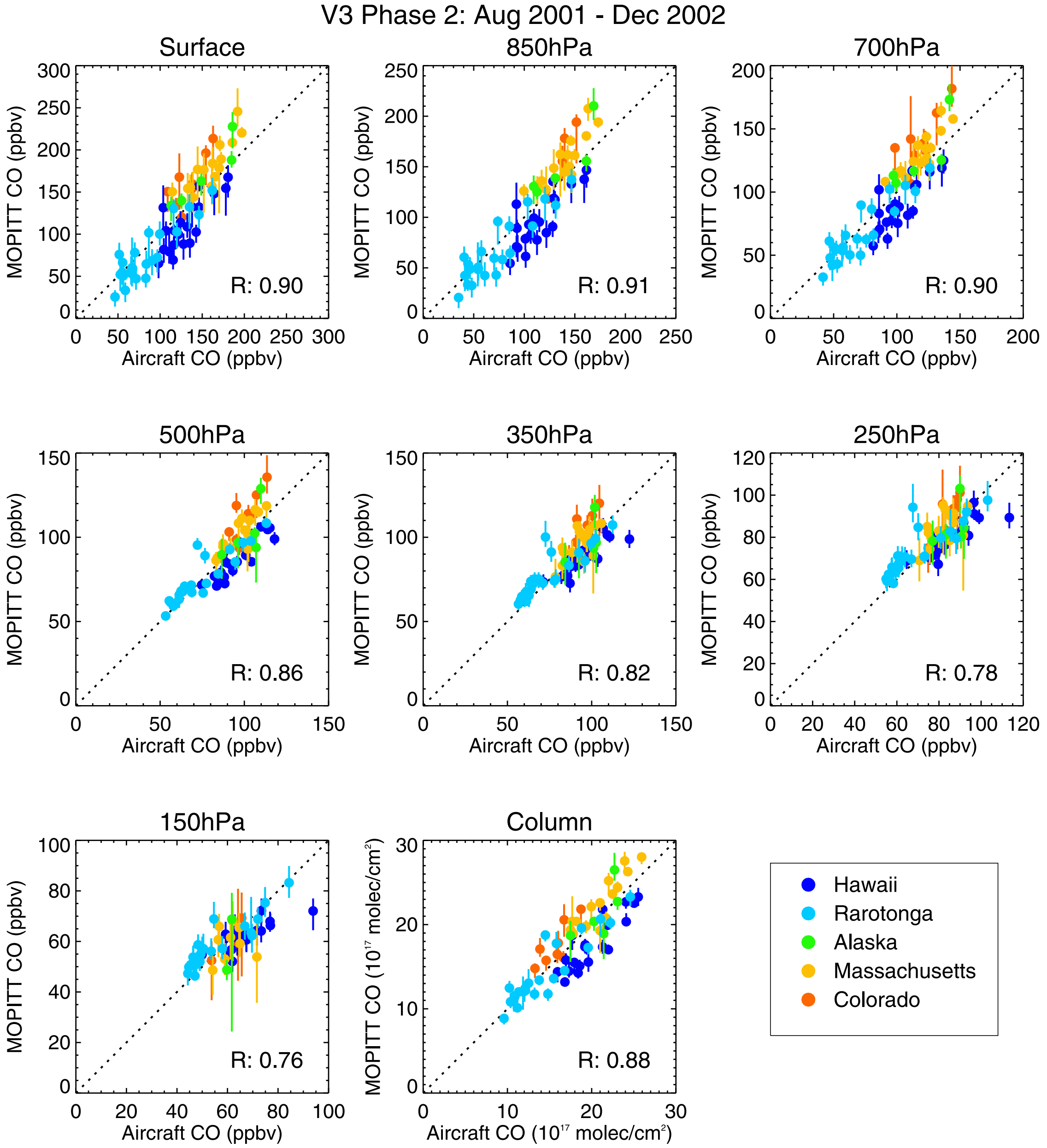

Figure 7. As Figure 3, for Phase 2 (August 2001–December 2002).