Progress Report For EOS Validation Contract S-97890-F:
Vertical Profiles of Carbon Monoxide and Methane in the Troposphere.

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14 August 2001

SUMMARY:

In support of the MOPITT validation, we measure the vertical distributions of carbon monoxide and methane in the lower and middle troposphere. Using charter aircraft and portable, computer-controlled sampling units, tropospheric air is collected on a regular basis above five geographically diverse sites: Poker Flats, Alaska; Harvard Forest, Massachusetts; Carr, Colorado; Molokai, Hawaii; and Rarotonga, Cook Islands. These samples are shipped to Boulder after each flight and analyzed for CO, CO_2, CH_4, H_2, N_2O, and SF_6. Trace gas mixing ratios have also been determined over the ARM/CART site in Lamont Oklahoma as part of the MOPITT Validation Experiments (March 1998 and May 2001). Flights above Northern Colorado have been coordinated with the DOC Citation to compare measurements made by CMDL and those made by the MATR instrument (NCAR). Vertical profiles determined over Southern Africa during SAFARI-2000 were timed to coincide with the TERRA overpass, and whenever possible, with over flights of the ER-2 (which carried MOPITT-A). Over the past year, more than 100 profiles have been measured at 7 geographically diverse sites (Figure 1). To date, these data have been the fundamental components of the MOPITT validation.

PROGRESS: JUNE 2000 – JULY 2001:

MOPITT ANCHOR SITES:

Sampling continued at all five vertical profile sites. Modification of the CMDL automated sampling units improved the efficiency of sample collection and the loss of samples was minimized. The new custom designed valve gears, motors, and flask pistons have all contributed to higher success rates (defined as the number of good samples analyzed per 20 flask package, typically = 90 ± 10%). Aircraft logistics and hardware are operational at all five sites (Table 1). Aircraft planning is coordinated with TERRA flight paths using an overpass predictor developed at NCAR (URL: http://mop1.eos.ucar.edu/cgi-bin/op/test/orb.pl). An oxygen system was provided to Air Rarotonga, permitting them to increase their maximum sampling altitude from 14,000 to 22,000 ft. All flights were scheduled to begin the descending profile about one half hour before the overpass, such that sampling was halfway completed as the satellite passed over.

TABLE 1: ANCHOR SITES: JUNE 2000 – JULY 2001

Location/ Site category	Lat/Long	Plane	Maximum Altitude (km)	No. Successful Flights
Poker Flats, Alaska Boreal Forest	65.1°N, 147.5°	Piper Chieftain	7.6	13
Carr, Colorado Background Continental	40.1°N, 104.1°W	Cessna 210	9.2	24
Beverly, Mass. Polluted Boundary Layer	42.5°N, 71.2°W	Cessna 421	8.7	13
Molokai, Hawaii Tropical Ocean, NH	21.4°N, 157.2°W	Cessna 414	7.6	19
Rarotonga, Cook Islands Tropical Ocean, SH	21.3°S, 159.6°W	Bandeirente	6.7	14

The long-term measurements made by this program can provide a rare view of trace gas climatology in the remote free troposphere. Considerable variation of trace gases with altitude is common. Often, profiles of different gases tract one another (Figure 2). These reflect regional scale pollution or the transport of polluted air to the background free troposphere. Back trajectory analysis can sometimes trace air parcels arriving at the MOPITT sites back to source regions, but many cases the source of the elevated concentrations is not so easily understood. Boundary layer mixing ratios, determined from aircraft at the MOPITT sites show trace gas levels and latitudinal gradients comparable to those measured at the surface (Figure 3). The highest levels of CO, CH_4, and CO_2, at all sites except Rarotonga, are typically found near the surface, and strongly correlated structure in mixing ratios of the various species is often found in the free troposphere. Trace gases above Poker Flats are influenced by transport of anthropogenic surface emissions from the northern mid-latitudes, and depending on the meteorology, may increase, decrease or remain constant with altitude. Near surface mixing ratios above Harvard Forest, MA. were the highest measured: up to 250 ppb in winter. From a surface maximum, trace gases typically then decreased with altitude (the boundary layer at HFM reflects the regional pollution of southern New England). Profiles determined above Molokai, Hawaii during late winter/early spring often show layers of strongly elevated trace gases. Mixing ratios above Rarotonga were generally the lowest observed, consistent with the well-defined decreasing north to south surface interhemispheric gradient.

In spite of the best attempts by all involved to obtain a profile every 2 weeks, scheduled flights could not always be flown, or the samples collected were contaminated. Each site had it's own unique problems:

Rarotonga, Cook Islands: Progressive contamination of the pump unit and intake lines corrupted samples from 4 flights in the fall of 2000 before the problem was resolved. Tropical cyclones during spring 2001 prevented several scheduled flights.

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Molokai, HI: The most successful MOPITT site, several sampling dates were cancelled when the aircraft was needed for emergency medical service.

Carr, CO. Primarily operated by NOAA, the frequency of flights was cut due to lack of funding.

Harvard Forest, MA: Unexpected aircraft maintenance extended from May to October 2000. Inclement weather forced cancellation of several flights during the winter of 2000-2001.

Poker Flats, AK: Weather often prevented scheduled flights. Aircraft overhauls in September-October 2000 interrupted regular sampling.

SAFARI-2000

Our participation in the dry season SAFARI-2000 initiative was successful. From base of operations in Pietersburg, South Africa, we flew aboard the second RSA Aerocommander (JRB) over the Indian Ocean, coastal Mozambique, South Africa, and Botswana. Flight plans specific for MOPITT validation were easily coordinated with the other demands on the aircraft. After some initial problems with the plane, vertical profiles extending from 28,000 ft. to 1000 ft above the surface were achieved. Shipping of the samples between Pietersburg and Boulder presented no problems.

Nine flights between 14 August and 07 September 2000 determined the vertical profiles of trace gases above environments defined as crucial to the MOPITT validation. These included: the open ocean, the polluted and background continental troposphere, and areas of biomass burning. All the NOAA profiles were coordinated with the TERRA overpasses. Two flights were arranged for overlap (in space and time) between our profiles, the ER-2 (and MOPITT-A) flights and the satellite overpass. A UV resonance fluorescence detector for continuous measure of CO was sent to Africa for SAFARI-2000. Unfortunately, one of the gases (0.5% Argon in CO_2) needed to run the instrument was delivered from the supplier empty, and it could not be replaced in time for the mission.

MOPITT Validation Exercises:

MOPITT Validation Exercises (MOVE) were designed to compare the various instrument and sampling techniques used in the MOPITT validation (airborne sampling + gas chromatography/HgO reduction, solar absorption FTIR, and airborne remote sensing (MATR)). PRE-MOVE (March 1998) was only a partial success, due in large part, to bad weather. A second MOVE experiment was scheduled for several 2-3 day periods in May 2000.

Experimental planning for MOVE 2000 was plagued by problems associated with aircraft scheduling, and was finally cancelled when the MATR platform (the DOE Citation) was needed for fighting fires in the Southwest.

MOVE 2001 took place at the ARM/CART site during April – May 2001. The goal, as in PRE-MOVE, was to compare several instruments involved in the MOPITT validation [NDSC FTIR (DU, F. MurCray), AERI (UMBC, W. McMillan, MATR (NCAR, M. Smith), and NOAA/CMDL (P. Novelli)]. The CMDL sampling equipment was installed aboard the Citation in order to reach altitudes greater those possible with the CMDL Cessna. Five profiles were collected from about 40,000 ft to 2,000 ft ASL. Surface samples were also collected at the CART site for comparison to the lowest aircraft measurements. These measurements, collected by DOE personnel using CMDL equipment and analyzed in our Boulder lab, provide direct comparison to the CMDL profiles. The surface measurements also provide information needed to estimate tropospheric column CO mixing ratios from the aircraft profiles. These can then be compared to spectroscopic column measurements.

COMPARISON OF CMDL and MATR:

The MATR instrument response and retrieval was studied using co-located flights of NOAA/CMDL sampling with CO measurements made by MATR. Flights above Northern Colorado in June 2000 and June 2001 used the NOAA charter to collect samples. MATR collected spectra from the Citation about one hour later. While the results from the 2 methods were in generally good agreement, the temporal difference in sampling introduced uncertainty to the comparison. Addition comparisons were made over Carr, CO, the CART site, and off the Pacific coast of California, in which the NOAA samples were collected from the Citation.

PERFORMANCE AND DATA:

Although there were occasional problems, sampling frequency was high. Of a total possible 120 flights over 1 year at the 5 anchor sites, 84 successful flights were achieved (69%). This total does not include profiles determined during SAFARI-2000, MOVE experiments, or those flown for comparison to MATR (another 18 flights). During the past year 101 successful profiles have been determined as part of the MOPITT validation program. Unfortunately, not all profiles are acceptable for MOPITT validation studies. Some are disqualified due to MOPITT instrument performance and cloud interferences. Mismatch of flight timing and the satellite overpass was sometimes a problem in early 2000; this was fixed in mid-year.

Our web site (URL = cmdl.noaa.gov/ccg/mopitt.html) outlines the work CMDL is conducting for the validation and provides links to other sites with information on EOS, MOPITT, and SAFARI-2000. The profile results are archived at CMDL along with data from our other aircraft-based sampling programs. The MOPITT data are provided to NCAR on a monthly basis, and NCAR will provide the final data to the NASA archives. These data will also be archived with other CMDL global measurements at international data centers such as the CDIAC and the WDCGG.

VALIDATION OF MOPITT RADIANCES:

The measurements are now being used to validate the MOPITT level 1 and level 2 products (L1 is radiances, and L2 is mixing ratios, respectively). The 8 MOPITT thermal channel signals, which are discussed here, are evenly split between Average (A) and Difference (D) signals. The "A" signals are sensitive to regions between the CO absorption lines, and are insensitive to CO in the atmosphere. Only the "D" channels are sensitive to CO absorption. The 1D and 5D channels are most sensitive to the low-mid troposphere, the 3D and 7D channels are sensitive to the mid to upper troposphere. Both "A" and "D" signals are sensitive to surface temperature. To evaluate the MOPITT radiances, the MOPITT forward radiative transfer model was run using the best available input data. The CMDL profiles were used as the a priori CO profile (extended from the highest measured altitude, ~ 350, mb to 50 mb), along with NCEP meteorological data and surface temperatures from the MOPITT "A" signals. These modeled radiances were then compared to the MOPITT L1. Merritt Deeter (NCAR) provided this overview of the L1 validation studies.

The "D" thermal radiances for the 5 CMDL anchor sites (only for clear sky and through December 2000) and for the SAFARI-2000 campaign (Figure 4) show that the model and satellite radiance agree quite well for 1D and 5D, but less so for the 3D and 7D channels. This may reflect inaccurate extension of the measured profiles to the tropopause. This hypothesis is supported by comparisons between measured and modeled L1 using CMDL profiles determined from the DOE Citation. This aircraft is capable of flight into the lower stratosphere; therefore, uncertainties introduced to the analysis during the extension of the anchor sites are eliminated.

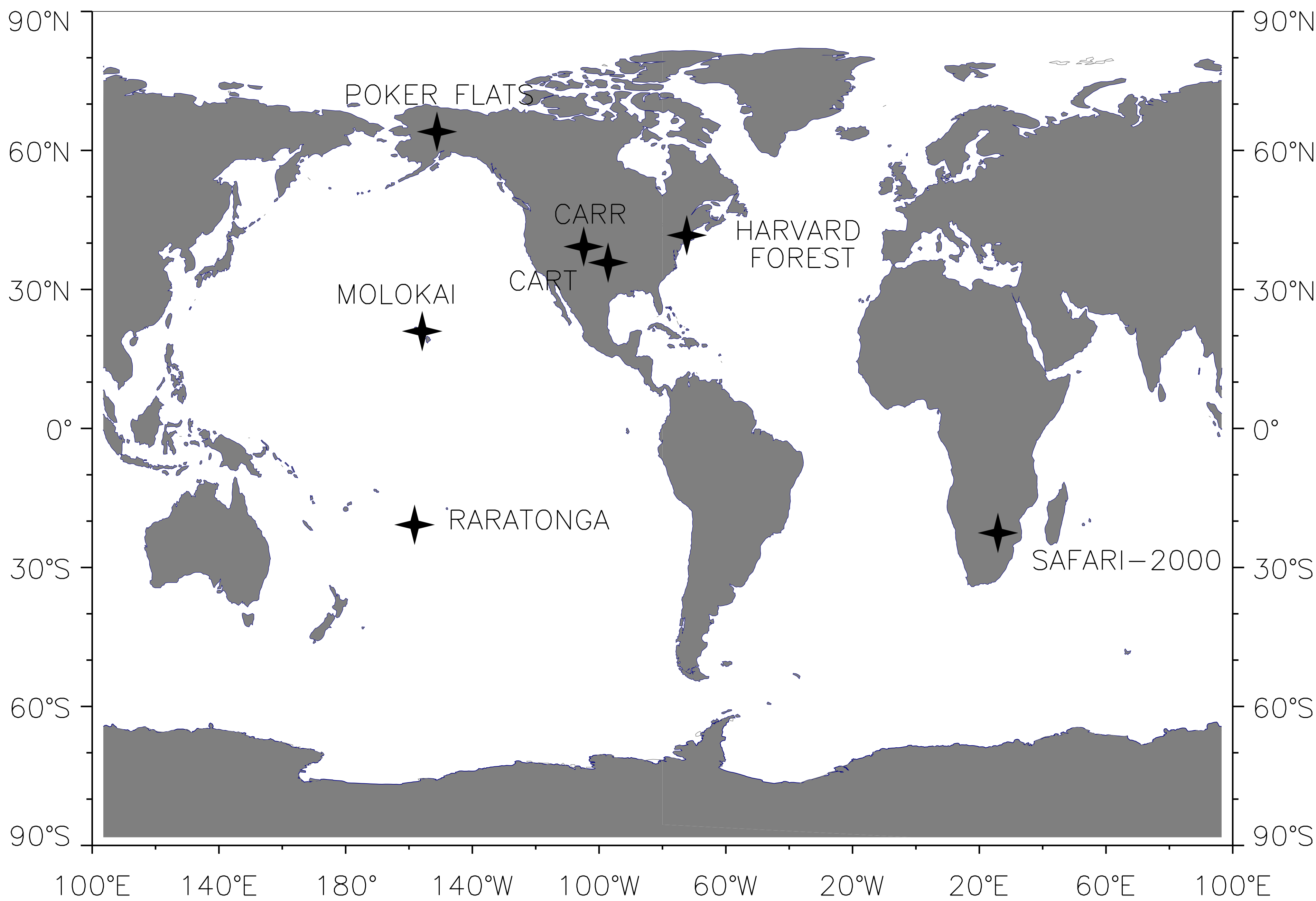
FIGURE CAPTIONS

Figure 1. Locations of the sites used in the NOAA/CMDL MOPITT validation program. The geographical distribution and various proximities to CO and CH_4 sources provide a wide range of environments for the MOPITT validation.

Figures 2: Mixing ratios of CO, CO_2, CH_4, H_2, N_2O, and SF_6 determined as a function of altitude on 08 March 2001 above Harvard Forest, MA. Elevated concentrations of traces gases emitted by industrial/technological sources are evident in the boundary layer. Each box represents the result for a single air sample.

Figure 3. CO mixing ratios above 4 MOPITT sites during 2 weeks in January 2001. Mixing ratios are generally greater in the Northern Hemisphere than in the South. CO measured at surface sites in the NOAA/CMDL surface program near the MOPITT sites (red dots) agree well with boundary layer measurements made from aircraft. Enhanced CO at ~4 km in the tropics may reflect seasonal biomass burning in the equatorial latitudes.

Figure 4. Comparison of modeled and measured radiances. A) Modeled radiances calculated using profiles from the 5 NOAA anchor sites plus SAFARI-2000, B) Profiles from high altitude flights determined from the Citation. The few high altitude samples suggest better agreement with the model compared to flights extending into the middle troposphere.


POKER FLATS
CARR
CART
HARVARD FOREST
MOLOKAI
RARATONGA
SAFARI–2000
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60°N
30°N
0°
30°S
60°S
90°S
100°E
140°E
180°
140°W
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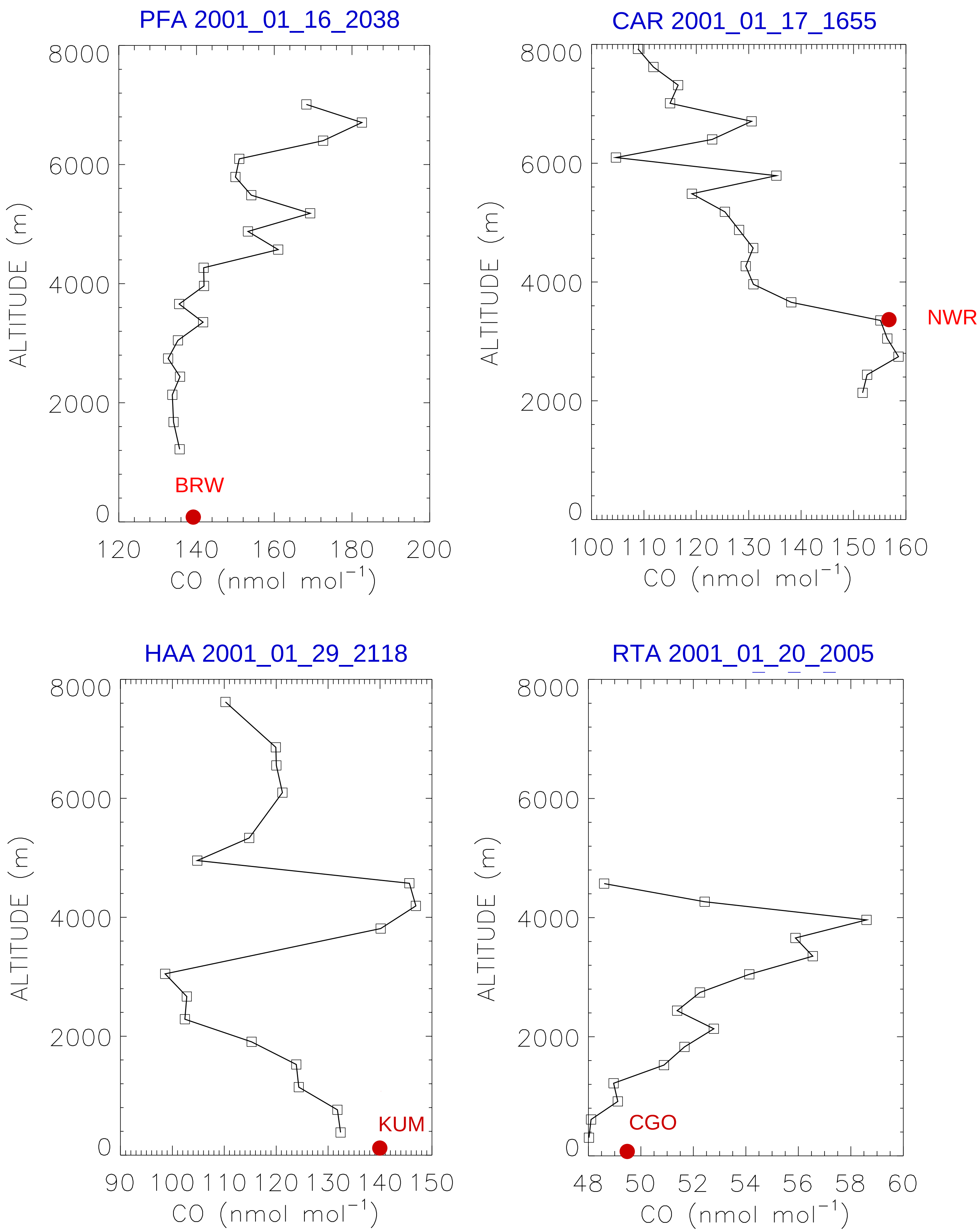
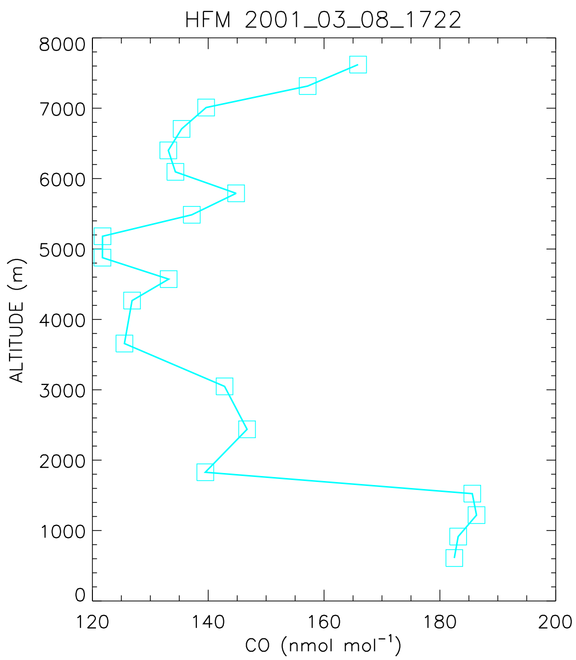
20°W
20°E
60°E
100°E

CO Mixing Ratios 65° N to 21°S During January 2001

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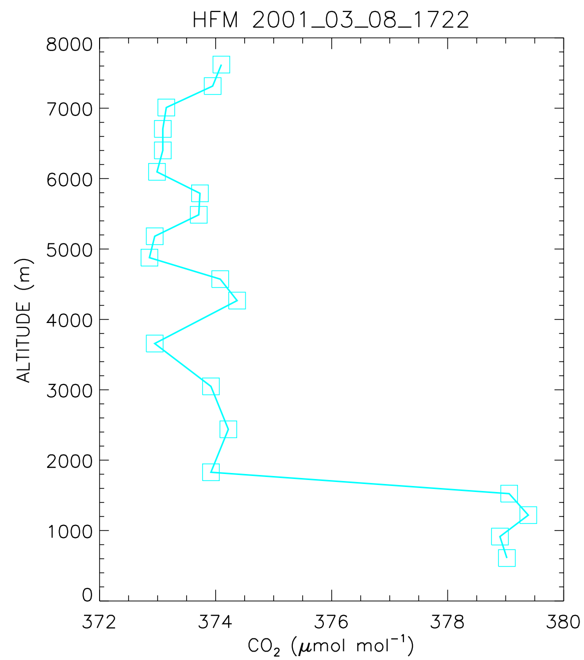
ALTITUDE (m)
CO (nmol mol^{-1})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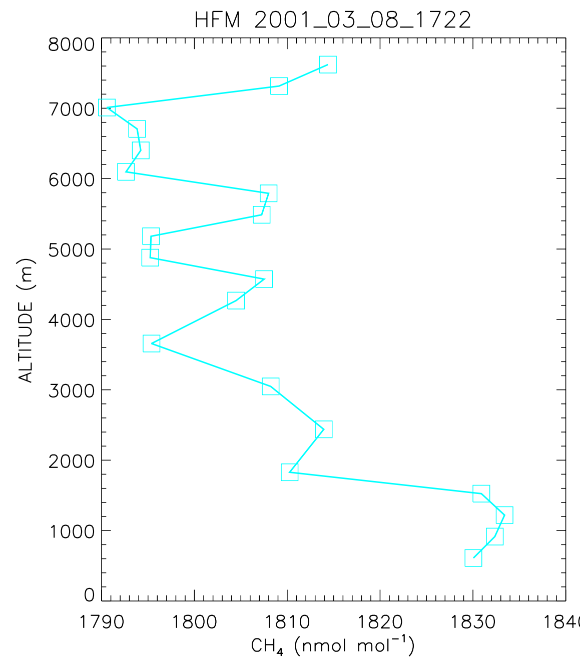
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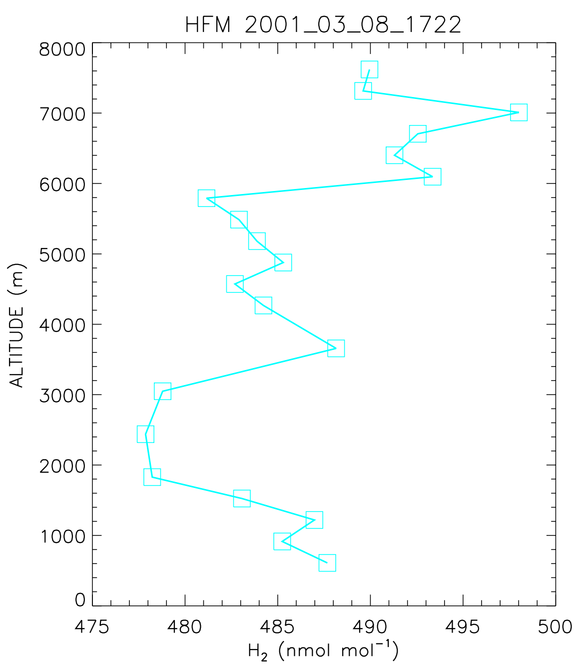
ALTITUDE (m)
CH_4 (nmol mol^{-1})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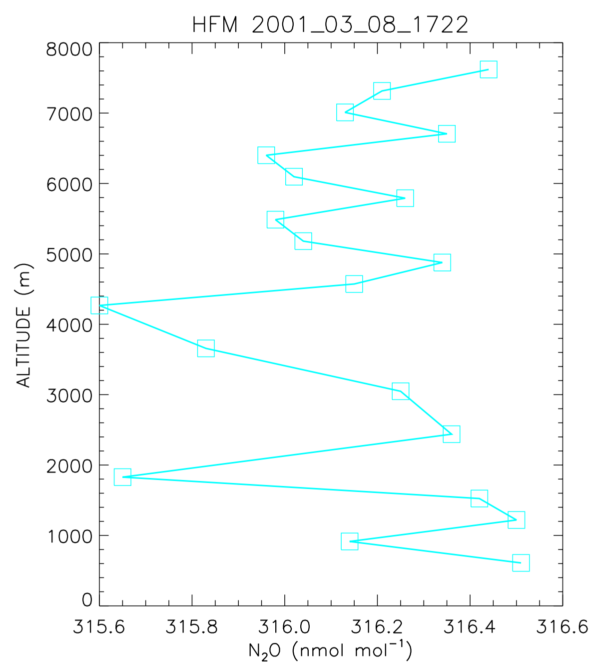
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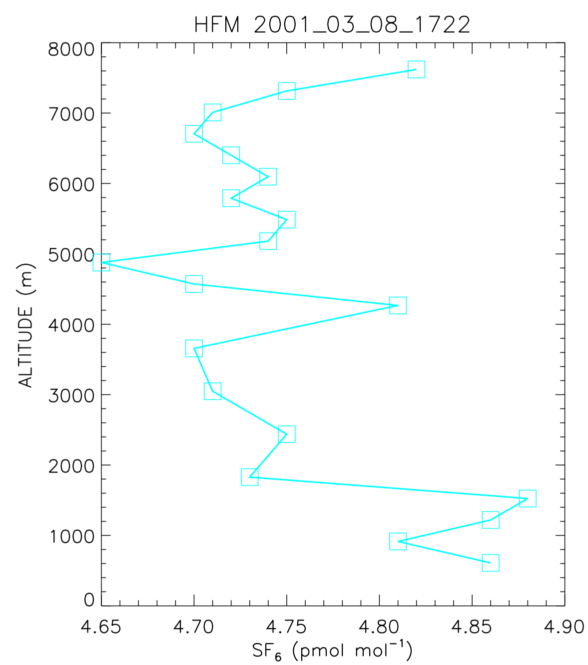
N_2O (nmol mol^{-1})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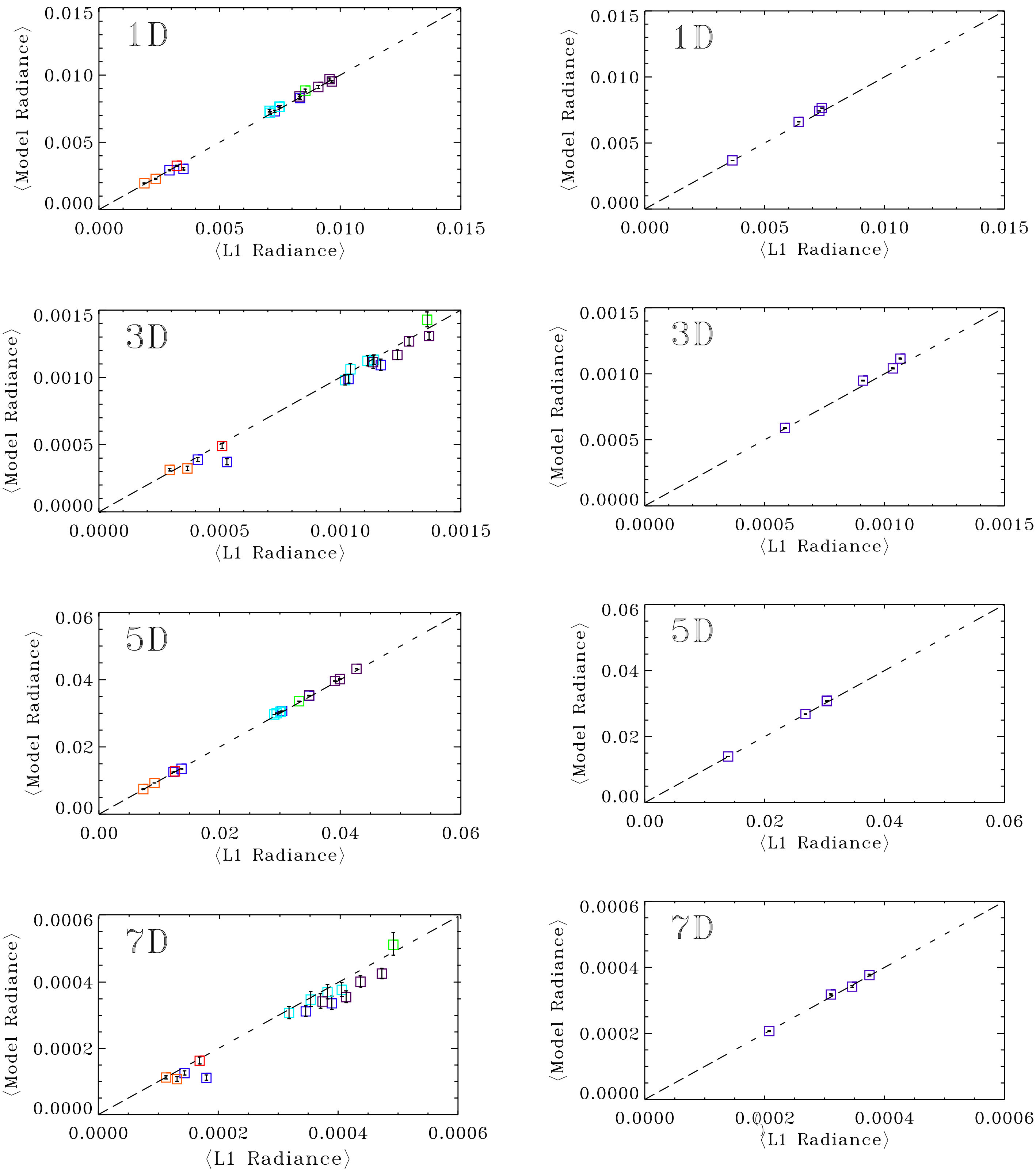
ALTITUDE (m)
SF_6 (pmol mol^{-1})

Validation of MOPITT Radiances

Standard Profiles (Anchor sites, SAFARI-2000)

High Altitude Profiles (Carr, Colorado)



Provided by M. Deeter,
as modified by P. Novelli