ALIAS (ER-2) and ALIAS-II (balloon) *in situ* measurements of N_2O, CH_4, CO, and HCl for SAGE III Validation Studies

FY01 Final Report

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FINAL ANNUAL REPORT:

A1. SUMMARY

Following the successful participation of both the Aircraft Laser Infrared Absorption Spectrometer (ALIAS-I) instrument for the ER-2 aircraft and the Airborne Laser Infrared Absorption Spectrometer (ALIAS-II) balloon instrument in the 1999/2000 SOLVE mission out of Kiruna, Sweden, we have focused our efforts on SOLVE data analysis and instrument improvements. Because this EOS/SAGE-III Validation investigation is a component of research jointly funded by both EOS/SAGE-III and UARP (Upper Atmospheric Research Program), this final report is duplicative of much of the FY01 report sent to UARP.

A2. DATA ANALYSIS

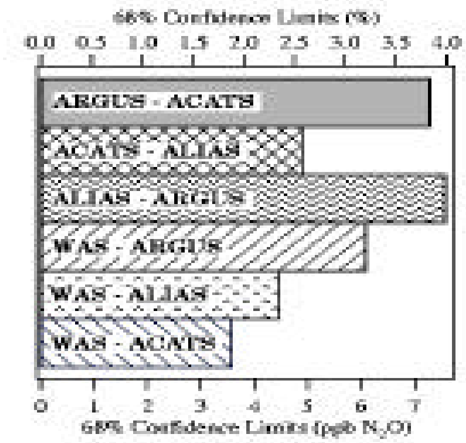
For numerous SOLVE flight data sets, we have re-processed spectral data (80,000 spectra per flight, and 400 Mbyte of data per flight), and conducted tracer-tracer correlation studies, measurement intercomparisons, final laboratory calibrations, flight data set statistics, consistency checks, and resubmitted final data. This has been done for N_2O, CH_4, CO, HCl, and CO_2. Considerable effort was devoted to the production of a unified N2O data set (described below in A3), in which the ALIAS N_2O was the most significant contribution. For several special issue and other publications, we have worked closely with other groups in preparing the results of scientific studies aimed at further understanding atmospheric transport and the PSC chemical processing.

A3. THE UNIFIED N_2O DATA SET PRODUCTION

The focus of our data quality assessment was N_2O, as part of the collaborative effort to create Unified N_2O, an error-weighted combination of *in situ* SOLVE N_2O measurements from ACATS, ALIAS, and Argus. Unified N_2O achieved its goal of better than 3 ppbv agreement with WAS on all but four SOLVE flights, with a typical agreement of 2.9 ppbv (1.5%), better than the typical agreement of any two ER-2 N_2O instruments [Hurst et al., 2001]. ALIAS provided the primary high temporal resolution measurement of N_2O during SOLVE and was thus a critical component of Unified N_2O. According to Hurst et al. [2001], the fractions of Unified N_2O data based on measurements by ALIAS, Argus, and ACATS were 92%, 53%, and 4%, respectively.

Four steps were taken by the ALIAS team to assess the N_2O measurements. First, we carefully examined each flight for correctable problems; none were found. Second, the ALIAS N_2O data from each SOLVE flight were shifted in time by a constant offset to maximize the correlation coefficient with the "expected" N_2O based on relations between ALIAS N_2O and Harvard CO_2.

Each of the N_2O instruments took this step to synchronize their timestamp to within 1 sec. Third, random error ("short-term precision") was estimated by measuring the root-mean-square amplitude of noise on the spectra recorded on the N_2O channel of ALIAS. This amplitude was converted from percent to equivalent N_2O volume mixing ratio, and averaged over each flight. The mean value of the random error was 1.21 ppbv (1 st. dev.) for all SOLVE flights. Since the mean N_2O mixing ratio during SOLVE was 182 ppbv, this translates into a mean measurement "short-term" precision of ±0.65% (1 st. dev.) for ALIAS N_2O. Fourth, we compared ALIAS N_2O with WAS measurements to estimate an accuracy of ±3.5%.



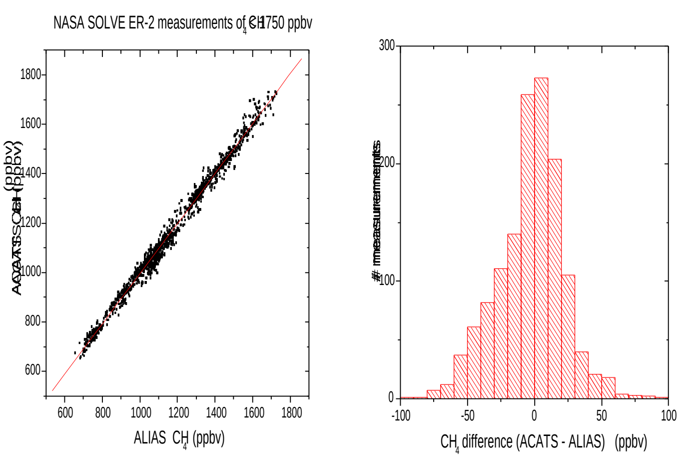
The figure to the left, from Hurst et al. [2001], demonstrates that ALIAS N_2O measurements from SOLVE were in excellent agreement with ACATS and WAS.

A4. THE CH_4 DATA SET

As shown below, there is excellent agreement between ALIAS measurements of CH_4 with those of ACATS during SOLVE (for CH_4 < 1750 ppbv). The linear fit is:

$$CH_4\ (ACATS) = -18.70096 \pm 3.32222 + 1.01358(\pm 0.0028) * CH_4\ (ALIAS) \qquad (1)$$

with a correlation coefficient R = 0.99476 and 1383 total data points. The slope is within 1.4% of a 1:1 slope. Bias between the two instruments is insignificant (<0.3%) at 1750 ppbv CH_4 and grows to typically 1.3% at 700 ppbv CH_4, the lowest CH_4 mixing ratio measured during SOLVE.



In the figure to the left, we show the difference between ACATS and ALIAS CH_4 from all SOLVE flights (10-sec mean data, CH_4 < 1750 ppbv, 10 ppbv-wide histogram bins). For the entire SOLVE dataset, the mean bias between ACATS and ALIAS CH_4 was only –2.9 ppbv, or 0.25% of the mean CH_4 mixing ratio, with a standard deviation of ±25.0 ppbv (±2.2%). This indicates that there was no significant bias between the two instruments, and that they agreed to within 2.2% (1 st. dev.).

A5. THE ALIAS-II DATA SET

The final SOLVE dataset of ALIAS II from the OMS balloon flight of 11/19/99 was prepared and submitted to the project archive. This dataset includes *in situ* measurements of N_2O, CH_4, and HCl.

A6. POST-MISSION ASSESSMENT OF IMPROVEMENTS MADE TO ALIAS-I AND -II FOR THE SOLVE MISSION

Implementation of several instrument modifications produced measurement sensitivities and channel capabilities for SOLVE that were much improved over those of POLARIS for the tracers N_2O and CH_4.

For both ALIAS-I (ER-2) and ALIAS-II (balloon), instrument and data processing modifications were made that increased the tracer (N_2O and CH_4) precision and accuracy for SOLVE over that attained during the POLARIS mission. Improved precision was achieved through instrument modifications that included electronics upgrading (cleaner power supplies, improved signal chains and laser drives) and faster sampling. Improved precision and accuracy were achieved from improved spectroscopic performance, including the reduction of spectral line drifts, the success of a new measurement strategy employing simultaneous recording of CO_2 lines as an in-flight calibration standard, and multi-line parallel data processing.

For SOLVE, spectral line drifts that have affected the performance of ALIAS during aircraft ascent, descent, and dives, were greatly reduced with the installation of a new vertically-mounted dewar that replaced the horizontally-mounted one of the past. The reduction in spectral line drift lead to better precision and accuracy.

For the duration of the SOLVE mission, a tunable Quantum-Cascade (QC) laser was flown on NASA's ER-2 high-altitude aircraft to produce the first atmospheric gas measurements using this newly-invented device, an important milestone in the QC laser's much-anticipated future planetary, industrial, and commercial application. From numerous aircraft flights from September 1999 through March 2000, measurements of methane (CH_4) and nitrous oxide (N_2O) gas were made from the ground to 70,000 ft in the stratosphere as part of a NASA tracer gas intercomparison mission.

Analog electronics originally designed in 1993 were upgraded to decrease background noise on the laser current supplies. Improved filtering and signal conditioning were also incorporated into high gain signal chains. Ultra-low noise transgalvanic signal pre-amplifiers were designed and built using solid state printed circuit boards. Improved layout and packaging of the current supply and the signal chain resulted in reducing jitter in the midpoint of the laser drive ramp from 2 ms down to 0.2 ms. This breakthrough in stability of the laser current supply manifested itself in the precision of the volume mixing ratio obtained by analyzing the second harmonic absorption spectra of atmospheric trace gases.

ALIAS-II was originally designed for installation on a RPV capable of achieving a height of 25 km. Due to development delays in the aircraft it was necessary to retrofit the spectrometer for use on a stratospheric balloon package. Engineering data from the first series of flights of this package revealed the optical stability of the laser beam steering mirrors that inject the beams into the openpath external Herriott Cell is less than satisfactory. At the higher altitudes, up to 32 km, attained by the Observation from the Middle Stratosphere (OMS) payload, return laser power decreased by unacceptable amounts. A complete redesign and build of a more robust optical enclosure was completed, and its improved capability demonstrated in the November 1999 OMS balloon flight out of Kiruna, Sweden.

ALIAS I and II were both fitted with precision digital quartz pressure transducers capable of absolute accuracy of .01%. Comparison with simultaneous measurements using existing capacitance manometers were excellent, with differences typically 0.1 mbar for most altitudes. Critical optical components were also upgraded to improve instrument performance. New Zinc Selenide lenses and windows were installed to improve the transmission of laser light at detection frequencies specific to N_2O, CH_4, CO and HCl.

A7. NEW LIGHTWEIGHT ELECTRONICS

As part of our ongoing effort to improve reliability, ease field operation demands, and reduce cost, we have committed to building improved lightweight electronics (laser current drives, signal chains, digital and analog boards, instrument computer boards etc.) that can be used for all our aircraft instruments (ALIAS, ALIAS-II, WISP, JPL-H2O). The electronics are based on PC104 computer board technology, but take advantage of all our SOLVE improvements (especially signal processing improvements). Rather than have different electronics for each spectrometer, these electronics provide 4-channel electronics that are the same system for ALIAS, WISP, ALIAS-II, and our water spectrometers. This project will be completed by early fall. This exciting development:

- provides interchangeable identical spare boards for all instruments (2- or 1-channel instruments just use one of the available 4-channels) reducing field operations, with cost savings;
- reduces the ALIAS electronics weight from ~40 lb to <8 lb, a huge reduction important for both the WB57F and ER-2;
- should provide improved precision for tracer measurements by providing increased spectral bit-resolution and data storage.
- allows either near-IR (JPL H2O), TDL, or QC lasers to be driven with a simple jumper change on the generic laser drive board;

Most of the boards have been built and populated, and are now undergoing test on the ALIAS instrument.

A8. SCIENTIFIC PUBLICATIONS

Several publications this year have resulted from our collaborations with other groups on SOLVE-related activities.

107. "Quantum cascade laser measurements of stratospheric methane (CH_4) and nitrous oxide (N_2O)", C.R. Webster, G.J. Flesch, D.C. Scott, J. Swanson, R.D. May, W.S. Woodward, C. Gmachl, F. Capasso, D.L. Sivco, J.N. Baillargeon, A.L. Hutchinson, A.Y. Cho, *Applied Optics*, 40, 321-326, 2001.
108. "Arctic ozone loss in 1999/2000 winter – largest local loss on record", M. Rex, et al.

109. "Seasonally-averaged ozone in the northern hemisphere lower stratosphere using a conserved tracer as a reference", M.H. Proffitt, K, Aikin, A.F. Tuck, M. Loewenstein, T.P. Bui, J.J. Margitan, C.R. Webster, G.C. Toon, and J.W. Elkins, submitted to *J. Geophys. Re*s., March (2000).
110. "The detection of large HNO_3 particles in the winter Arctic stratosphere", D.W. Fahey, R.S. Gao, K.S. Carslaw, J. Kettleborough, P.J. Popp, M.J. Northway, J.C. Holecek, S.C. Ciciora, R.J. McLaughlin, T.L. Thompson, R.H. Winkler, D.G. Baumgardner, B. Gandrud, P.O. Wennberg, S. Dhaniyala, K. McKinney, T. Peter, R.J. Salawitch, T.P. Bui, J.W. Elkins, C.R. Webster, E.L. Atlas, H. Jost, J.C. Wilson, R.L. Herman, A. Kleinbohl, M. von Konig, *Science*, *291*, 1026-1031, (2001).
111. "The Construction of a Unified, High-Resolution Nitrous Oxide Data Set for ER-2 Flights During SOLVE", Dale Hurst, Sue Schauffler, Jeffery B. Greenblatt, Hansjurg Jost, Bob Herman, Jim Elkins, Pavel Romashkin, Elliot Atlas, Stephen Donnelly, Jim Podolske, Max Loewenstein, Chris Webster, Greg Flesch, and Dave Scott, submitted to SOLVE/THESEO special issue *J. Geophys. Res*. (2001).
112. "Mean ages of stratospheric air derived from *in situ* observations of CO_2, CH_4 and N_2O", A.E. Andrews, K.A. Boering, B.C. Daube, S.C. Wofsy, M. Loewnstein, H. Jost, J.R. Podolske, C.R. Webster, R.L. Herman, D.C. Scott, R.D. May, E.J. Moyer, J.W. Elkins, G.S. Dutton, D.F. Hurst, F.L. Moore, E.A. Ray, P.A. Romashkin, P.R. Wamsley, S.E. Strahan, submitted to SOLVE/THESEO special issue *J. Geophys. Res*. (2001).
113. "Severe and Extensive Denitrification in the 1999-2000 Arctic Winter Stratosphere", P.J. Popp, M.J. Northway, J.C. Holecek, R.S. Gao, D.W. Fahey, J.W. Elkins, D.F. Hurst, P.A. Romashkin, G.C. Toon, B. Sen, S.M. Schauffler, R.J. Salawitch, C.R. Webster, R.L. Herman, H. Jost, S.C. Wofsy, T.P. Bui, P.A. Newman, L.R. Lait, submitted to SOLVE/THESEO special issue *J. Geophys. Res*. (2001).
114. "Simulation of ozone depletion in spring 2000 with the Chemical Lagrangian Model of the Stratosphere (ClaMS)", Jens-Uwe Groob, Gebhard Gunther, Paul Konopka, Rolf Muller, Daniel S. McKenna, Fred Stroh, Barbel Vogel, A. Engel, M. Muller, K. Hoppel, R. Bevilacqua, E. Richard, R.M. Stimpfle, C.R. Webster, J.W. Elkins, D.F. Hurst, P.A. Romashkin, submitted to SOLVE/THESEO special issue *J. Geophys. Res*. (2001).
115. "Modeling the Effect of Denitrification on Arctic Ozone Depletion During Winter 1999/2000", S. Davies, K.S. Carslaw, M.P. Chipperfield and B.M. Sinnhuber, submitted to SOLVE/THESEO special issue *J. Geophys. Res*. (2001).
116. "Definition of the vortex boundary from an N_2O:theta correlation vs. the Nash criterion: A comparison", Jeffery B. Greenblatt, Hans-Jurg Jost, Max Loewenstein, James R. Podolske, T. Paul Bui, Dale F. Hurst, James W. Elkins, Robert Herman, Christopher Webster, Sue Schauffler, Eliot Atlas, Paul A. Newman, Leslie R. Lait, Melanie Muller, Andreas Engel, Ulrich Schmidt, submitted to SOLVE/THESEO special issue *J. Geophys. Res*. (2001).
117. "Quantifying the rate of heterogeneous processing in the Arctic polar vortex with *in situ* observations of OH and HO_2", T.F. Hanisco, J.B. Smith, R.M. Stimpfle, D.M. Wilmouth, E.J. Lanzendorf, K.K. Perkins, J.R. Spackman, and J.G. Anderson, D. Baumgartner, et al., C.R. Webster, et al., P.O. Wennberg, et al., D.W. Fahey et al., R.J. Salawitch, E.C. Richard, T.P. Bui, submitted to SOLVE/THESEO special issue *J. Geophys. Res*. (2001).
118. "Inorganic chlorine partitioning in the summer lower stratosphere: Modeled and measured [$ClONO_2$]/[HCl] during POLARIS", P.B. Voss, R.M. Stimpfle, R.C. Cohen, T.F. Hanisco, G.P. Bonne, K.K. Perkins, E.J. Lanzendorf, J.G. Anderson, R.J. Salawitch, C.R. Webster, D.C. Scott, R.D. May, P.O. Wennberg, P.A. Newman, L.R. Lait, J.W. Elkins, and T.P. Bui, submitted to *J. Geophys. Res*., *106*, 1713-1732 (2001).

CR Webster, August 14, 2001