Final Summary of Research: A Climatology of Tropospheric CO over the Central and Southeastern United States and the Southwestern Pacific Ocean Derived from Space, Air, and Ground-based Infrared Interferometer Spectra

P.I.: W. Wallace McMillan (Physics Dept. UMBC, 410 455-6315, mcmillan@umbc.edu)
Co-I.'s: L. Larrabee Strow (Physics and JCET UMBC), H. Revercomb and R. Knuteson (CIMSS UW), and A. Thompson (NASA GSFC)

Submitted to

Phil DeCola
ACMAP Program Manager

1 Summary

This final report summarizes all research activities and publications undertaken as part of NASA Atmospheric Chemistry and Modeling Analysis Program (ACMAP) Grant NAG-1-2022, *A Climatology of Tropospheric CO over the Central and Southeastern United States and the Southwestern Pacific Ocean Derived from Space, Air, and Ground-based Infrared Interferometer Spectra.* Major project accomplishments include: (1) analysis of more than 300,000 AERI spectra from the ARM SGP site yielding a 5-year (1998-2002) timeseries of CO retrievals from the Lamont, OK AERI; (2) development of a prototype CO profile retrieval algorithm for AERI spectra; (3) validation and publication of the first CO retrievals from the Scanning High-resolution Interferometer Sounder (SHIS); and (4) development of a prototype AERI tropospheric O_3 retrieval algorithm. Compilation and publication of the 5-year Lamont, OK timeseries is underway including a new collaboration with scientists at the Lawrence Berkeley National Laboratory. Public access to this data will be provided upon article submission. A comprehensive CO analysis of the archive of HIS spectra of remains as the only originally proposed activity with little progress. The greatest challenge faced in this project was motivating the University of Wisconsin Co-Investigators to deliver their archived HIS and AERI00 data along with the requisite temperature and water vapor profiles in a timely manner. Part of the supplied HIS dataset from ASHOE may be analyzed as part of a Master's Thesis under a separate project. Our success with the SAFARI 2000 SHIS CO analysis demonstrates the utility of such aircraft remote sensing data given the proper support from the instrument investigators. In addition to the PI and Co-I's, personnel involved in this CO climatology project include one Post Doctoral Fellow, one Research Scientist, two graduate students, and two undergraduate students. A total of fifteen presentations regarding research related to this project were delivered at eleven different scientific meetings. Thus far, three publications have resulted from this project with another five in preparation. No subject inventions resulted from this research project.

2 Original Aims and Outcomes

The following five specific aims were set forth in the original proposal with the analysis of IMG data removed in the revised scope of work due to limited funding. Following this list, the project outcome for each of the five aims are presented in brief subsections with appropriate detailed discussion of some of our results.

1. Develop a climatology of free tropospheric and boundary layer CO for the central, southeastern, and mid-Atlantic regions of the United States and the extreme southwestern Pacific Ocean using our existing, validated CO retrieval algorithm[3-5] and archived spectra from HIS, AERI, and IMG. Thus far, we have only analyzed CO abundances for portions of 3 of the total 114 HIS flights and only 10 of the thousands of AERI spectra.

2. Provide public access for the scientific community to our derived CO climatology. This climatological database will be invaluable to the community for comparisons to other CO measurements, comparisons to models, and validation of future EOS instruments.

3. Examine the derived CO climatology for interannual variability of CO where the data permit. The HIS and AERI archives will enable examination of the interannual variability of CO over the southeastern and mid-Atlantic United States during late summer of 1991, 1993, and 1995 and over the central United States from 1993 to the present.

4. Employ trajectory analysis techniques for in-depth analysis of a few specific cases. In addition to the above mentioned interannual variability cases, we will investigate the impact of the south polar vortex on CO abundances using data from 7 HIS flights over the extreme southwestern Pacific Ocean in 1994.

5. With an eye towards future deployments of ground-based and satellite high-spectral resolution instruments, we will investigate the development of a CO profile retrieval algorithm for AERI and IMG spectra.

2.1 Aim 1: CO Analysis of Archival Data

2.1.1 AERI

By shear bulk of numbers of spectra analyzed, our CO analysis of the archive of AERI spectra from the ARM SGP Lamont, OK site was overwhelmingly successful. Prior to this project, only 10 AERI spectra had ever been analyzed for CO abundances. During this project, five years of data from the Lamont, OK AERI alone yielded more than 300,000 spectra (one spectrum every 10 minutes). University of Wisconsin (UW) Co-Investigators were instrumental with access to and analysis of AERI data from the ARM archive as well as ongoing data collection and temperature/water vapor retrievals from the ARM AERIs. Applying a first generation cloud detection algorithm to remove low and thick clouds cut the number of spectra by about half. A more refined cloud and haze detection scheme further reduces the number of good CO retrievals by another 25% to about 15,000 per year. This 5-year CO timeseries is discussed in more detail in a subsequent subsection. At this point, it is sufficient to state that the compiled CO climatology for this site forms a unique resource soon to be available to atmospheric chemistry modelers. Prior to 1998, the routine temperature and water vapor retrievals from AERI spectra were confined to the lowest 3 km. This severely limits the utility of these data for trace gas retrievals where characterization of the entire troposphere is required. Thus, our analysis concentrated on AERI data acquired since January 1, 1998. See the subsection on CO Climatology Analysis under Aim 3 for details.

2.1.2 HIS

Unfortunately, the success with the permanently deployed AERIs thus far has not been duplicated with the archive of field deployment AERIs nor with the archive of spectra from HIS aircraft flights. Due to the untimely delivery of these spectral archives with requisite temperature and water vapor retrievals from the UW Co-Investigators, analysis of this data was not possible within the confines of this project.

Delivery of the archive of HIS and AERI00 data occurred on March 31, 2002, after the end date of this project on March 11, 2002. Moreover, the delivered HIS archive only includes data from 4 of the possible 12 field experiments, and the delivered AERI archive only includes data from 3 of the proposed 12 field experiments but does contain data from 4 campaigns occurring after proposal submission. Concerned with the UW progress early in this project, the PI limited their funds in an attempt to spur action. In the last year, the PI offered to send a UMBC paid employee to UW to expedite archival data processing. This offer was refused. Adding to this disappointment, numerous billing irregularities with the UW sub-contract took nearly one year to resolve concluding with the revelation this project had been improperly billed for another project's expenses. The PI hopes a Masters Student funded by a separate project will perform CO analyses on the ASHOE HIS spectra from the Southwestern Pacific Ocean.

2.1.3 SHIS

Although not specifically mentioned in the original proposal, our subsequent annual reports did envision analysis of IR spectra from additional aircraft instruments such as SHIS and NAST-I as they commenced operation and data became available as part of the archive. Once again, the PI met with resistance from the UW Co-Investigators regarding delivery of SHIS spectra and requisite temperature and water vapor retrievals. Of the six SHIS field deployments each with numerous flights, UW has provided temperature and water vapor retrievals for spectra from only two SHIS flights from the SAFARI 2000 (S2K) field experiment.

These two flights have been thoroughly analyzed by the PI and one of his graduate students in collaboration with 6 teams of scientists involved with 5 different in situ CO instruments flying on 3 other aircraft during S2K and one team of trajectory calculation scientists. While this analysis does not fit in the general climatology theme of this project, the results represent significant achievements paving the way toward future analysis of the burgeoning SHIS data archive and demonstrate the utility of such aircraft remote sensing measurements with subsequent supported analysis.

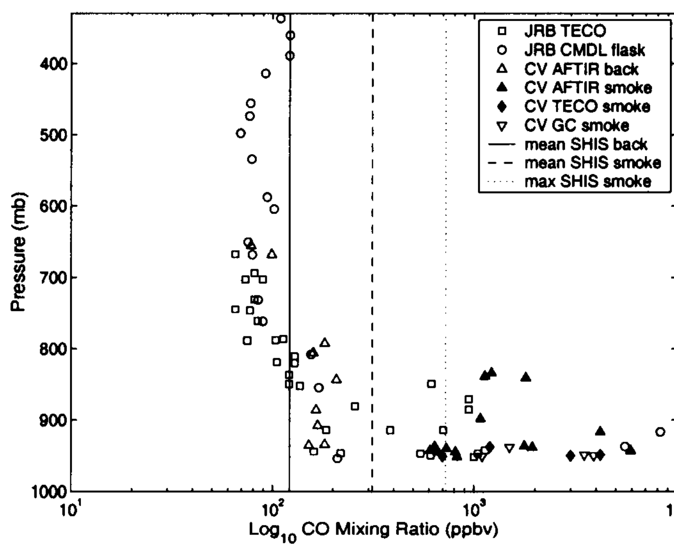

Figure 1: A comparison of all the available *in situ* and remote sensing measurements of CO in the vicinity of the Timbavati fire is presented. Note the good agreement between the JRB TECO, JRB CMDL flask, and AFTIR measurements in background air, pressures < 850 mb and near the AFTIR open triangles for pressures > 850 mb. The larger CO values occurred in the smoke plume and illustrate the temporal and spatial variability of the fire and resultant smoke. The SHIS CO retrieval algorithm uses a constant tropospheric CO mixing ratio profile. Figure 10 of McMillan *et al.* [2003]. Darker background colors indicate lower AI values.

Already accepted for publication in a special issue of JGR dedicated to S2K, highlights of our results from the 7 September 2000 SHIS flight include: (1) excellent validation of column CO retrievals from SHIS by comparison to in situ measurement, (2) detection of biomass burning and associated boundary layer smoke plumes with quantitative CO retrievals, and (3) dramatic spatial variations of column CO due to large scale transport of biomass burning known as the *River of Smoke*.[2] Examples of each of these three are shown

Table 1: Quantitative comparison of SHIS retrieved total tropospheric column CO and CMDL+AFTIR *in situ* measured/extrapolated total tropospheric column CO in the vicinity of the Timbavati fire for both background air and qualitatively for the smoke plume are given. The uncertainty listed for the SHIS background columns represents the 1σ variations in retrieved background columns near the fire and for the several SHIS spectra apparently impacted by the fire and smoke. Table 1 of McMillan *et al.* [2003].

Instrument	background Column CO $\times 10^{18}$ cm^{-2}	smoke plume Column CO $\times 10^{18}$ cm^{-2}
CMDL		15
CMDL+AFTIR	2.1	
SHIS mean	2.3	4.6
SHIS 1σ	0.25	3.5
SHIS max	3.1	15
SHIS min	1.6	2.0

in Figures 1, 2 , and 3 and Table 1. Preliminary results were presented at the First S2K Data Workshop in Siovanga, Zambia with more details shown at the 2001 Fall AGU Meeting in San Francisco, CA and the 2002 Spring AGU Meeting in Washington, DC.

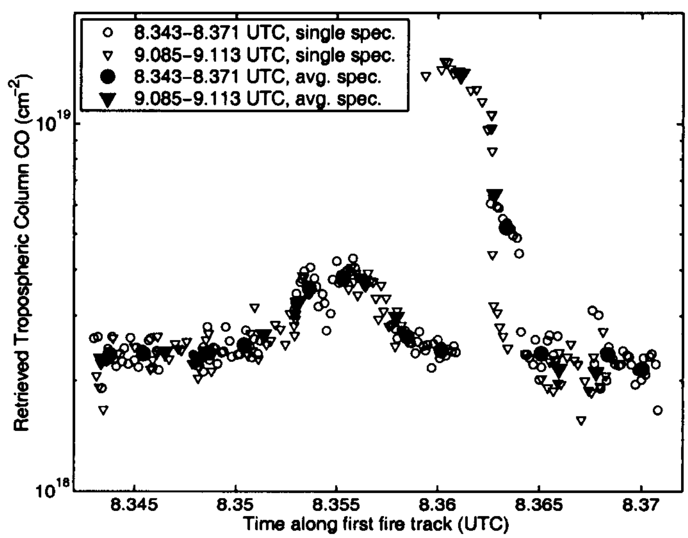

Figure 2: Tropospheric CO columns retrieved from 144 individual SHIS spectra along each of the first and third fire tracks over the Timbavati fire are shown. There is general agreement of the retrievals along the two tracks, including the bump centered at 8.355 UTC. However, the detailed shape of the maximum retrieved CO columns closest to the active fire, 8.36-8.364 UTC, differs substantially between the two over-passes. Also plotted are the corresponding CO columns retrieved from the averages of 12 SHIS spectra along the same tracks. The SHIS FOV locations along the third ER-2 fire track were interpolated to the times of the first track so the observations from these two spatially coincident tracks could be overlaid to illustrate inter-track variations. Figure 5 of McMillan *et al.* [2003].

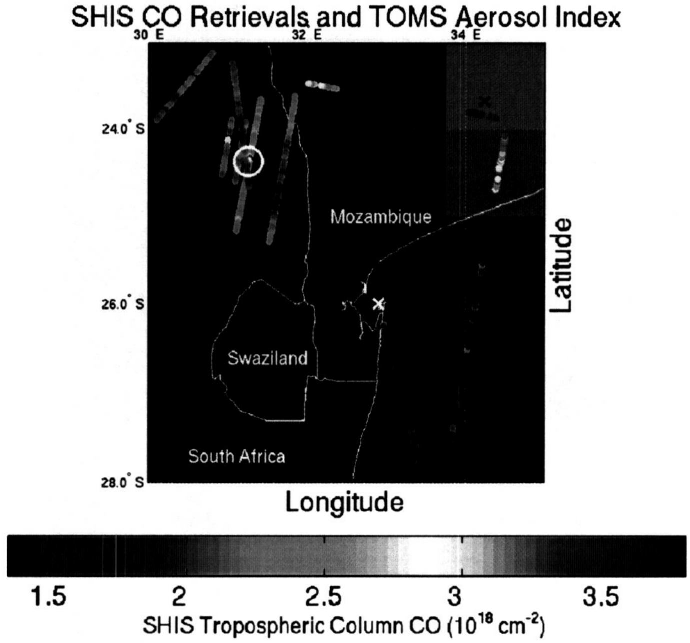

Figure 3: SHIS retrievals of tropospheric column CO for 742 averages of 12 spectra are overlaid in the colored circles on the TOMS AI map. The location of the Timbavati fire lies at the center of the large white circle in the upper left of the image; note the two larger CO columns at the fire. Locations of the three *in situ* CO profiles from the JRA aircraft are noted by the colored X's. White denotes Inhaca Island, blue south central Mozambique, and red northeastern South Africa. Figure 8 of McMillan *et al.* [2003].

Continuing our CO analysis on to the 6 September 2000 SHIS flight, graduate student Michele McCourt is preparing a manuscript describing (1) additional in situ validation, (2) detection not only of biomass burning in CO but retrieval of sub-SHIS-pixel information on the biomass fires themselves, and (3) additional measurements over the *River of Smoke* in east-central southern Africa. Examples of results from (1) and (3) are shown in Table 2 and Figure 4. Item (2) is a matter of ongoing research. Early results were presented at both the 2002 Spring AGU Meeting in Washington, DC and the Second S2K Data Workshop in Charlottesville, VA. Submission for publication is expected by mid-2003.

2.2 Aim 2: Public Release of CO Climatology

Following publication of our initial interpretation and analysis of the 5-year CO timeseries for Lamont, OK, we will make this full dataset publicly available online and we will submit the data to the appropriate NASA DAAC for permanent archiving. As mentioned in the Summary, we presently are engaged in final analyses of this dataset including a new collaboration with scientists at the Lawrence Berkeley National Laboratory who recently started CO in situ sampling at the Lamont, OK SGP site. We expect initial article submission to occur by mid-2003, with data becoming accessible in NetCDF and HDF formats shortly thereafter.

Table 2: Quantitative comparison of S-HIS retrieved total tropospheric column CO and AFTIR *in situ* measured/extrapolated total tropospheric column CO.

	Column Co × 10^{18} cm^{-2}			
	Mongu Airfield		Biomass Burning	
	9:04:48 UTC	9:30:00 UTC	10:12:00 UTC	10:22:12 UTC
Number of Spectra	156	145	19	9
S-HIS mean	4.47	4.63	12.80	7.15
S-HIS 1σ	.280	.250	1.90	.660
S-HIS max	5.51	5.33	15.0	7.19
S-HIS min	3.74	3.95	6.34	5.50
AFTIR	4.67	4.67		

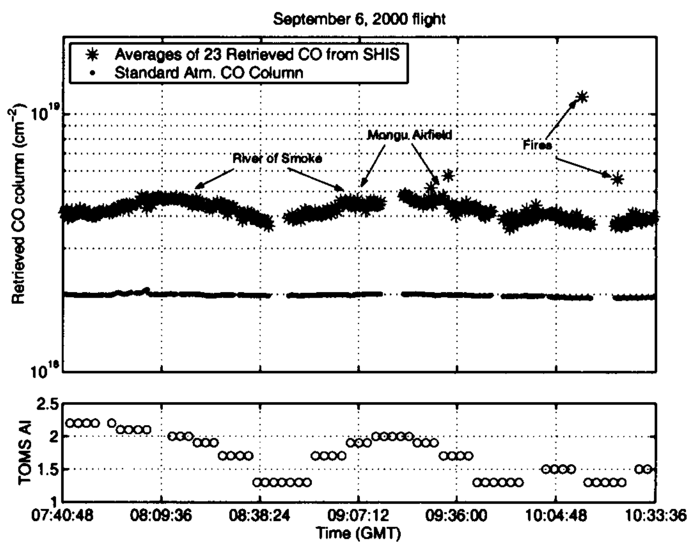

Figure 4: In the top panel the asterisks plot the total column CO retrievals from S-HIS spectra and the dots plot a standard atmosphere CO column for the ER-2 flight. The lack of variability of the dots enforces that the changes seen in the retrieved CO from S-HIS is due to actual changes in CO and not changes in aircraft elevation. Two out-lying points after 10 UTC correspond to fires in the vicinity of Kaoma, Zambia. The bottom panel shows the TOMS Aerosol Index (AI) during the ER-2's flight, approximately 9:50 GMT. The increase in CO between 8-8:30 and 9-9:30 UTC in the top panel loosely correspond to higher AI values in the bottom panel implying the aircraft's encounter with the *river of smoke.*

2.3 Aim 3: CO Climatology Analysis

As previously discussed, insufficient aircraft data (spectra and associated temperature and water vapor retrievals) were available to form any kind of climatological compilation. However, the analyzed AERI spectra from the ARM SGP Lamont, OK site provide a 5-year timeseries of CO showing dramatic variations on daily, synoptic (3-5 day), super synoptic (10-20 day), seasonal, and interannual timescales. UW Co-Investigators cooperated well with access to ARM SGP AERI spectra and temperature and water vapor profiles they process as part of their ongoing DOE ARM research. The first publications of AERI CO retrievals pertain to data from the ARM SGP Lamont AERI analyzed in coordination with MOPITT validation activities.[1,6]

Figure 5 presents the full 5-year timeseries of retrieved tropospheric CO for Lamont, OK with more than 75,000 retrievals. Examples of the different temporal variations are shown in Figures 6 and 7. The most pronounced event evident in the full timeseries occurred in May 1998 and is associated with transport of biomass burning products from extensive brush fires in southern Mexico and Central America across the Gulf of Mexico and as far north as Minnesota as illustrated in the TOMS AI image in Figure 8. Some additional fire events evident in 2000 correlate with back trajectories indicating source regions associated with fires burning near Los Alamos, NM (see Figure5). Separate manuscripts are in preparation to look at the full timeseries and the fire events.

Analysis is underway to quantify the amplitudes and phase of the different timescales present in the data. Complicating this analysis are aperiodic sampling and numerous gaps caused by clouds and other weather events. Many of the cloud-gaps occur with timescales related to expected and observed CO variations. Thus, one cannot simply average or interpolate the timeseries to fill the gaps, nor can one ignore the gaps. By eye, the timeseries show distinct changes in the character of variations, specifically the amplitudes of synoptic and super-synoptic variations appear to be a function of season with their largest amplitudes in late spring and particularly late summer. In addition, a comprehensive back trajectory analysis will soon commence to examine possible source regions over the full 5-year timeseries.

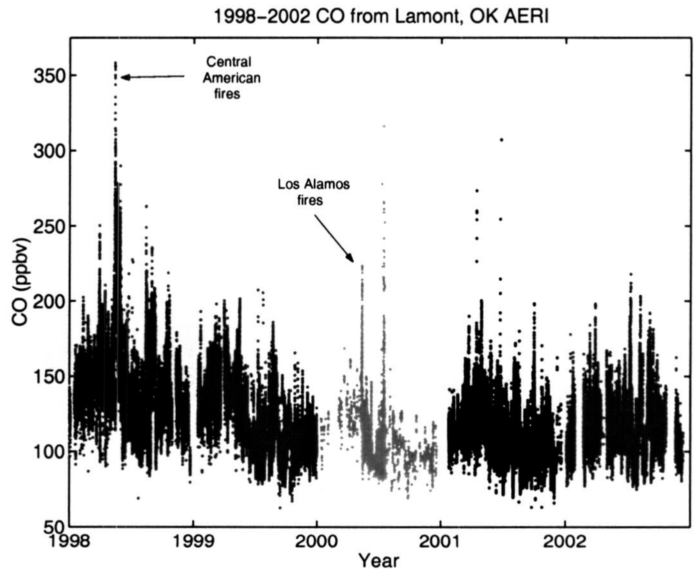

Figure 5: Retrievals of boundary layer CO from the ARM AERI located at Lamont, OK from 1998-20002. Two major fire events are noted.

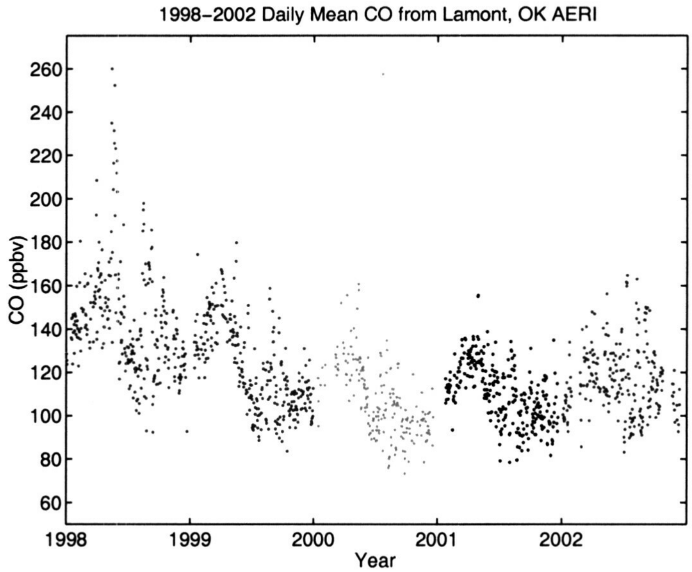

Figure 6: Daily averaged CO retrieved from the ARM AERI at Lamont, OK from 1998-2002. Interannual variations in the seasonal cycle are evident from 1998-2001, less so in 2002.

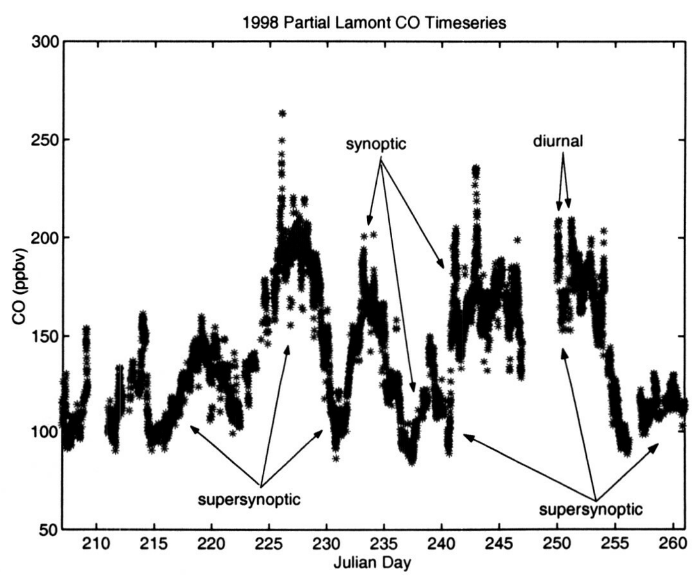

Figure 7: Detailed view of about 50 days of CO retrievals from the Lamont, OK ARM AERI from 1998 showing variations on diurnal, synoptic, and super-synoptic timescales.

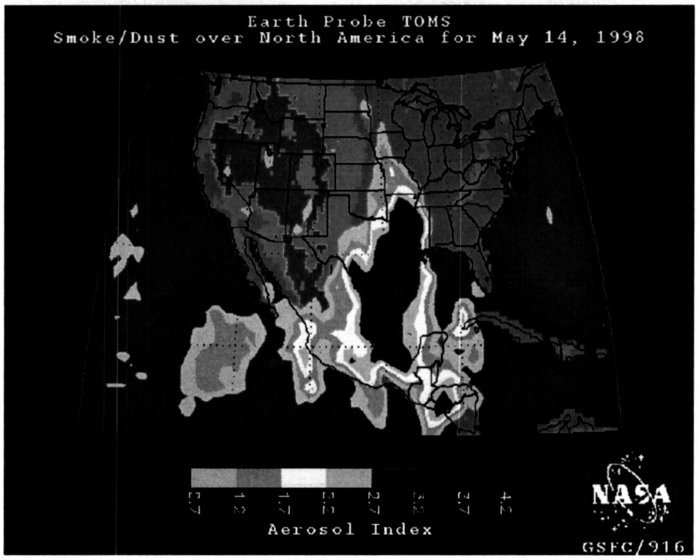

Figure 8: Map of TOMS Aerosol Indices over North America on May 14, 1998, showing transport of biomass burning products from southern Mexico and Central America as far north as Minnesota.

2.4 Aim 4: Trajectory Analysis

The previously discussed SHIS CO results made use of back-trajectory calculations for identification of source regions as originally envisioned in the original proposal. Future analysis of HIS and SHIS data will continue to use these trajectory techniques to assess transport impacts on the observations. As part of the ongoing Lamont, OK CO climatology, we will be undertaking extensive trajectory analyses to identify different temporal variations in source regions on the broad timescales, as well as for specific events such as the Central American fires of 1998.

2.5 Aim 5: CO Profile Retrievals

With the funds not spent by the UW Co-Investigators for archival compilation, the PI directed members of his research team to investigate CO and O_3 profile retrieval algorithms from AERI spectra. The CO profile retrievals were part of the original proposal as stated in Aim 5. O_3 profile retrievals are more risky, but offer great return if successful. Correlations and anti-correlations of tropospheric CO and O_3 indicate different source regions and pollution effects. AERI's ability to provide routine, autonomous monitoring of both these gases in some vertical distribution would be enormously useful to modelers and for air quality assessment.

2.5.1 Prototype AERI CO Profile Retrievals

At the PI's direction, Research Scientist Dr. Gyula Molnar began development of an AERI CO profile retrieval algorithm. As illustrated in this report, development has progressed through sensitivity testing with the algorithm now ready for its first validation tests. After validation testing, we plan to move on to development of a new fast radiative transfer model to enable more rapid reanalysis of all AERI data.

After exploring more standard regression and covariance matrix retrieval schemes, we determined only a physically based algorithm could provide substantial improvement over our previous column based CO

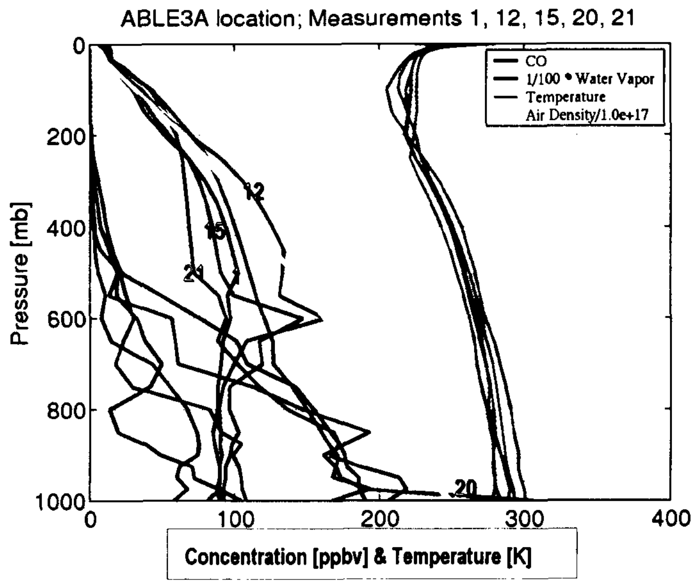

Figure 9: Five of the 21 profiles in the MOPITT training set for ABLE3A are shown as examples of some of the CO variations tested.

retrieval algorithm. For sensitivity studies, a set of 263 CO (and other atmospheric parameter) profiles from the MOPITT training set were used to compute synthetic AERI spectra. This set was further extended with 12 sets of different perturbations each, yielding a total of 3419 profiles for testing. The original 263 profiles are globally distributed from a number of field campaigns providing extreme variations in most atmospheric layers of not only CO but also temperature and water vapor. Five of the profiles from the ABLE3A experiment are shown in Figure 9.

Figure 10 illustrates the spectral changes due to the CO perturbations of Figure!11 with the two lines marked A and B to the high wavenumber side of 2150 cm^{-1} showing different correlations for CO variations in the planetary boundary layer (PBL) vs. the mean free troposphere (MFT). Although all CO profiles do not show such extreme sensitivity, approximately 70% of the 263 profiles do. CO profile retrieval in each layer proceeds by minimizing the differences between computed and observed spectra first with the PBL, then the MFT. For the five profiles shown in Figure 9, CO retrieval results are summarized in Table 3. Although, the large PBL CO abundances in profiles 20 and 21 cause problems with accurate MFT retrievals, the algorithm does identify the large differences between the PBL and MFT.

Sensitivity analyses to inevitable temperature and water vapor profile errors, ± 1K, and ± 10% biases, respectively, demonstrate the robustness of the simple approach described above. These errors are typical of temperature and water vapor retrievals from AERI measurements, alone. Temperature profile biases cause less than 2% retrieval errors, while the water vapor profile errors were, on average, approximately 3%. Next, we plan to assess whether a similar approach could be used to enable retrievals of separate lower and upper PBL and MFT perturbations, thus obtaining a four-layer tropospheric CO mixing ratio profile. In addition, we plan to investigate the use of a Backus-Gilbert approach to assess the feasibility of even higher vertical resolution CO-profile retrievals. We also plan to assess whether higher vertical resolution PBL retrievals are possible using slant path soundings.

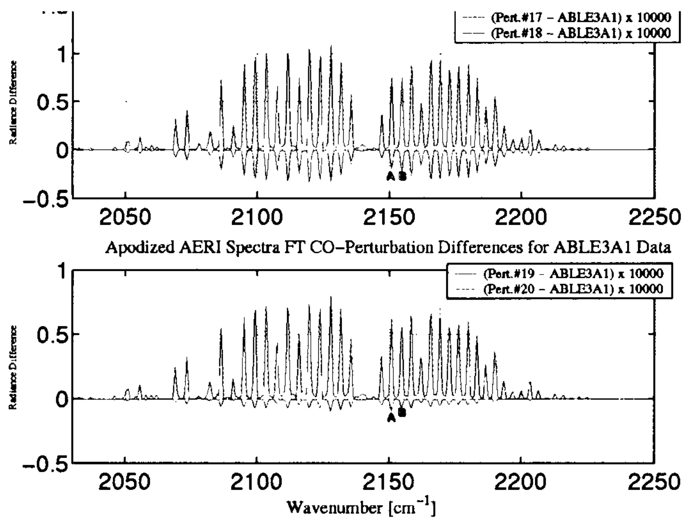

Figure 10: Illustration of the first-step of the CO profile retrieval algorithm demonstrating the sensitivity of compute spectra to the vertical perturbations in CO abundance shown in 11. The two lines marked A and B are discussed in the text.

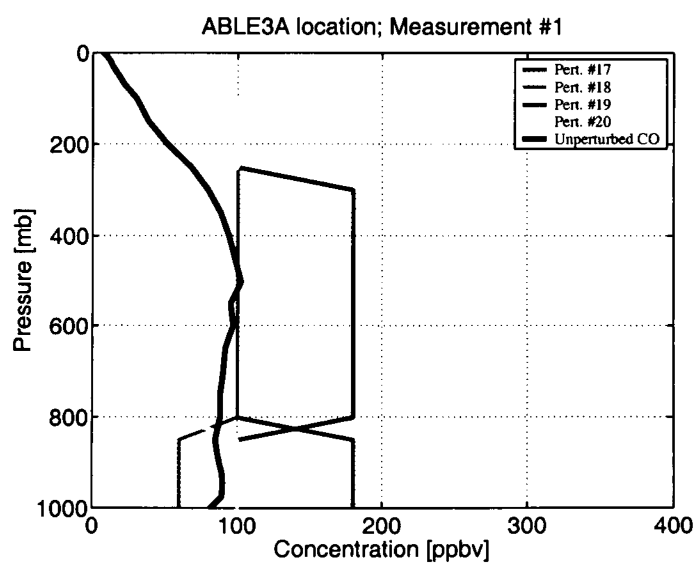

Figure 11: Four of the 12 initial perturbations in CO profiles tested in algorithm development are shown. These four perturbations performed the best in PBL and MFT CO profile retrievals.

Table 3: Comparison of Retrieved and Actual CO abundances in the PBL and MFT for 5 ABLE3A profiles.

ABLE3A profile	Actual PBL CO (ppbv)	Retrieved PBL CO (ppbv)	Actual MFT CO (ppbv)	Retrieved MFT CO (ppbv)
1	88	90	92	88
12	94	100	112	115
15	98	103	100	105
20	198	198	116	123
21	189	180	94	103

2.5.2 AERI O_3 Retrievals

One of the PI's graduate students, Kurt Lightner, started development of a prototype AERI tropospheric O_3 retrieval algorithm as his Ph.D. research. Preliminary results with regression based retrievals have proven quite promising, see Figure 12, and development now is focused on physically-based retrieval techniques with extensive sensitivity testing. As part of another research project, we have obtained spectra with the PI's BBAERI coincident with several ozonesonde launches at the University of Alabama, Huntsville in collaboration with Dr. Mike Newchurch. This data will be invaluable for validation of our AERI O_3 retrieval algorithm. Additional validation datasets may be acquired in the summer of 2003 under other funding. A publication is in preparation summarizing the sensitivity testing and preliminary validation.

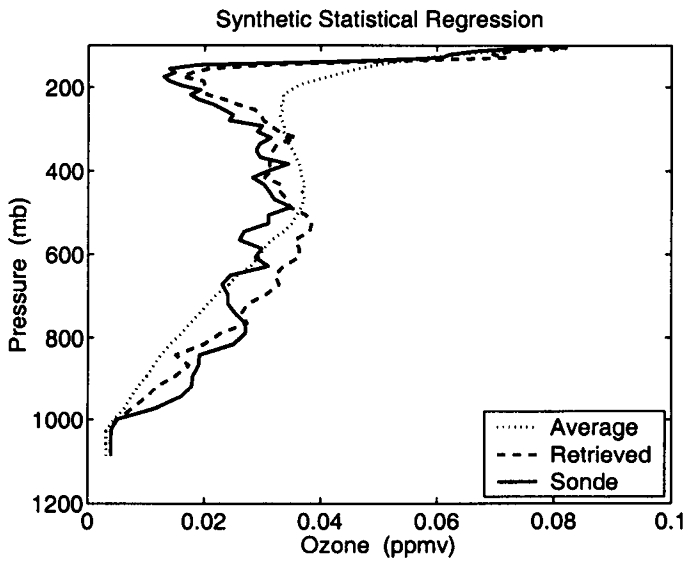

Figure 12: An example result of O_3 retrieval from a synthetic AERI spectrum via statistical regression based on a set of 1000 tropical ozonesondes. The ability of the regression retrieval to deviate from the mean profile and find true structure is impressive, though not precisely accurate.

3 Future Directions

In addition to completing and publishing the 1998-2002 Lamont CO timeseries and associated detailed studies of specific cases, we plan to pursue CO retrievals for spectra from the other 4 ARM AERI's deployed at the SGP boundary sites since 1999. Furthermore, the older AERI archive (prior to 1998) and AERI00 field deployments will be investigated for interesting cases and climatological benefit. Several other AERIs are also in use worldwide including the PI's BBAERI, the M-AERI's at the University of Miami, the Polar-AERI, and another commercial Bomem AERI in Australia. We are developing collaborations with these instrument PIs for future CO data analysis. In addition to CO retrievals from AERI spectra, the PI's research group currently is working on O_3, CO_2, and CH_4 retrievals with ongoing validation and manuscript preparation. The carbon gases are useful for carbon cycle research and satellite validation while CO/O_3 correlations are particularly useful for pollution and tropospheric/stratospheric exchange studies.

As previously mentioned, the PI hopes to have a Masters student analyze the ASHOE HIS spectra and perhaps the other delivered HIS archive data. Analysis will continue with SHIS and NAST-I data as they become available. Particularly any data useful for validating MOPITT, AIRS, and TES.

4 Presentations

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2. McCourt, M., W. McMillan, H. Revercomb, R. Knuteson, R. Yokelson, Remote Sensing Observations of Carbon Monoxide in the River of Smoke on September 6, 2000 and Comparisons to In Situ Measurements, SAFARI 2000 Synthesis Workshop, Charlottesville, VA, 10/7-11/02.
3. McMillan, W. W., M. McCourt, L. Sparling, J. Lukovich, H. Revercomb, R. Knuteson, P. Antonelli, Tropospheric Carbon Monoxide Measurements from the Scanning High-Resolution Interferometer Sounder during SAFARI 2000 on September 7, 2000, EOS, 83, Supplement to No. 19, p. S18, Abstract A21B-02, 2002.
4. McCourt, M., W. McMillan, L. Sparling, J. Lukovich, H. Revercomb, R. Knuteson, P. Antonelli, Intercomparison of In Situ and Remote Sensing Observations of Tropospheric Carbon Monoxide Abundances During SAFARI, EOS, 83, Supplement to No. 19, p. S19, Abstract A21B-04, 2002.
5. McMillan, W. W., M. McCourt, R. Knuteson, P. Antonelli, H. Revercomb, Retrievals of Tropospheric Carbon Monoxide Abundances from the Scanning High-Resolution Interferometer Sounder on-board the ER-2 During SAFARI 2000, EOS, 82, Supplement to No. 47, p. F110, 2001.
6. McMillan, W, M. McCourt, R. Knuteson, H. Revercomb, P. Antonelli, SAFARI 2000 First Data Workshop, Siovanga, Zambia, Invited poster, Retrievals of Tropospheric Carbon Monoxide Abundances from the Scanning High-Resolution Interferometer Sounder on-board the ER-2 During SAFARI 2000, Siovanga, Zambia, 8/28-9/1/01.
7. McMillan, W. W., K. Lightner, G. Molnar, R. Knuteson, W. Feltz, Carbon Monoxide Retrievals from ARM AERIs for MOPITT Validation, In Optical Remote Sensing of the Atmosphere, Optical Society of America 2001 Technical Digest Series, p. 158-160, 2001.
8. McMillan, W. W., H. He, L. Strow, B. Doddridge, R. Knuteson, W. Feltz, Sensitivity Testing of Carbon Monoxide Retrievals from Up-looking FTIR Atmospheric Thermal Emission Spectra, EOS, 81, Supplement to No. 19, p. S160, 2000.

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