Final Report

NAG 5 - 6503

Validation of the Measurement of Pollution in the Troposphere (MOPPITT) Experiment by Ground-Based Infrared Solar Spectroscopic Measurements of Carbon Monoxide (CO and Methane (CH4)

By

Dr. Nikita Pougatchev
Christopher Newport University
1 University Place Newport News, VA 23606

NASA Langley Research Center
Hampton, VA 23681

January 22, 2003

Dr. David O'C Starr

EOS Validation Scientist

From: Dr. Nikita S. Pougatchev, Principal Investigator

"Validation of the Measurement Of Pollution In The Troposphere (MOPITT) experiment by ground-based infrared solar spectroscopic measurements of carbon monoxide (CO) and methane (CH4)"

Final Report

INTRODUCTION

The goal of the MOPITT experiment is to enhance our knowledge of the lower atmosphere system and particularly how it interacts with the surface/ocean/biomass systems. The particular focus is the distribution, transport, sources and sinks of carbon monoxide and methane in the troposphere. The MOPITT instrument was launched on EOS TERRA satellite December 18, 1999.

After the launch and until March 22, 2000 the MOPITT instrument was in engineering and calibration mode. Beginning March 23, 2000 through May 6, 2001 the instrument was in a science measurement mode with some calibration breaks. On May 7, 2001 a criocooler on a side B died and channels 1 – 4 became inoperational. The MOPITT resumed its scientific measurements on August 25, 2001 with channels 5 – 8. With some calibration breaks the instrument currently provides the data.

The project has three elements to it: hardware, data analysis and modeling. The MOPITT instrument, on the NASA EOS Terra satellite, measures the upwelling infrared radiance. Using the technique of correlation spectroscopy, information regarding the distribution of atmospheric CO and CH_4 can be extracted. By using appropriate data analysis techniques, concentration profiles of CO are currently obtained on a global basis at a reasonably high horizontal (~22km) and vertical resolution (~3km). Column amounts of methane will be derived over the sunlit side of the orbit. These profiles are assimilated into models to study the chemistry and dynamics of CO, CH_4 and other constituents of the lower atmosphere.

The measurement of CO profiles has been identified as being of primary importance in an effort to improve our understanding of the global system. This has been recognized by the EOS Science Steering Committee - *"The fate of carbon monoxide, remotely detected from space, in conjunction with a few other critical meteorological and chemical parameters, is crucial to our understanding of the chemical reaction sequences that occur in the entire troposphere and govern most of the biogeochemical trace gases"* . This view is supported by the World Meteorological Organization - *"Definition of trends and distributions for tropospheric CO is essential. A satellite-borne CO sensor operating for extended periods could help enormously"*

MOPITT Level 1 data products include: (1) eight calibrated and geo-located instrument difference radiances for each stare (~ 400 ms); and (2) eight calibrated and geo-located instrument average radiances for each stare (~ 400 ms). MOPITT Level 2 data products include: (1) tropospheric CO profiles, which are currently defined as average mixing ratios of five tropospheric layers (1000 — 700 mb, 700 — 500 mb, 500 — 400 mb, 400 — 300 mb, 300 —

200 mb) for each nominal 22-km-by-22-km pixel; (2) total CO for each atmospheric column over a nominal area of 22-km-by-22-km; (3) total CH_4 for each atmospheric column over a nominal area of 22-km-by-22-km. The column amount of CH_4 will only be available on the sunlit side of the orbit as a standard Level 2 MOPITT product.

In any remote-sensing experiment, validation of algorithms and data products is essential to ensure the quality of the data products for archiving and use by scientific communities. To complement the validation activities of each Terra instrument team, the EOS Project Science Office developed a NASA Research Announcement (NRA) that was issued in March 1997. As a result of this NRA, a number of investigators using different instruments and techniques were selected to provide correlative measurements for MOPITT data validation (Wang *et al.* 1998; http://eospso.gsfc.nasa.gov/validation/valinfo.html). One of the selected validation approaches is ground-based solar Fourier Transform InfraRed (FTIR) spectroscopy. This technique is capable to measure CO vertical profiles and CH_4 total column. Observatories equipped with FTIR instruments are operated on a regular basis what makes FTIR technique a valuable tool for a long term MOPITT validation. The list of the observatories is presented in Table 1.

Table 1

Spectroscopic station	Location	Coordinates	Altitude above sea level	Spectrometer
International Scientific Station	Jungfraujoch, Switzerland	47° N, 8°E	3580 m	FT, Homemade
Jet Propulsion Laboratory	Table Mountain near Pasadena, CA	34.2°N, 118.2°W	360m	MARK-IV, Homemade, FT
Atmospheric Environment Service	Egbert, Canada	44.2°N, 79.8°W	251m	FT, Bomem DA5
Institute of Atmospheric Physics	Zvenigorod, Russia	55.2°N, 36.5°E	200m	Homemade, grating
Institute of Atmospheric Physics	Kislovodsk, Russia	43.5°N, 42.4°E	2100m	Homemade, grating
National Solar Observatory at Kitt Peak	Kitt Peak near Tucson, AZ, USA	32°N, 111.5°W	2090m	FT, Homemade
University of Nagoya	Moshiri, Japan	44.4°N, 142.3°E	17m	FT, Bruker 120M
University of Nagoya	Rikubetsu, Japan	43.5°N, 143.8°E	215m	FT, Bruker 120M
National Institute of Water and Atmosphere	Lauder, New Zealand	45°S, 169.8°E	370m	FT, Bruker 120M
National Center of Atmospheric Research (USA)	Sondre Stromfjord, Greenland	67°N, 52.6°E	400m	FT, EOCOM
Alfred Wegner Institute for Polar and Marine Research (Germany)	Spitsbergen, Norway	78.9°N, 11.9°E	10m	FT, Bruker 120M

CO profile retrieval method

The method is based on fitting of the calculated spectrum to the recorded one by means of adjustment to the CO profile and supplementary instrument-related parameters, such as background line, but not the instrument response function. The Rodgers [1976] optimal estimation technique has been used for the inversion. Following the analysis of Rodgers [1990], we have characterized our retrievals and performed an error analysis.

In this section the forward model and inverse method are discussed and an error analysis is performed to evaluate the uncertainties in the retrieved profiles.

Forward model

A multilayer, multi-species line-by-line radiative transfer model developed at the NASA Langley Research Center for the analysis of Fourier transform infrared solar spectra has been used in the present investigation [Rinsland et al., 1982, 1984, 1985]. The key assumptions and characteristics of this model are (1) homogeneous layers in local thermodynamic equilibrium, (2) a Voigt lineshape function computed with the algorithm of Drayson [1976], (3) refractive ray-tracing calculations with subroutines from Gallery et al. [1983], and (4) instrumental line shape function calculations with a Fourier transform technique including the effects of apodization, maximum optical displacement, and the finite field of view. Additional instrumental parameters are included to model, for example, the variation of the instrument response function with wavenumber and wavenumber shifts between the measured and calculated spectra.

Recently, total columns obtained with this forward model and a modified Levenberg-Marquardt nonlinear least squares fitting procedure (the "SFIT" retrieval code) have been reported and compared with values obtained with other algorithms [Zander et al., 1993].

The forward model described above has been extended to allow simultaneous analysis of multiple spectral regions in a series of spectra. However, for the initial study reported in this paper, we have restricted its application to the analysis of one spectral interval in a single spectrum at a time. We used here a total of 29 atmospheric layers, with 1-km vertical thicknesses in the troposphere, increasing to 2 km in the lower stratosphere, 5 to 10 km from 35 km to 80 km, and a final layer from 80 to 100 km altitude. The spectroscopic line parameters given on the 1992 HITRAN compilation [Rothman et al., 1992] have been used.

Inverse method

The measured spectrum may be conceptually described as

$$\mathbf{y} = \mathbf{F}(\mathbf{x},\mathbf{b}) + \varepsilon_y \qquad (1)$$

where $\mathbf{y}$ is the vector of measurements, $\mathbf{F}$ is the forward function,

which relates the true state of the atmosphere and the properties of the observing system to $\mathbf{y}$, $\mathbf{x}$ is the 'state vector' containing the quantities to be retrieved (the CO

profile, the signal corresponding to the 100% transmission level in the spectrum, and instrument related parameters), **b** is a vector containing other atmospheric and instrumental specifications which are not to be retrieved (such as spectroscopic parameters), and ε_y is a vector of direct errors of measurement (noise, etc).

The retrieval employs the method of Newtonian iteration of optimal estimation where the (n+1)st iterate is given by Rodgers [1976 (Eq. 100)

$$\mathbf{x}_{n+1} = \mathbf{x}_a + (\mathbf{S}_a^{-1} + \mathbf{K}_n^T\mathbf{S}_\varepsilon^{-1}\mathbf{K}_n)^{-1}\mathbf{K}_n^T\mathbf{S}_\varepsilon^{-1}[(\mathbf{y} - \mathbf{y}_n) - \mathbf{K}_n(\mathbf{x}_a - \mathbf{x}_n)] \qquad (2)$$

and $\mathbf{x_a}$ is the a priori state vector, $\mathbf{S_a}$ is its covariance, **S** is the covariance of ε_y from Eq. 1, and

$$\mathbf{y}_n = \mathbf{f}(\mathbf{x}_n, \mathbf{b}) \qquad (3)$$

where **f** is a forward model, which is distinguished from forward function **F** because **F** embodies the true physics of the atmosphere (and instrument), while **f** embodies our best attempt to model it.

The first point to note is the generality of this approach. No assumptions are made about the physics in **F** or about the nature of either **x** or **y**. Thus, in our implementation, **y** may consist of measurements in multiple spectral windows recorded at multiple solar zenith

angles and **x** may consist of vertical profiles of several species as well as key instrument parameters.

In practice, the number of elements of **y** is large so that its error covariance $\mathbf{S}_\varepsilon$ is awkward to store and invert; thus we assume that it is diagonal, i.e. that the measurement errors at each wavenumber are independent.

Ideally, the $\mathbf{x_a}$ would represent the mean state of the atmosphere and $\mathbf{S_a}$ its covariance, but in practice neither of these are well known, so we have adopted an approach whereby we use a set of simulated spectra (calculated from actual aircraft in situ CO profiles measurements) to test the sensitivity of the retrievals to both $\mathbf{x_a}$ and $\mathbf{S_a}$, and to enable us to choose values for $\mathbf{S_a}$ which provide satisfactory results for the full range of profiles observed in the atmosphere. This is the 'semi-empirical' modification to Rodgers' technique referred to earlier.

Retrieval of CO vertical profile from ground-based solar infrared spectra is an ill-posed inverse problem. A priori information in form of a priori CO profile and its covariances as well as an assumption about a signal-to-noise ratio are used to stabilize the retrieval. Unfortunately, there is some ambiguity in a process of selection of above mentioned parameters. To avoid any disagreements caused by wrong tuning of the SFIT2 algorithms by different researchers, a workshop of the SFIT2 users (vast majority of them provides the data for the MOPITT validation) was held in Brussels in 2001.

During the workshop a theoretical background as well as practical hands on training were presented to the participants. Lectures on the SFIT2 theory were presented by Clive Rodgers (Oxford University), and practical training was provided by Nicholas Jones and Stephen Wood from NIWA, New Zealand. A copy of source and executable codes of the SFIT2 was distributed to all the workshop participants and the MOPITT validation Co-Is.

VALIDATION

1. Validation of FTIR CO retrievals against CMDL standards.

It is important to have confidence in the correlative data and associated data-processing algorithms before they are used to validate the MOPITT data products. It is also useful to test the intercomparison techniques that are to be used in post-launch MOPITT data validation. The Pre-launch MOPITT Validation Exercise (Pre-MOVE) was a validation campaign at the Southern Great Plains (SGP) Cloud and Radiation Testbed (CART) site of the Department of Energy Atmospheric Radiation Measurement (DOE/ARM) program (Stokes and Schwartz 1994) in Lamont, Oklahoma, March 2-6, 1998. The primary reasons for conducting Pre-MOVE at the CART site are: (1) validate correlative measurement data-processing algorithms by comparing retrieved CO columns and tropospheric profiles from ground-based FTIR interferometer with *in situ* CO profile measurements by the NOAA/CMDL; and (2) test intercomparison techniques and protocols and prepare for future validation experiments after the MOPITT launch in 1999.

The CO and CH_4 profiles were measured by NOAA/CMDL using their aircraft sampling system on March 6, 1998 at the ARM site in Lamont, Oklahoma. The NOAA/CMDL aircraft sampling

unit is an automated package that collects samples of air using a small pump and 20 glass flasks. The samples are then returned to the NOAA/CMDL laboratory in Boulder, Colorado for analysis. Two successful sets of profiles for CO, CH_4, and CO_2 were obtained from 1-8 km with a vertical resolution of ~0.3 km. CO measurements made using the CMDL automated flask system have a typical precision of 1 ppbv (Novelli *et al.* 1994).

Ground-based FTIR remote sensing measurements were made with SORTI FT spectrometer operated by University of Denver. Figure 1 shows the retrieved CO profile using ground-based solar absorption FTIR measurements (SORTI) on March 3, 1998 with a spectral resolution of 0.013 cm. The retrieval was carried out by N. Jones of NIWA and N. Pougatchev of CNU (Pougatchev and Rinsland 1995). The agreement with the airborne *in situ* measurement is within about 10% in the middle troposphere, but the agreement is not that good in the lower and upper troposphere. This could be due to the lack of coincidence (difference of three days) of both location and time for the interferometer observations and the *in situ* measurements.

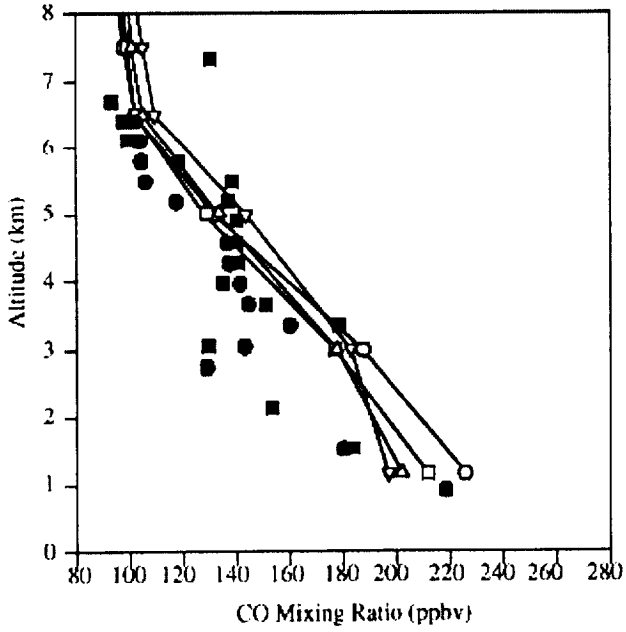

Figure 1.

In Fig. 1 black circles (squares) are the *in situ* CO profile obtained during the morning (afternoon) of March 6, 1998. Open circles, squares, and triangles are the retrieved CO profile using SORTI data (Nikita Pougatchev and Nicholas Jones)

2. MOPITT validation against FTIR measurements.

FTIR measurements were performed during all the MMOPITT's science phases and are in progress now. The FTIR data were used to validate the satellite CO retrievals and some results are presented in Figure 2. Agreement is good and systematic differences between two data sets are statistically insignificant.

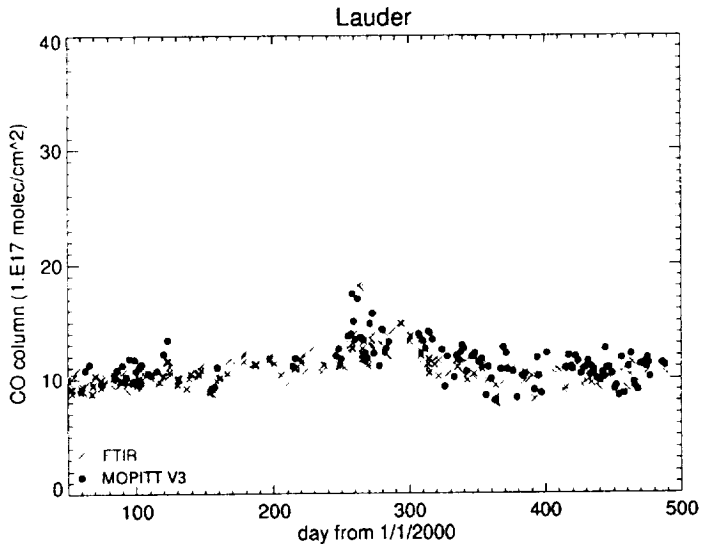

Figure 2a. CO total column measured by the MOPITT (version 3 of Level 2 data processing) and ground-based FTIR technique at Lauder, New Zealand, 45° S, 169.8° E.

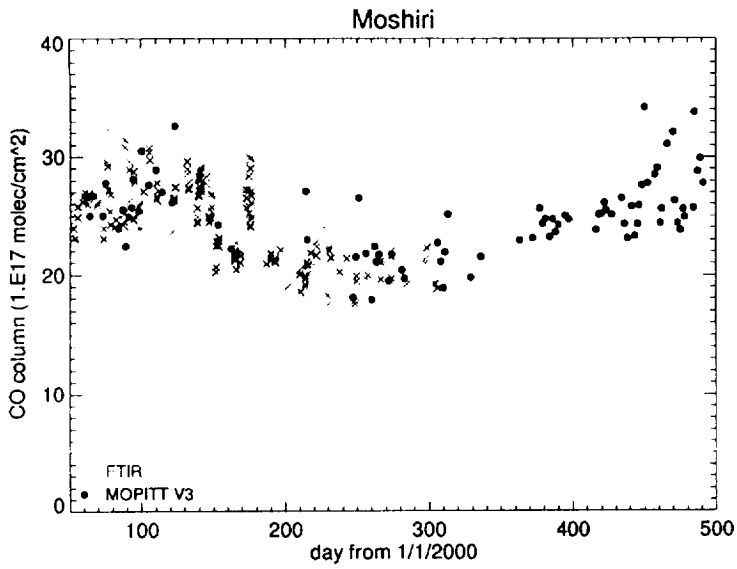

Figure 2b. CO total column measured by the MOPITT (version 3 of Level 2 data processing) and ground-based FTIR technique at Moshiri, Japan, 44.4° N, 142.3° E.

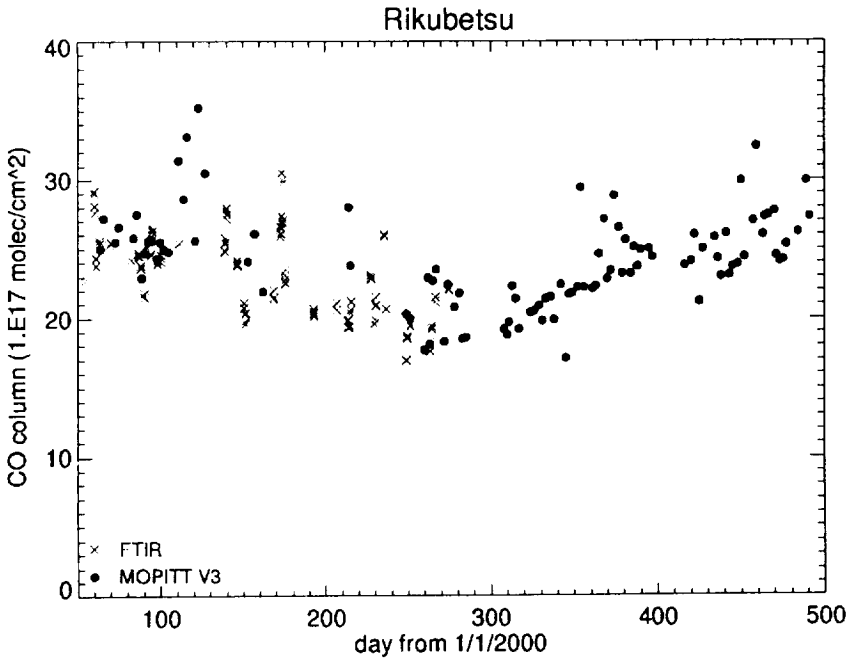

Figure 2c. CO total column measured by the MOPITT (version 3 of Level 2 data processing) and ground-based FTIR technique at Rikubetsu, 43.5° N, 143.8° E.

The displayed results characterize seasonal and irregular variations of the CO total column in the northern and southern hemispheres. Work on the validation of the profile retrievals is in progress now.

The MOPITT instrument continues to provide the data, hence, there is a need for continuing of the validation activity. To preserve the continuity, a joint session of the MOPITT science team and FTIR validation team was held in Boulder in May, 2002. Both teams discussed the acquired experience and problems they encountered. An agreement was reached to continue collaboration as long as there is a need for the validation of the MOPITT Level 2 data products.

References

Drayson, S. R., Rapid computation of the Voigt profile, J. Quant. Spectrosc. Radiat. Transfer, 16, 611-614, 1976.

Gallery, W. O., F. X. Kneizys, and S. A. Clough, Air mass computer program for atmospheric transmittance/radiance calculation: FSCATM, Environ. Res. Paper 828 (AFGL-TR-83-0065), 145 pp., Air Force Geophysics Lab., Bedford, Mass., 1983.

Rinsland, C. P., M. A. H. Smith, P. L. Rinsland, A. Goldman, J. W. Brault, and G. M. Stokes, Ground-based infrared spectroscopic measurements of atmospheric hydrogen cyanide, J. Geophys. Res., 87, 11,119-11,125, 1982.

Rinsland, C. P., R. E. Boughner, J. C. Larsen, G. M. Stokes, and J. W. Brault, Diurnal variations of atmospheric nitric oxide: Ground-based infrared spectroscopic measurements and their interpretation with time-dependent photochemical model calculations, J. Geophys. Res., 89, 9613-9622, 1984.

Rinsland, C. P., J. S. Levine, and T. Miles, Concentration of methane in the troposphere deduced from 1951 infrared solar spectra, Nature, 318, 245-249, 1985.

Rodgers, C. D., Retrieval of the atmospheric temperature and composition from remote measurements of thermal radiation, Rev. Geoph. Space Phys., 14, 609-624, 1976.

Rodgers, C. D., Characterization and error analysis of profiles retrieved from remote sounding measurements, J. Geoph. Res., 95, 5587-5595, 1990.

Stokes, G.M., and S.E. Schwartz, 1994: The Atmospheric Radiation Measurement (ARM) Program: Programmatic background and design of the cloud and Radiation Testbed. *Bull. Amer. Meteor. Soc.*, 75, 1201-1221.

Rothman, L. S., R. G. Gamache, R. H. Tipping, C. P. Rinsland, M. A. H. Smith, D. C. Benner, V. Malathy Devi, J.-M. Flaud, C. Camy-Peyret, A. Perrin, A. Goldman, S. T. Massie, L. R. Brown, and R. A. Toth, The HITRAN molecular database: editions of 1991 and 1992, J. Quant. Spectrosc. Radiat. Transf., 48, 469-507, 1992.

Wang, J., J. Gille, P. Bailey, M. Smith, D. Edwards, J. Drummond, G. Davis, and H. Reichle, 1998: MOPITT Data Validation Plan, Version 4.0. Available from http://eospso.gsfc.nasa.gov/validation/valplans.html.

Zander, R., P. Demoulin, E. Mahieu, G. P. Adrian, C. P. Rinsland, and A. Goldman, ESMOS II/NDSC-IR spectral fitting algorithms. Intercomparison exercise, in Proceedings of ASA Workshop 1993 Reims France, 7-12, Publ. by Univ. of Reims Champagne Ardenne, 1993.