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Earth and Space Science

Supporting Information for

Atmospheric Carbon and Transport – America (ACT-America) Datasets: Description, Management, and Delivery

Y. Wei[1], R. Shrestha[1], S. Pal[2], T. Gerken[3], J. McNelis[1*], D. Singh[1], M. M. Thornton[1], A. G. Boyer[1†], M. A. Shook[4], G. Chen[4], B. C. Baier[5], Z. R. Barkley[6], J. D. Barrick[4], J. R. Bennett[7], E. V. Browell[4], J. F. Campbell[4], L. J. Campbell[6], Y. Choi[8], J. Collins[8], J. Dobler[9], M. Eckl[10], S. Feng[6], A. Fiehn[10], A. Fried[11], J. P. Digangi[4], R. Barton-Grimley[4], H. Halliday[4‡], T. Klausner[10], S. Kooi[8], J. Kostinek[12], T. Lauvaux[13], B. Lin[4], M. McGill[14], B. Meadows[4], N. L. Miles[6], A. R. Nehrir[4], J. B. Nowak[4], M. Obland[4], C. O'Dell[15], R. M. P. Fao[16], S. J. Richardson[6], D. Richter[11], A. Roiger[10], C. Sweeney[17], J. Walega[11], P. Weibring[11], C. A. Williams[18], M. M. Yang[7], Y. Zhou[18], and K. J. Davis[6]

[1] Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN.
[2] Department of Geosciences, Atmospheric Science Division, Texas Tech University, Lubbock, TX.
[3] School of Integrated Sciences, James Madison University, Harrisonburg, VA.
[4] NASA Langley Research Center, Hampton, VA.
[5] University of Colorado Boulder/NOAA GML, Boulder, CO.
[6] Department of Meteorology and Atmospheric Science, Penn State University, State College, PA.
[7] Bay Area Environmental Research Institute, Moffett Field, CA.
[8] Science Systems & Applications, Inc., NASA Langley Research Center, Hampton, VA.
[9] Spectral Sensor Solutions LLC, Fort Wayne, IN (formerly at Harris Corporation).
[10] Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany.
[11] Institute of Arctic and Alpine Research - INSTAAR, University of Colorado, Boulder, CO.
[12] Institute of Atmospheric Physics, German Aerospace Center, Kühlungsborn, Germany.
[13] Laboratory for Sciences of Climate and Environment, Paris, France.
[14] Earth Sciences Division, NASA Goddard Space Flight Center, Greenbelt, MD.
[15] Cooperative Institute for Research in the Atmosphere, Colorado State University, Fort Collins, CO.
[16] Science Systems & Applications, Inc., NASA Goddard Space Flight Center, Greenbelt, MD.
[17] Global Monitoring Laboratory, NOAA, Boulder, CO.
[18] Graduate School of Geography, Clark University, Worcester, MA.

[*] Now at Jet Propulsion Laboratory, Los Angeles, CA
[†] Now at Reviewbox, Knoxville, TN
[‡] Now at U.S. Environmental Protection Agency, Washington, D.C.

Corresponding author: Yaxing Wei (weiy@ornl.gov)

Contents of this file

Text S1: Summary of the calibration and QA/QC procedures used by the major instruments and measurements of ACT-America.

Introduction

This document contains supporting Information (Text S1), which provides a summary of the calibration and quality-assurance and quality-control (QA/QC) procedures used by the major instruments and measurements of ACT-America.

Text S1. Summary of the calibration and QA/QC procedures used by the major instruments and measurements of ACT-America.

- **Airborne PICARRO GHG measurements**

In situ CO_2, CH_4, and CO were measured via cavity ringdown spectroscopy using a PICARRO G2401-m analyzer, while H_2O(v) was measured using a similar PICARRO G2301-m analyzer. Ambient air was sampled using a modified Rosemont total air temperature gas sampling probe (Buck Research Inst. LLC) sampling 12″ from the fuselage to avoid the aircraft boundary layer. The flow was split, with one branch proceeding directly to the G2301-m analyzer and the other dried using a PermaPure Nafion dryer (PD-200T-24-MSS). The latter was then compressed with a diaphragm pump (Vacuubrand, Inc.) to a constant pressure of ~1070 mbar maintained using an absolute pressure proportional relief valve (Tavco, Inc.), then sampled by the G2401-m analyzer. An onboard cylinder standard was used to perform hourly in-flight single point offset calibrations of CO_2, CH_4, and CO. This calibration gas was introduced at the inlet with a flow greater than the total system flow so as to avoid pressure disruption. Calibration slopes of CO_2, CH_4, and CO were calculated from weekly three point ground calibrations using cylinder standards. All standards were obtained from NOAA ESRL with concentrations traceable to WMO standards (CO_2: X2007; CH_4: X2004A; CO: X2014A). H_2O(v) was calibrated between campaigns with a water source measured simultaneously by the analyzer and a NIST-traceable frost-point hygrometer (Edgetech).

- **Ozone measurements**

In situ O_3 was sampled with a ½″ OD (3/8″ ID) FEP tube sheathed inside a 5/8″ OD (1/2″ ID) stainless steel forward facing J-probe inlet. This flow was introduced into a 9″ x 1.5″ diameter PFA sampling manifold. Flow exiting the manifold was exhausted from the aircraft through a static exhaust port. Thus, flow through the manifold was driven by the differential between the impact pressure from the forward facing inlet and the static pressure from the exhaust, typically ~30 L/min during flight. O_3 concentrations were measured from air subsampled from the upstream side of the manifold via UV absorption using a 2B Technologies Model 205 analyzer. The air from the analyzer was then exhausted to the downstream side of the manifold to minimize pressure differentials across the analyzer. O_3 background offsets were corrected using zeros measured hourly in flight by scrubbing incoming air with a potassium iodide cartridge. Calibration of the analyzer was performed between campaigns using a NIST-traceable UV photolysis source (Model 306, 2B Technologies).

- **Onboard NOAA Programmable Flask Packages**

NOAA Programmable Flask Packages (PFPs) were sampled in-flight and promptly returned to NOAA's Global Monitoring Laboratory in Boulder, CO for analysis of greenhouse gases, carbon isotopes, halocarbons and hydrocarbon species. A first sample aliquot was analyzed on the Measurement of Atmospheric Gases that Influence Climate Change (MAGICC) system for dry-air mole fractions of CO_2, CH_4, CO, N_2O, SF_6, and H_2 (https://www.esrl.noaa.gov/gmd/ccgg/aircraft/analysis.html). MAGICC gases were calibrated to standard scales maintained at NOAA/GML (Dlugokencky 2005; Hall 2007; Novelli 1991; Zhao and Tans 2006). A second aliquot of sample air was analyzed on a custom-built GC/MS (PR1) system for approximately 50 additional non-methane hydrocarbons, halocarbons, and other sulfur-containing compounds and reported on NOAA absolute calibration scales derived in-house from pure components and high-precision gravimetric techniques. Remaining flask sample air was transferred to the Stable Isotope Laboratory at University of Colorado-Boulder's Institute of Arctic and Alpine Research (INSTAAR) for stable isotopic measurements of carbon dioxide and methane ($d^{13}C$-CO_2 and ($d^{13}C$ –CH_4). For these species, INSTAAR maintains standards that tie sample measurements to the local realization of the VPDB-CO_2 scale (Miller et al, 2002, Trolier et al., 1996, Vaughn et al., 2004). A subset of the ACT-America flask samples has undergone CO_2 graphitization at INSTAAR's Laboratory for AMS Radiocarbon Preparation and Research (Turnbull et al., 2009; Turnbull et al. 2007) and subsequent analysis for radiocarbon ($^{14}CO_2$) by the University of California at Irvine.
Flask data have undergone several quality control measures to assess analysis errors, sampling errors, or storage biases. Analysis errors in measurement systems were detected through drift in target or standard gas measurements or via abnormal initial flask pressure or humidity before during measurement of sample aliquots. Similar to Sweeney et al. (2015), flask sampling errors were identified using onboard PFP data logs used to both record flask mass flow rates and flushing times, and to ensure that the flask target storage pressures were reached for each sample. Potential contamination or leaks in sample lines were identified using indicator species such as CO, tetrafluoroethane (Freon 134a, $C_2H_2F_4$) and bromochlorodifluoromethane (Halon-1211, $CBrClF_2$), emitted from engine exhaust, onboard air-conditioners, and aircraft fire extinguishers respectively (Sweeney et al., 2015). Enhanced mole fractions of the aforementioned gases in flask samples, when unaccompanied by co-enhanced species indicative of anthropogenic emissions plumes, were flagged for potential sample contamination.

- **Cloud Physics Lidar (CPL)**

CPL is a multi-wavelength (355, 532 and 1064 nm) elastic backscatter lidar that enables a comprehensive analysis of radiative and optical properties of clouds and aerosols [McGill et al., 2002]. CPL data have been used for cloud properties analysis [McGill et al., 2003; McGill et al., 2004] and validation of satellite retrievals [McGill et al., 2007; Yorks et al., 2011b; Hlavka et al., 2012]. CPL measures the total attenuated backscatter (e.g., aerosol plus Rayleigh) as a function of altitude at each wavelength. Additional cloud and aerosol properties include the particle depolarization ratio for phase discrimination, lidar ratio, extinction coefficient, optical depth, and backscatter color ratio. Final CPL data product accuracy depends upon the number of laser pulses averaged and the aerosol loading of the atmosphere.

The steps to producing calibrated CPL profiles of normalized relative backscatter (NRB) are: (1) geo-locate the raw CPL data; (2) correct for detector nonlinearity, range, and instrument artifacts; (3) normalize to laser energy; and (4) subtract solar background signal. The ancillary information included in the Level-1 data file is the navigation data from the aircraft and coincident meteorological data (i.e., temperature, pressure, relative humidity). CPL uses Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) reanalysis data for meteorological variables. The calibration method for CPL backscatter data at all three wavelengths is the Rayleigh normalization technique, which normalizes the CPL signal to the actual atmospheric signal from molecular (Rayleigh) scattering [McGill et al., 2007]. The molecular backscatter and extinction coefficients are computed using temperature and pressure from MERRA-2. No in-flight calibration is required. CPL Level-2 data products (i.e., cloud phase, backscatter and extinction coefficients, cloud optical depth, etc.), which are provided in HDF5 format, are derived using the algorithms outlined in Yorks et al. [2011a], Yorks et al. [2011b], and Hlavka et al. [2012].

The CPL QA/QC activities have three stages: (1) in-field data assessment, (2) preliminary QA/QC, and (3) final data products. First, the CPL team applies the calibration and data processing algorithms, as described above, to the raw data acquired during flight. The team produces initial data products within 24-48 hours of each flight. In-field data assessment is performed using browse image analysis (images available at the CPL website http://cpl.gsfc.data.gov). If the browse images pass the in-field QA/QC, the preliminary data products are produced using standard atmospheric profiles for calibration. Once MERRA-2 reanalysis data become available (approximately 1 month after a campaign), the CPL data is reprocessed to produce the final data products. Final data products are examined by the CPL team to assess data quality before the data is archived at the DAAC.

ABL depths have been derived from the CPL backscatter data using a wavelet algorithm (Davis et al, 2000) and quality-checked by hand. This data set is in the process of being documented and added to the ACT-America data archive.

- **Multifunctional Fiber Laser Lidar (MFLL)**

The Multifunctional Fiber Laser Lidar (MFLL) was developed by Harris Corporation and further advanced through significant test flights and collaboration with NASA Langley Research Center, as a testbed for the Active Sensing of CO_2 Emissions over Nights Days and Seasons (ASCENDS) Mission. MFLL uses an Intensity Modulated Continuous Wave (IMCW) measurement method which allows simultaneous transmission and reception of two or more closely spaced (50 pm) wavelengths precisely positioned on the CO_2 line at

1571.1192 nm to acquire the differential absorption over the column of air between the aircraft and the target of interest (e.g. clouds, ground) (Dobler et al. 2013; Lin et al. 2013). Systematic assessments of MFLL lidar data with certain flags are performed to assure and control the quality of MFLL data during lidar data processing. The most critical QA/QC procedures are lidar power check, aircraft attitude evaluation, and cloud determination. A constant threshold for the total power of all channels of MFLL signals is applied to ensure high enough signal-to-noise ratio (SNR) for XCO_2 retrieval. To avoid measurement errors caused by aircraft attitude variations aircraft pitch and roll angles only within ± 5 degrees are tolerated in data processing. Furthermore, thin and thick clouds are identified based on lidar ranging capability. These clouds could reduce lidar SNR or totally block lidar returns from the ground. In these cases, partial column CO_2 retrievals are also reported.
MFLL data processing uses two basic procedures in calibrating MFLL measurements for its XCO_2 retrieval. The first one is MFLL instrument short-path measurements. This calibration compensates the wavelength-dependent throughput of the internal optics of the instrument and accounts for the differences in lidar signal path lengths within and outside the instrument. After this calibration, MFLL data and differential absorption optical depth (DAOD) measurements are further calibrated with in situ derived DAOD values obtained from aircraft spiral CO2 observations (Campbell et al. 2020). This calibration procedure not only makes remote sensing and in situ measurements consistent but also reduces the impacts of various potential error sources on XCO_2 retrievals. With these calibrations, along with QA/QC, MFLL XCO_2 retrievals reach high accuracy (0.8 ppm) and precision (0.26 ppm with 1-min integration; Campbell et al., 2020).

- **High-Altitude Lidar Observatory (HALO)**

NASA Langley Research Center has developed the High-Altitude Lidar Observatory (HALO) system to address the observational needs of NASA's weather, climate/radiation, carbon cycle, and atmospheric composition focus areas. HALO is a modular and multi-function airborne lidar developed to measure atmospheric H_2O and CH_4 mixing ratios and aerosol, cloud, and ocean optical properties using the differential absorption lidar (DIAL) (Nehrir et al., 2017) and high spectral resolution lidar (HSRL) (Hair et al. 2008) techniques, respectively. To respond to a wide range of airborne process studies, HALO can be rapidly reconfigured to provide either, H_2O DIAL/HSRL, CH_4 DIAL/HSRL, or CH_4 DIAL/H_2O DIAL measurements using three different modular laser transmitters and a single multi-channel and multi-wavelength receiver. For the summer 2019 ACT-America campaign HALO employed the CH_4_DIAL/HSRL configuration and archived the standard suite of aerosol extensive and intensive products as described in Hair et al. 2008 as well as the mixed layer height as described in Scarino et al., 2014. HALO also measured, for the first-time, distributions of column weighted XCH_4 during this campaign and will archive those products once the development and validation effort has been completed. HALO data are sampled at 0.5-s temporal and 1.25-m vertical resolutions. The vertical resolution for the aerosol measurements is increased to 15 m in post-processing to increase the SNR of the aerosol intensive and extensive retrievals. Aerosol backscatter and depolarization products are averaged 10 s horizontally and aerosol extinction products are averaged 60 s horizontally and 150 m vertically. The polarization and HSRL gain ratios are calculated as described in Hair et al., 2008. Operational retrievals also provide mixing ratio of non-spherical-to-spherical backscatter (Sugimoto and Lee, 2006), aerosol type (Burton et al., 2012) and aerosol mixed-layer height (Scarino et al., 2014). The raw data are quality controlled by applying a cloud screening mask to remove attenuated signals below clouds. The data are further screened when the aircraft is within 2 km of the surface or when the lidar profile does not reach within 1km of the surface. For the mixing layer height product which is the principal lidar observable for ACT-America, the retrievals are quality controlled beyond the methods described in Scarino et al. (2014) by applying a user defined and time dependent threshold on the wavelet transform. All data products are archived in an H5 file format with 10 second horizontal resolution. Future methane column products will be archived at the 0.5 second native resolution.

- **Quantum Cascade Laser Spectrometer (QCLS)**

Observations are referenced to calibration gas mixtures every 10 mins in flight using a two-point calibration procedure (zero and target mixing ratios) for all measured species (except H_2O). The target calibration gas mixtures (resembling mole fractions close to atmospheric ambient values) have been cross-calibrated against NOAA standards using a cavity ring-down spectrometer (Picarro G2301) and are thus traceable to WMO standards for CH4 and CO2 (WMO X2004A for CH4 – Dlugokencky et al., 2005, WMO X2007 for CO2 – Zhao and Tans, 2006). C2H6, CO and N2O are compared to NOAA flask samples traceable to WMO standards (PI: Colm Sweeney) taken during the ACT-America field campaigns. QA/QC procedures further include manual review of every flight and removal of spurious data associated with in-cavity pressure anomalies.

- **Compact Atmospheric Multi-Species Spectrometer (CAMS-2) Ethane Measurements**

Weibring et al. (2020) discuss comprehensive details of the 2nd generation Compact Atmospheric Multispecies Spectrometer (CAMS-2) employed in acquiring high precision 1-second ethane measurements on the B-200 airplane autonomously without an onboard operator. Ethane mixing ratios were determined by sampling ambient air through a multipass absorption cell where a mid-IR laser operating at a wavelength of 3.34 microns (2996.86 cm-1) was directed back and forth to achieve an optical pathlength of 47.6 m. At this wavelength, the laser is absorbed by a manifold of strong ethane lines and the retrieved ambient mixing ratios are determined employing the Beer-Lambert Absorption Law. To validate the direct absorption results, known calibration mixtures of ethane in air were introduced into the inlet before and after every flight, and the resulting direct absorption determinations were in agreement with the retrieved calibration values to within 6%. The final reported data employed the pre- and post-flight calibrations to correct the data. Comparisons of the continuous CAMS ethane data with time-coincident Portable Flask Package ethane data acquired on the B-200 resulted in agreement in the 4 to 5% range. Post mission exchange of ethane standards also produced agreement in this same range and given the uncertainties in the assigned standards values as well as the spectroscopic parameters, this level agreement was considered quite good. With each successive campaign, the ambient ethane performance was improved, and during 4th and 5th campaigns we routinely achieved 1-second (1σ) ethane precisions in the 30 to 40 pptv range during flight.

- **Tower-based GHG measurements**

Prior to deployment, the instruments were calibrated in the laboratory using 4 NOAA-calibrated tanks. A field calibration tank was sampled daily and used to apply a zero-offset correction. Round robin tests using 3-4 NOAA-calibrated tanks were conducted every 1-2 years. NOAA flask measurements were used for comparison at the Mildred, Greenfield, and Mooresville sites. The averaging interval standard deviation and uncertainty derived from periodic flask sample to in-situ measurement comparisons are provided in the data files. Based on flask to in-situ comparisons and round robin testing presented in Richardson et al. (2017), the estimated compatibility of these measurements is approximately 0.18 ppm CO2 and 0.6 ppb CH4.

- **Meteorological and navigation data products**
  - **C-130**

Two levels of post-collection data quality control are performed. The first occurs within 24 hours of the end of the research flight, and the second within 6 months after the last research flight. These two distinct phases are often referred to as "field/preliminary" and "public/publication quality" data.

The preliminary phase of quality control is intended to capture, highlight, and remove errors in the original signal recordings obtained from the instruments, to prevent error propagation into the derived quantities which are reported in the meteorological and navigation file. Automated checks include plausible value (does the instantaneous signal make sense), plausible rate of change (are the changes between instantaneous values realistic and physically explainable), and internal consistency (are the instantaneous values sensible in relation to other measured values). Each of these processes occur within the field/preliminary phase.

The second phase of data quality control consists of further analysis that expands to an evaluation of instrument biases and long-term stability of the sensors, as well as instrument inter-comparisons with redundant sensor networks (both in situ and model comparisons, where applicable).

The instrumentation supported by NSRC is regularly calibrated by the NASA Armstrong metrology calibration laboratory or within-house by the NSRC Instrumentation Engineer. The calibration records are applied within the second phase of the data quality control process, prior to the submission of publication quality data. Additionally, for ACT-America, a set of aircraft calibration maneuvers were performed regularly throughout the mission to assess the fidelity and stability of the aircraft pitot-static and inertial navigation system used to derive horizontal winds. These maneuvers allow for the quantification of errors in the measurement of static pressure and the true heading alignment of the inertial navigation system. Calibration factors derived from the aircraft maneuvers are applied in the second phase of quality control to aircraft true heading, aircraft static pressure, calculated wind speed, and calculated wind direction.

- **B-200**

Navigational and meteorological measurements obtained on board the LaRC B-200 aircraft have been carefully processed and screened to preserve their integrity and accuracy. Lab characterization, scheduled maintenance, and ground tests procedures in a static environment were performed on all instrumentation involved in making required airborne supporting measurements (Stickney et al., 1990; Edgetech Vigilant 137 Operation Manual). Navigational and meteorological measurements were also calibrated and verified using dynamic airborne flight maneuvers (Barrick et al. 1996; Haering, 1985). This was especially critical to meet the desired accuracy of horizontal winds. Reversed heading maneuvers were performed several times during each field mission to verify accurate derivation of horizontal winds. Static pressure position error and heading alignment were determined and corrections applied to wind calculations. Intercomparison flight legs between the LaRC B-200 and WFF C-130 were also conducted for additional correlative quality assurance purposes. Respective navigational parameters were verified during ground and airborne flights utilizing inertial and GPS techniques via two well documented systems. Navigational and attitude parameters from both a differential GPS (DGPS) and Applanix Pos/AVTM direct georeferencing system (DG) were recorded during all flights for redundancy and quality assurance purposes. The DGPS technique enhances the accuracy limits of GPS receivers by removing selective availability, atmospheric conditions, timing, and satellite orbit errors. The Applanix DG system integrates DGPS measurements with an inertial measurement system for added stability and accuracy (Mostafa et al. 2001).