Final Technical Report for Interagency Agreement No. DE-SC0005453 "Characterizing Aerosol Distributions, Types, and Optical and Microphysical Properties using the NASA Airborne High Spectral Resolution Lidar (HSRL) and the Research Scanning Polarimeter (RSP)"

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#### **OBJECTIVE:**

Measurements of the vertical profile of atmospheric aerosols and aerosol optical and microphysical characteristics are required to: 1) determine aerosol direct and indirect radiative forcing, 2) compute radiative flux and heating rate profiles, 3) assess model simulations of aerosol distributions and types, and 4) establish the ability of surface and space-based remote sensors to measure the indirect effect. Consequently the ASR program calls for a combination of remote sensing and in situ measurements to determine aerosol properties and aerosol influences on clouds and radiation. As part of our previous DOE ASP project, we deployed the NASA Langley airborne High Spectral Resolution Lidar (HSRL) on the NASA B200 King Air aircraft during major field experiments in 2006 (MILAGRO and MaxTEX), 2007 (CHAPS), 2009 (RACORO), and 2010 (CalNex and CARES). The HSRL provided measurements of aerosol extinction (532 nm), backscatter (532 and 1064 nm), and depolarization (532 and 1064 nm). These measurements were typically made in close temporal and spatial coincidence with measurements made from DOEfunded and other participating aircraft and ground sites. On the RACORO, CARES, and CalNEX missions, we also deployed the NASA Goddard Institute for Space Studies (GISS) Research Scanning Polarimeter (RSP). RSP provided intensity and degree of linear polarization over a broad spectral and angular range enabling column-average retrievals of aerosol optical and microphysical properties. Under this project, we analyzed observations and model results from RACORO, CARES, and CalNex and accomplished the following objectives.

- 1. Identified aerosol types, characterize the vertical distribution of the aerosol types, and partition aerosol optical depth by type, for CARES and CalNex using HSRL data as we have done for previous missions.
- 2. Investigated aerosol microphysical and macrophysical properties using the RSP.
- 3. Used the aerosol backscatter and extinction profiles measured by the HSRL to characterize the planetary boundary layer height (PBL) and the transition zone thickness, for the RACORO and CARES and CalNex campaigns as we have done for previous campaigns.
- 4. Investigated how optical properties measured by HSRL vary near clouds.
- 5. Assessed model simulations of aerosol spatial distributions and optical and microphysical properties.

## **Accomplishments:**

1. Identified aerosol types, characterize the vertical distribution of the aerosol types, and partition aerosol optical depth by type, for CARES and CalNex using HSRL data as we have done for previous missions.

We used HSRL data to characterize the vertical and horizontal distribution of aerosols, provide the vertical context for the airborne in situ measurements acquired from these other aircraft, and to derive the height of the mixing layer (ML). We employed lidar intensive observables (extinction-to-backscatter ratio, backscatter color ratio, depolarization ratio, and spectral ratio of depolarization) to infer aerosol types (e.g., urban, smoke, dust, etc.) and determine the fraction of aerosol optical thickness (AOT) contributed by these types. The aerosol type results are described in Burton et al. [2012]. We also compared aerosol type to the PALMS in situ instrument based on Pasadena during CalNex for a coarse mode (sea salt) case and found good agreement. Armed with comparisons with PALMS for cases showing sea salt and sea salt mixtures, we were able to show that the HSRL measurements for a CARES case in which in situ measurements at the ground sites T0 and T1 were observing coarse mode were consistent with a mix of sea salt and organics. Investigation of HSRL-derived aerosol types for CalNex and CARES led to improvements in our aerosol typing algorithm and the use of typing result to assess compositional distributions predicted by models. For example, urban aerosols in California tend to have a smaller lidar ratio than those near East Coast urban centers. We also computed aerosol types and apportioned aerosol optical thickness to type for DOE CARES and TCAP missions. We also automated the aerosol typing so that it is now a standard HSRL product. Aerosol classification results are discussed in Burton et al. (2012).

# 2. Investigated aerosol microphysical and macrophysical properties using the RSP.

RSP is the airborne prototype for the Aerosol Polarimetry Sensor (APS) instrument built for the Glory satellite mission. The RSP team has decided to take advantage of the extensive data processing infrastructure developed for Glory to generate level-2 data products from the RSP level-1 radiance data archived for the past ASP/ASR field missions. The team focused their efforts this year on modifying algorithms developed for the Glory. This included modifications required to process data acquired from the 8-9 km altitude of the B200 aircraft: algorithms for a top-ofatmosphere satellite viewing geometry had to be modified for the in-the-atmosphere aircraft viewing geometry to account for light scattering both above and below the instrument. This work included generating extensive look-up tables required to make processing computationally efficient. In addition the last step in the retrieval process requires exact radiative transfer calculations together with functional derivatives to drive the final iterative retrieval steps and improve retrieval accuracy beyond what is possible using a look-up-table. The team has recently completed a modification to a vector doubling/adding radiative transfer code that allows the functional derivatives to be calculated from a single run of the radiative transfer code for both external sources (i.e. solar illumination and satellite observations) and internal sources (i.e. the adjoint radiative transfer solution for aircraft observations). Data from RACORO has been used to evaluate partial cloud fraction effects on aerosol retrievals using polarized reflectance observations (Cairns et al. 2011) and the capability to simultaneously retrieve aerosol and cloud properties in cumulus cloud fields. The capability to retrieve aerosols over snow was evaluated using data from CARES (Ottaviani et al. 2012). The level-2 products (i.e., spectral optical depth, particle size distribution, complex index of refraction, single scatter albedo, etc.) will be used for future analyses of aerosol microphysical and macrophysical properties.

In order to effectively perform a combined analysis of RSP and HSRL data it is necessary to have a non-spherical aerosol model that is compatible with the observed depolarization and spectral depolarization ratios that have been observed by HSRL (Burton et al. 2012) and also the

multi-angle multi-spectral polarized radiances observed by the RSP since coarse mode aerosols appear to be frequently non-spherical in shape. We have identified an equi-probable aspect ratio distribution of prolate and oblate spheroids as being compatible with the RSP observations (Chowdhary et al. 2012). We have completed an extensive tabulation of the scattering properties of such particles as a function of their size and complex refractive index for the RSP spectral bands and the lidar wavelengths of 355, 532 and 1064 nm, and have demonstrated that this type of particle is compatible with the aerosol types identified using the HSRL (Burton et al. 2012). These non-spherical particle models are now being incorporated into the aerosol retrieval scheme and products.

Evaluating the effects of aerosols on clouds requires robust and accurate remote sensing techniques for determining cloud microphysical properties. We have developed a method for determining the cloud droplet size distribution using RSP data that requires no assumptions regarding the form of the droplet size distribution (Alexandrov et al. 2012a) and can therefore be used to evaluate whether droplets are evaporating homogeneously, or heterogeneously near cloud top. We have also evaluated the robustness of a parametric retrieval of cloud droplet size distribution against multiple scattering and three-dimensional effects (Alexandrov et al. 2012b).

A simplified RSP retrieval scheme with and without HSRL constraints was developed in parallel with the advanced retrieval efforts — to retrieve aerosol microphysical properties for TCAP scenes over water. The aerosols are assumed to be bimodal and log-normally distributed in one-or two-layer configurations. By using non-linear optimal estimation and vector doubling/adding simulations of the Stokes parameter I and DLP, the degree of linear polarization, column-averaged retrievals will be performed on the following retrieval parameters: f, the fraction of coarse-to-fine mode;  $\tau_{865}$ , the column aerosol optical depth at 865 nm;  $r_{eff}$ , the coarse mode effective radius;  $v_{eff}$ , variance of the coarse mode; and n the coarse mode refractive index. The initial HSRL constraints will be the aerosol extinction and the location of the aerosols. We expect that additional retrieval parameters can be added (e.g. vertically resolved aerosol properties, scattering by non-spherical particles) and that the retrieval can be readily extended to scenes over land by taking the appropriate surface boundary conditions into effect and by using the I, Q, and U Stokes parameters separately in the inversion.

# 3. Used the aerosol backscatter and extinction profiles measured by the HSRL to characterize the planetary boundary layer height (PBL) and the transition zone thickness, for the RACORO and CARES and CalNex campaigns as we have done for previous campaigns.

We focused our analyses on two products which have a more straightforward definition in terms of the lidar products we use to estimate them: (1) the mixing layer height and (2) the height of the maximum aerosol gradient. We determine these heights by finding and quantifying gradients in aerosol backscatter using a Haar wavelet covariance transform. The mixing layer height is the height of gradient we find closest to ground level and generally corresponds with PBL heights as defined by traditional meteorological definitions (e.g., gradient in potential temperature). The mixing layer height is often not the maximum gradient we see in the lidar signal. That gradient is often significantly higher (1 or more km) and can be thought of as the scale height of the tropospheric aerosols, including residual layers. We produced a data product with mixing layer height and height of maximum aerosol gradient for the CARES, CalNex, and RACORO data sets. Most of the subsequent analysis focused on the CARES and CalNex data sets. We compared

our mixing layer heights with PBL heights derived from radiosondes from the CARES T0 and T1 sites and found excellent agreement: the correlation analysis exhibited  $R^2 = 0.86$ , with slopes of 0.9 and a mean difference of  $\sim 7\%$  for data acquired within 30 minutes and 15 km of the sonde launch site. For CalNex, our mixing layer heights were also compared to PBL heights derived from a ceilometer with a correlation of  $R^2$ =0.98. We also made extensive comparisons to WRF-Chem estimates of PBL height and describe those under objective 5 below.

The NASA LaRC airborne High Spectral Resolution Lidar (HSRL) was deployed to California on board the NASA LaRC B-200 aircraft to aid in characterizing aerosol properties during the Carbonaceous Aerosol and Radiative Effects Study (CARES) field campaign in 2010. The HSRL ML heights were used to evaluate the performance in simulating the temporal and spatial variability of ML heights from the Weather Research and Forecasting Chemistry (WRF-Chem) community model. Hourly WRF-Chem simulations were extracted along the HSRL flight track using the Aerosol Modeling Testbed software tools. Scarino et al. [2013] demonstrated that the WRF-Chem under-predicted the ML heights when the ML height was low, but tended to over predict when the ML height was large. There was generally good agreement over the flat terrain in the Central Valley, but on certain days WRF-Chem did not correctly represent the diurnal growth of the mixed layer and distributed aerosol over a much taller ML than measurements indicated, up to twice the measured ML height. In contrast, the complex terrain and bodies of water in the San Francisco Bay and Sierra Nevada regions introduced larger uncertainties in the simulated interaction of surface fluxes, boundary layer mixing, and ambient winds or there were local variations in the ML depth that the model did not resolve using a grid spacing of 4 km. The HSRL aerosol ML heights provided additional information for validating and improving model ML heights by providing the means to distinguish between biases due to BL parameterizations from those due to other factors such as interaction with synoptic meteorology.

# 4. Investigated how optical properties measured by HSRL vary near clouds.

HSRL data acquired on several days in June 2009 during the RACORO mission were used to examine the behavior of aerosol optical properties near clouds. Aerosol backscatter, extinction, optical thickness, depolarization, extinction/backscatter, and wavelength dependence were examined for changes occurring with increasing proximity to clouds. Initial results show 5-40% increases in aerosol backscatter for samples within 1-2 km of clouds; smaller changes and more variability were observed in aerosol extinction and optical thickness. Aerosol depolarization generally decreased within a few kilometers of clouds. These changes are likely due to aerosol humidification causing the aerosols to become more spherical in the vicinity of clouds. Backscatter wavelength dependence and aerosol extinction/backscatter ratio did not show a consistent behavior near clouds.

# 5. Assessed model simulations of aerosol spatial distributions and optical and microphysical properties.

The performance of the WRF-Chem in simulating the spatial and temporal variations in aerosol mass, composition, and size over California was quantified using the extensive meteorological, trace gas, and aerosol measurements collected during the California Nexus of Air Quality and Climate Experiment (CalNex) and CARES campaigns conducted during May and June of 2010. Fast et al. [2014] compared WRF-Chem output with NASA LaRC airborne HSRL and in situ

measurements, and found that long-range transport of aerosols from the global model was too high in the free troposphere even though their concentrations were relatively low. This bias led to an over-prediction in aerosol optical depth by as much as a factor of 2 that offset the under prediction of boundary-layer extinction resulting primarily from local emissions. Lowering the boundary conditions of aerosol concentrations by 50% greatly reduced the bias in simulated aerosol optical depth for all regions of California.

We have also examined and evaluated the ECMWF/MACC aerosol model products using aerosol profiles from HSRL. The ECMWF model parameters and HSRL measurements were compared the King Air flight tracks for 17 field missions, including RACORO, CalNex and CARES. Comparisons include AOT in the 0-7 km column, aerosol extinction profiles, fraction of AOT and extinction due to natural and anthropogenic aerosols, PBL height (mixed layer height from HSRL used as proxy for PBL height), and fraction of AOT within the PBL. For the aerosol extinction profile comparisons, there was considerable variability in the profiles and the best agreement was found within the PBL. ECMWF often had higher extinction in the free troposphere, especially over the western United States. Larger differences in AOT and aerosol extinction over Los Angeles and California are noted and likely associated with small scale variability, accuracy and availability of assimilated MODIS AOT, local emission sources not well resolved and a lack of nitrates in the model. Overall, ECMWF PBL heights are generally 100 to 200 m higher than the HSRL mixed layer heights. As for the comparisons of natural and anthropogenic aerosols, there is very good agreement in fractions of AOT and aerosol extinction contributed by natural and anthropogenic aerosols. The HSRL anthropogenic aerosol fraction is about 5-10% higher than ECMWF and most missions saw high (greater than 75%) fraction of anthropogenic aerosols.

## **PUBLICATIONS**

- Alexandrov, M.D., B. Cairns, and M.I. Mishchenko: Rainbow Fourier transform. J. Quant. Spectrosc. Radiat. Transfer, 113, 2521-2535, doi:10.1016/j.jqsrt.2012.03.025, 2012.
- Alexandrov, M.D., B. Cairns, C. Emde, A.S. Ackerman, and B. van Diedenhoven: Accuracy assessments of cloud droplet size retrievals from polarized reflectance measurements by the research scanning polarimeter. *Remote Sens. Environ.*, 125, 92-111, doi:10.1016/j.rse.2012.07.012, 2012.
- Burton, S. P., Ferrare, R. A., Hostetler, C. A., Hair, J. W., Rogers, R. R., Obland, M. D., Butler, C. F., Cook, A. L., Harper, D. B., and Froyd, K. D.: Aerosol classification using airborne High Spectral Resolution Lidar measurements methodology and examples, Atmos. Meas. Tech., 5, 73-98, doi:10.5194/amt-5-73-2012, 2012.
- Fast, J. D., Gustafson Jr., W. I., Berg, L. K., Shaw, W. J., Pekour, M., Shrivastava, M., Barnard, J. C., Ferrare, R. A., Hostetler, C. A., Hair, J. A., Erickson, M., Jobson, B. T., Flowers, B., Dubey, M. K., Springston, S., Pierce, R. B., Dolislager, L., Pederson, J., and Zaveri, R. A.: Transport and mixing patterns over Central California during the carbonaceous aerosol and radiative effects study (CARES), Atmos. Chem. Phys., 12, 1759-1783, doi:10.5194/acp-12-1759-2012, 2012.
- Fast, J.D., and coauthors, 2014: Modeling regional aerosol and aerosol precursor variability over California and its sensitivity to emissions and long-range transport during the 2010 CalNex and CARES campaigns, *Atmos. Chem. Phys.*, **14**, 10013-10060, doi:10.5194/acp-14-10013-2014.

- Ottaviani, M., B. Cairns, R.R. Rogers, and R. Ferrare: Iterative atmospheric correction scheme and the polarization color of alpine snow. *J. Quant. Spectrosc. Radiat. Transfer*, 113, 789-804, doi:10.1016/j.jqsrt.2012.03.014, 2012.
- Scarino AJ, MD Obland, JD Fast, SP Burton, RA Ferrare, CA Hostetler, LK Berg, B Lefer, C Haman, JW Hair, RR Rogers, C Butler, AL Cook, and DB Harper, 2014: Comparison of mixed layer heights from airborne high spectral resolution lidar, ground-based measurements, and the WRF-Chem model during CalNex and CARES. *Atmos. Chem. Phys.*, 14, doi:10.5194/acp-14-5547-2014
- Turner, D. D., Ferrare, R. A, Wulfmeyer, V. and Scarino, A. J.: Aircraft Evaluation of Ground-based Raman Lidar Water Vapor Turbulence Profiles in Convective Mixed Layer, J. Atmos. Ocean Tech., Submitted 15 Mar 2013.