MOPITT Validation Using Ground-Based IR Spectroscopy

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ABSTRACT

MOPITT is a nadir-viewing gas correlation radiometer due to be launched aboard the EOS Terra platform. The feasibility of MOPITT data validation using ground-based sun-viewing spectrometers of moderate resolution is investigated. Several instruments with a spectral resolution of approximately 0.2 cm^{-1} are now operating in Russia and in China for the monitoring of CO and CH_4. A spectrometer of this type has been tested and improved at the University of Toronto. It has also been compared with other spectroscopic instruments in field conditions. The results of these comparisons, and the prospects for further work are presented and discussed.

Keywords: carbon monoxide, methane, water vapour, MOPITT, retrieval, validation

1. INTRODUCTION.

MOPITT is a nadir-viewing gas correlation radiometer due to be launched aboard the EOS Terra platform. It will provide global, long-term measurements of CO and CH_4 in the troposphere[1]. It will measure these species using Pressure Modulated Cells (PMCs) and Length Modulated Cells (LMCs). While PMCs have been used for several space-based measurements [2], LMCs have only been used for a few ground-based studies [3], with little existing inter-comparison data to validate their performance.

The MOPITT radiometers are sensitive to several parameters that vary significantly across the globe. Key parameters include surface reflectivity, interfering gasses (water vapour, N_2O, O_3), clouds, aerosols, and the atmospheric temperature profile. Consequently, a large validation program is planned to identify and correct any discrepancies in the MOPITT data. This validation will be carried out by a variety of ground-based and air-borne instruments. Given the varying vertical sensitivity of the MOPITT instrument, air-borne sampling with subsequent gas chromatographic analysis is the most accurate technique for the present time. This delivers high vertical resolution measurements without the uncertainties inherent in retrievals based upon remote sensing techniques. However, current *in-situ* sampling is limited to below a ceiling of 9-km altitude, and it is also quite expensive. Only a few sites could reasonably be funded for this sampling. These sites might well supply sufficient data to validate MOPITT measurements of methane, since methane is distributed much more evenly in the Earth's atmosphere than carbon monoxide. However carbon monoxide shows significant temporal and spatial variability[11] and will require much wider global coverage.

FTIR instruments at the NDSC (Network for a Detection of Stratospheric Change) stations will be used for CO validation. They have a high spectral resolution (typically 0.003 cm^{-1}) and are routinely used to monitor two dozen atmospheric trace gasses, including CO and CH_4 [4]. However, there are many regions on the globe where air sampling up to tropopause height and FTIRs are both absent. Our team is involved in improving, characterising and operating medium resolution spectrometers for MOPITT validation. We plan to supply data on CO and CH_4 total column amounts for Euro-Asian and South American sites. This will be done with the expected active participation of Dr. E. Grechko (Russia) and Dr. Van Genchen (People's Republic of China), who are making routine measurements in their countries using medium resolution spectrometers (Sarcophagus) designed by the Institute of Atmospheric Physics, Moscow, Russia. One of the instruments of this type is currently in Canada. It has been improved and compared to other instruments. There are plans to deploy it in Patagonia, Southern Argentina, at a newly organised monitoring station. It will be operated by personnel from the University of Buenos-Aires (Dr. P.O. Canziani). This report contains a description of the instrument and the improved spectral fitting algorithm, as well as several results of comparisons against other instruments.

2. INSTRUMENT AND RETRIEVAL

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Sarcophagus is a solar tracking Ebert-Fastie type grating spectrometer. It has a spectral resolution of approximately 0.2-0.3 cm^{-1} between 2.8 and 4.8 microns, a focal length of 822 mm and a 15 x 14 cm grating[5]. It has been in operation since 1973, and there are similar instruments still in operation in Russia (Zvenigorod, near Moscow, Kislovodsk, Caucasus)[6], and in China, near Beijing.

The synchronous scanning motor was replaced by a micro-stepping drive; an old lock-in amplifier of Russian production was replaced by a Stanford Research Systems SR510 single-phase analog lock-in amplifier; data acquisition is now through GPIB connection to a PC with LabVIEW software.

Previously, retrieval was carried out by calculating the equivalent width of the CO R(3) line and interpolating this against a forward calculation for the equivalent width (EQW) as a function of column amount. This technique provided reliable results and many important studies have been carried out using this method [7]. The main disadvantage of this approach is that it is one-parameter fitting. This requires one to assume constant reasonable values for several other parameters (spectral resolution, interfering gasses, etc.), and to apply some post-hoc correction for humidity.

A new retrieval algorithm has now been developed using non-linear least squares minimisation. We now simultaneously fit for concentrations of several gases in addition to carbon monoxide; H_2O absorption being the most important interfering species. We assume the so called "refmod 95" [4,6] set of standard profiles for N_2O and ozone, and the solar radiation spectrum measured from orbit by the ATMOS instrument [8]. The forward line-by-line calculation is carried out by the GENASIS program[16] with $0.005cm^{-1}$ spectral resolution, and a vertical resolution of 1 km up to 25 km altitude, and 5 km vertical resolution to 105 km. The a priori CO profile is 100 ppbv up to 12 km altitude, and 40 ppbv above. This profile is then scaled to determine the total column amount. The a priori H_2O profile is taken from balloon soundings where available, otherwise we use the refmod 95 profile. The spectral resolution of the instrument depends slightly upon the alignment of the spectrometer; therefore we need to retrieve the instrument function width. The shape of the instrument function was measured several years ago using a blue line of a mercury lamp in the 6th diffraction order. We scale this measurement by a retrieved stretching factor. Work on a more precise characterisation of the instrument function is currently under way [9]. We also retrieve two parameters to determine the wavelength axis (intercept and slope) and a 100% transmittance signal level (gain and slope). We use the Levenberg-Marquardt minimisation algorithm as coded for MATLAB.

Figure 1 shows a spectrum recorded on June 9 in Egbert Ontario, alongside the fitted spectrum and residual. A persistent residual in our retrievals indicates that we do not have the correct instrument function shape (it is slightly more asymmetric than the original measurement), and we plan to re-measure this curve. Despite this, we still achieve RMS residuals less than 1.5% of the 100% transmittance signal. Each retrieval takes roughly 2 minutes to perform.

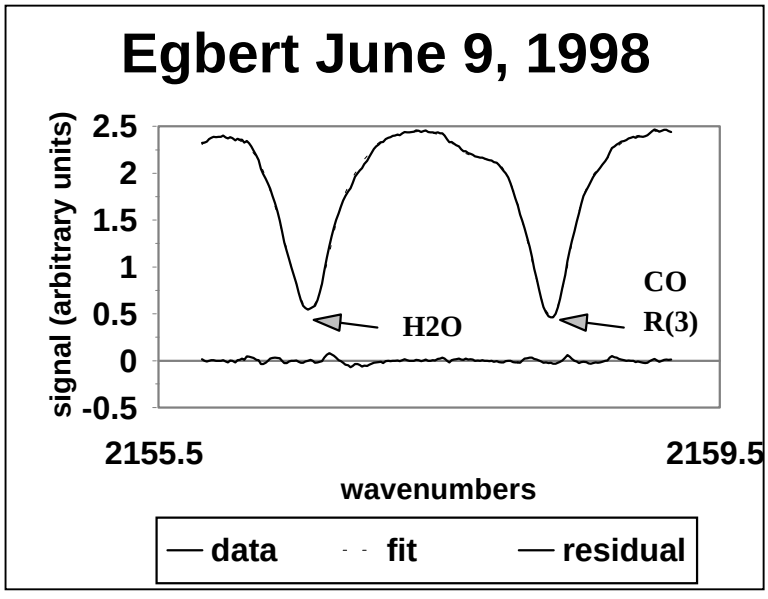

Figure 1. A spectrum recorded by Sarcophagus at 11:46 AM EST, with the fitted spectrum and residual. There is a water vapor absorption feature near 2156.6. cm^{-1}. The RMS residual is 0.88%.

For comparison, an FTIR spectrum is shown in Figure 2. The apparent asymmetry in the CO R(3) lineshape is due to an overlapping H_2O feature at 2158.1 cm^{-1}.

3. VALIDATION CAMPAIGNS

In preparation for MOPITT validation, we have participated in several pre-launch validation campaigns. The purpose of these campaigns is to understand how measurements made by different ground- and air-based instruments compare with each other before comparing their data products with MOPITT itself. These campaigns also serve as test runs of the equipment, when we can optimise their operation.

One such campaign was conducted at the Atmospheric Radiation Monitoring Climate And Radiation Testbed (ARM CART) in Lamont

Oklahoma from March 3 to March 10, 1998 [10]. During this Pre-launch MOPITT Validation Exercise (Pre-MOVE) there were four CO monitoring instruments deployed in addition to our grating spectrometer. SORTI (Solar Radiance Transmission Interferometer) is a sun tracker equipped Brucker 120M with a resolution of 0.0035 cm^{-1} which measures the atmospheric absorption of solar radiation, and is operated by the University of Denver, with support from CART personnel. AERI (Atmospheric Emitted Radiance Interferometer) is a zenith viewing radiometer manufactured by BOMEM (model MB-100) which measures atmospheric emission. It is supervised by the University of Wisconsin. NOAA conducted flights overhead where they took in-situ gas samples with 500 m vertical resolution between 1 and 8 km. Finally, NOAA provided a portable gas sampling device enabling us to take surface measurements as well. The NOAA samples were analysed using a gas chromatograph equipped with a mercury oxide detector [11].

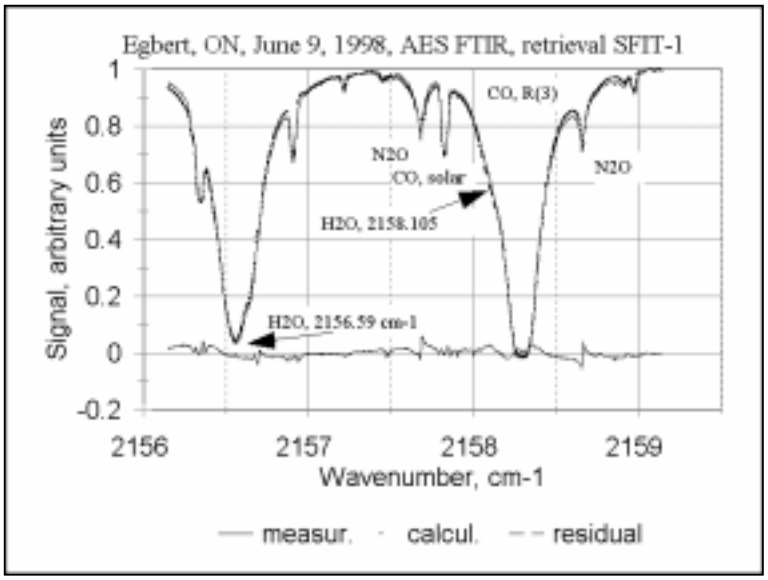

Figure 2. An FTIR spectrum recorded on June 9, 1998, 11:45 AM EST, in Egbert, Ontario, with the fitted spectrum and residual. The RMS residual is 1.5%.

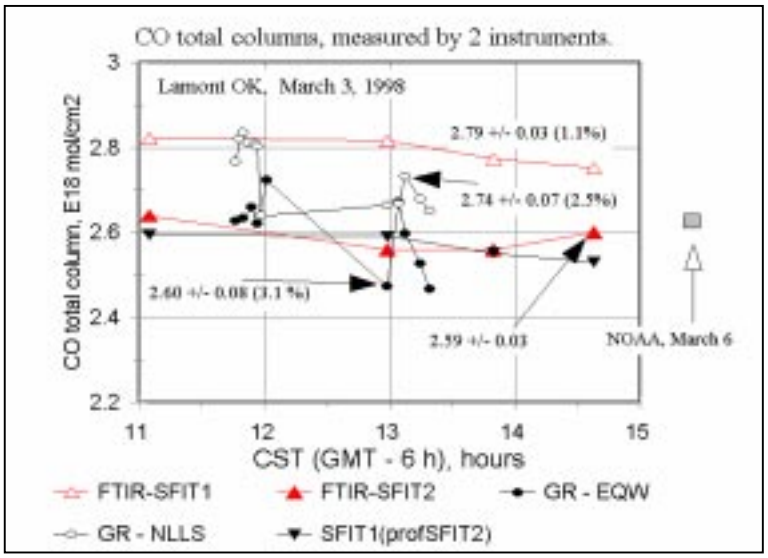

Figure 3. Measurements of CO total column using the grating spectrometer (circles) and FTIR (triangles). SFIT2 revealed a decreasing CO mixing ratio with height, unlike the assumed a priori profile of constant CO in the troposphere. Recalculation with a new a priori profile (upside down triangles) restores agreement between the FTIR retrievals.

To provide temperature profile data for the retrievals, there were balloon soundings four times per day.

From June through August of 1998 the Toronto Sarcophagus was installed at the CARE/AES (Center for Atmospheric Research and Experiments of the Atmospheric Environment Service of Canada) near Egbert, Ontario. Egbert is a remote village roughly 100 miles north of Toronto. Stationed in Egbert is a solar tracking BOMEM DA-8 FTIR with a spectral resolution of 0.003cm^{-1}. Bad weather during that summer permitted just 3 days of concurrent measurements.

4. RESULTS OF COMPARISONS

As discussed previously, the NOAA profile measurements represent the closest reasonable approximation to the true atmospheric state. Unfortunately we did not achieve simultaneous measurements by NOAA and the ground based solar transmittance spectrometers due to cloud cover and equipment malfunctions during Pre-MOVE; though there was a NOAA flight on March 6 -- 3 days after a clear day on March 3. Integration of this profile against the Sarcophagus averaging kernel yields a predicted column measurement of 2.62E18 ± 0.06 mol/cm^2. The actual retrieval using the new NLLS technique produced an average column of 2.74E18 ± 0.07 mol/cm^2 for March 3. So far this is our only non-spectroscopic independent measurement to compare with our data.

The SORTI data were analysed using two different techniques. The SFIT1 analysis [12] is similar to the NLLS technique used for Sarcophagus, though the higher resolution FTIR spectra allows for explicit fitting for interfering gasses such as N_2O, O_3, and solar CO. The newer SFIT2 algorithm [13] uses multi-spectral fitting techniques and statistical estimation to determine the profile shape as well as the total column amount. The retrievals were performed by N. Jones and N. Pougatchev. Note that performing a SFIT1 retrieval using the profile produced by SFIT2 yields a similar column amount

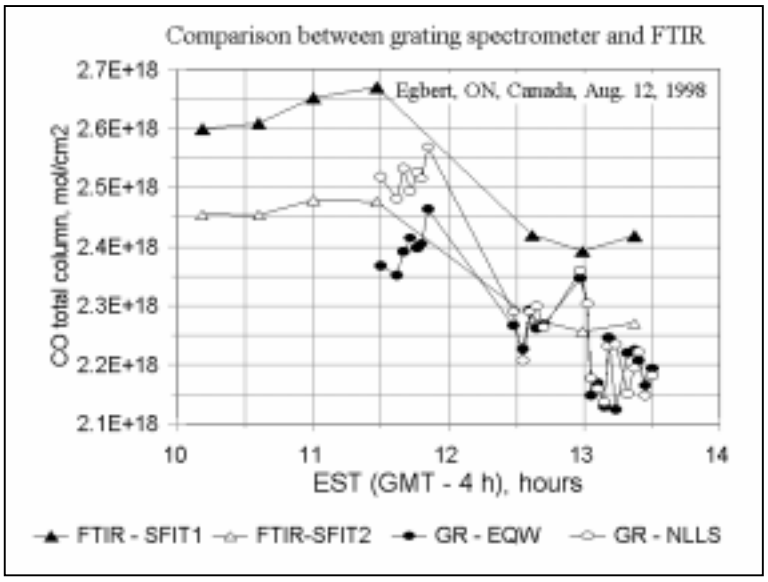

Figure 4. CO retrievals for August 12, 1998. For the EQW technique water vapor total column was kept constant. This explains the minor differences among grating fitting techniques between 11:30 am and noon.

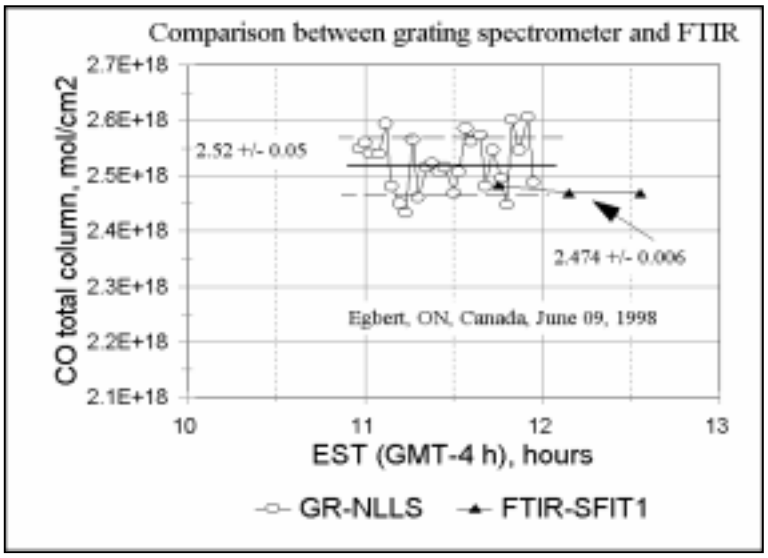

Figure 5. Total columns on June 9, 1998 retrieved with an a priori profile of constant CO in the troposphere, and in the stratosphere (initially 100 ppb and 40 ppb, respectivelyly).

to that from SFIT2, offering confidence in the techniques. The remaining slight discrepancy is likely due to the fact that we used the average profile from the 4 retrievals rather than the 4 different profiles themselves.

As can be seen in the March 3 retrievals, the uncertainty in the shape of the atmospheric profile leads to significant (in this case ~6 %) systematic error in the CO column amount retrieved using the NLLS technique. Obviously this error depends upon the degree to which the actual profile differs from the assumed one. There are some methods to extract information on the shape of the CO profile from medium resolution spectra, and we are currently investigating this possibility so as to improve the absolute accuracy of the retrievals.

Figure 4 displays retrievals from measurements made in Egbert, Ontario, on August 12, 1998. This was a calm summer day, with measured total column of water vapour in the range of 1.2-1.4 g/cm^2. Shown are the NLLS & EQW retrievals for the Sarcophagus data, and the SFIT1 & SFIT2 (profile method) retrievals for the FTIR. The two instruments with their different retrieval techniques show close agreement within the scatter of the data. The changes in CO total column during the day are evident. According to SFIT2 (the retrieval was made by N. Jones) these changes were due to changes in mixing ratios below 4 km altitude.

Figure 5 displays retrievals from Egbert measurements made on June 9, 1998. Again, the NLLS retrievals for Sarcophagus spectra agree within scatter with the SFIT1/FTIR retrievals (SFIT2 retrievals were not available). Comparisons of grating retrieval techniques are illustrated in Figure 6. The retrievals were performed using the EQW technique for the Sarcophagus spectra. The EQW retrievals relied upon the balloon sonde H_2O profile measured above Buffalo, some 200 km to the south, with a water vapour column of 2.44 g/cm^2. The CO tropospheric mixing ratio (squares) shows an apparent diurnal variation, with a minimum around 11:30 AM. This correlates well with an H_2O minimum seen in retrievals performed using the 2156.6 cm^{-1} line (Figure 7). Also shown in Figure 5 are CO columns obtained from an earlier technique[7] (filled circles) which makes a correction to CO depending on the actual water vapour total column for a given spectrum. These retrievals show no diurnal variation; hence, variable water vapour content can explain the EQW diurnal variation.

These significantly lower CO amounts obtained using the 1989 technique are quite understandable. This earlier technique relies upon different line parameters (HITRAN 1986) and a different a priori profile (constant CO throughout the entire atmosphere). Despite these discrepancies it produces a satisfactory correction for changes in humidity. The results of earlier published retrievals[14] are therefore internally consistent. Reprocessing with up to date parameters and a more realistic a priori CO profile should improve the absolute column accuracy, but is not expected to affect reported trends.

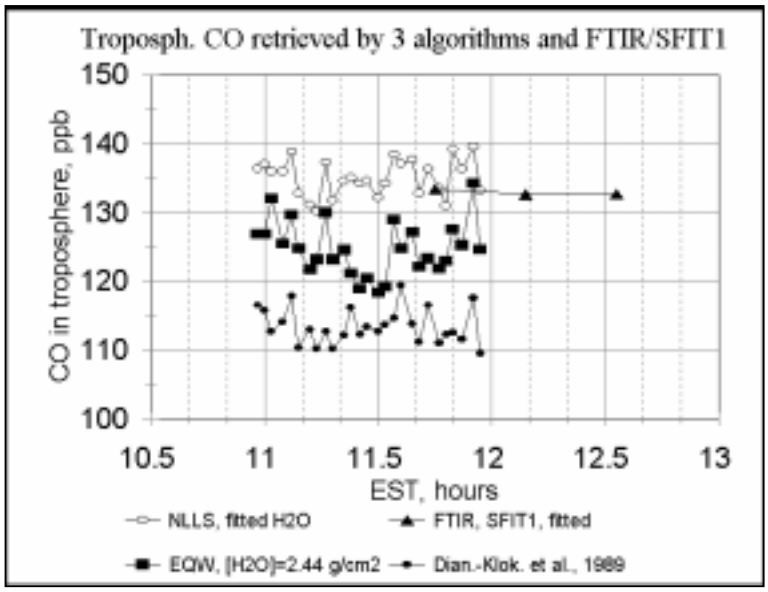

Figure 6. Egbert, ON, June 9, 1998, again using a priori profiles with constant tropospheric and stratospheric CO mixing ratio. For the EQW technique (squares) the Buffalo water vapour profile was assumed.

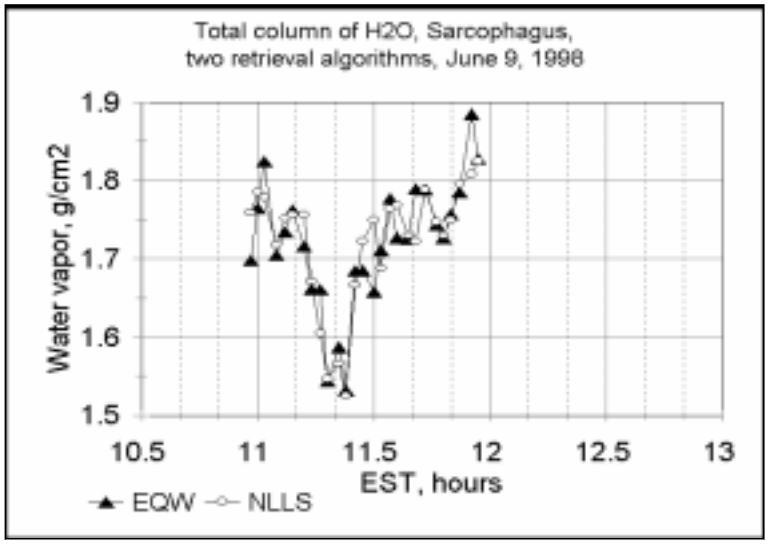

Figure 7. Total column water vapour measured by Sarcophagus. The two retrieval algorithms yield very similar results. Note that these values are significantly lower than for Buffalo (2.44 g/cm^2 according to the sounding).

Fig. 7 displays the H_2O columns retrieved by the EQW and NLLS techniques. The two techniques generate similar retrievals for H_2O. The H_2O variability resembles the CO variability seen in the EQW retrieval using the reference Buffalo H_2O profile (Fig. 4). This can be explained by H_2O contamination of the CO R(3) line. Fig. 2 shows a high resolution FTIR spectrum around the R(3) line. The adjacent H_2O line near 2158.1 cm^{-1} can be seen, but it is not resolved even with a high-resolution instrument. For medium resolution spectra, such as those obtained by the Sarcophagus instrument, the two lines appear as one overlapped feature (Fig 1). The adjacent H_2O line at 2156.59 cm^1 must therefore be included in the fitting interval. Otherwise the fitting is inaccurate due to the overlapping of CO and H_2O lines.

A discussion of the AERI retrievals and its comparisons to other measurements can be found elsewhere[15].

5. DISCUSSION AND CONCLUSIONS

The presented results demonstrate a satisfactory agreement between measurements of carbon monoxide total column amounts using spectrometers of different resolution and design. These limited data indicate that non-linear least squares column retrieval using the Sarcophagus, a grating spectrometer of moderate resolution, yields retrieved column amounts of CO that are within 6% of those retrieved using FTIR interferometers. This difference is well within the claimed accuracy of MOPITT (±10%). Earlier retrieval techniques based upon equivalent width determination are significantly less accurate.

The final objective of the MOPITT project is to supply data, which can be directly compared to *in situ* measurements. Ground-based spectrometers, which are sensing the atmospheric state remotely (as does MOPITT itself), should be considered as important, but not sufficient validation tools. The final decision on the validity of the MOPITT measurements can best be made from a comparison with *in situ* air-borne sampling.

In this respect, the aim of preliminary validation experiments such as Pre-MOVE, and the upcoming Pre-MOVE II (scheduled for August 9-20, 1999 in the Boulder/Denver area) is to establish an agreement between spectroscopic and direct measurements. We have not yet collected sufficient data to comment decisively on these comparisons. This report mainly outlines concurrent observations using spectroscopic instruments. Additional work should be carried out for comparing spectroscopic data with in situ sampling.

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