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Two-Column Aerosol Project (TCAP) Field Campaign Report

LK Berg

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LK Berg, Pacific Northwest National Laboratory Principal Investigator

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Executive Summary

The Two Column Aerosol Project (TCAP) was conducted from June 2012 through June 2013 and included the deployment of the Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facility's Mobile Facility (AMF), ARM Mobile Aerosol Observing System (MAOS) and the ARM Aerial Facility (AAF). The study was a collaborative effort involving scientists from DOE national laboratories, NOAA, NASA, and universities. The AAF and MAOS were deployed for two approximately month-long Intensive Operational Periods (IOPs) conducted in June 2012 and February 2013. Seasonal differences in the aerosol chemical and optical properties observed using the AMF, AAF, and MAOS are presented in this report. The total mass loading of aerosol is found to be much greater in the summer than in the winter, with the difference associated with greater amounts of organic aerosol. The mass fraction of organic aerosol is much reduced in the winter, when sulfate is the dominant aerosol type. Surprisingly, very little sea-salt aerosol was observed in the summer. In contrast, much more sea salt aerosol was observed in the winter. The mass loading of black carbon is nearly the same in both seasons. These differences lead to a relative increase in the aerosol light absorption in the winter and an associated decrease in observed single-scattering albedo. Measurements of aerosol mixing state were made using a single-particle mass spectrometer, which showed that the majority of the summertime aerosol consisted of organic compounds mixed with various amounts of sulfate. A number of other findings are also summarized in the report, including: impact of aerosol layers aloft on the column aerosol optical depth; documentation of the aerosol properties at the AMF; differences in the aerosol properties associated with both columns, which are not systematic but reflect the complicated meteorological and chemical processes that impact aerosol as it is advected away from North America; and new instruments and data-processing techniques for measuring both aerosol and cloud properties that were deployed for the first time during the TCAP.

Key lessons learned during TCAP include the need for closer coordination between the AMF, MAOS, and the AAF so that all AMF instruments can be online and functioning during the AAF IOPs. Based on experiences from TCAP, it is also important for instrument mentors, or other relevant individuals, to review data on a regular basis to ensure that data quality remains high during the entire deployment.

TCAP was marked by two important meteorological events including the passage of Hurricane Sandy at the end of October 2012 and the occurrence of one of the largest New England blizzards in recorded history. During Sandy the AMF received some, generally minor, damage and was largely functional a short time after the storm. The blizzard led to extensive power outages on Cape Cod and a multi-day interruption of measurements by the AMF, MAOS, and AAF. In each case, however, the ARM Facilities were returned to service and functioning as soon as was reasonably possible.

Acronyms and Abbreviations

4STAR	Spectrometer for Sky-Scanning Sun-Tracking Atmospheric Research
AAF	ARM Aerial Facility
ACSM	Aerosol Chemical Speciation Monitor
AERONET	Aerosol Robotic Network
AMF	ARM Mobile Facility
AOD	Aerosol Optical Depth
ARM	Atmospheric Radiation Measurement Climate Research Facility
CU	University of Colorado
DOE	U.S. Department of Energy
G-1	ARM Gulfstream aircraft
GVAX	Ganges Valley Aerosol Experiment
HSRL-2	Second-Generation High Spectral Resolution Lidar
IOP	Intensive Operational Period
km	kilometer
kPa	kilopascal
MAOS	Mobile Aerosol Observing System
MAX-DOAS	Multi Axis Differential Optical Absorption Spectroscopy
miniSPLAT	Single-Particle Laser Ablation Time-of-Flight mass spectrometer
NASA	National Aeronautic and Space Administration
NOAA	National Oceanic and Atmospheric Administration
PDF	Probability Density Functions
PILS	Particle Into Liquid Sampler
SACR	Scanning ARM Cloud Radar
SOA	Secondary Organic Aerosol
ТСАР	Two-Column Aerosol Project

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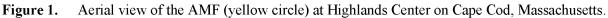
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1.0 Background

The Two-Column Aerosol Project was conducted from June 2012 through June 2013 and included the deployment of both the ARM Mobile Facility (AMF) and the ARM Aerial Facility (AAF). The study was a collaborative effort involving scientists from DOE national laboratories, NOAA, NASA, and universities (including the University of Colorado and the Massachusetts Institute of Technology). The AMF was deployed, in collaboration with staff from the Cape Cod National Seashore, near the eastern edge of Cape Cod at the Highlands Center (Error! Reference source not found.). This was the first combined deployment of the AMF and the ARM Mobile Aerosol Observing System (MAOS). The AAF was based in Hyannis, Massachusetts for two Intensive Operational Periods (IOPs), one in June of 2012 and one in February of 2013. Two aircraft were deployed during the summer IOP, including the ARM Gulfstream 1 (G-1) and NASA King Air B200, while only the G-1 was deployed in the second IOP.





The primary goal of TCAP was to investigate changes in the chemical and optical properties of aerosol as they are transported away from North America. Due to the wide range of cloud types observed at Cape Cod, the AMF and AAF deployments also provide a unique opportunity to document details of cloud properties in the vicinity of Cape Cod and to look for evidence of cloud-aerosol interactions.

The TCAP flight pattern (Figure 2) was designed to be relatively simple, sampling in the two columns at a range of heights to enable easy comparison with global- and regional-scale models. The orientation of the flight pattern was determined based on observations made at the AMF and weather forecasts so that the aircraft would either sample approximately downwind of Boston, Massachusetts or across aerosol gradients. As shown in Figure 2, the orientation of the flight pattern changed from day to day, with the maritime column (indicated by the gray shading in the figures) being located approximately 200 kilometers (km) from the AMF and MAOS in each case.

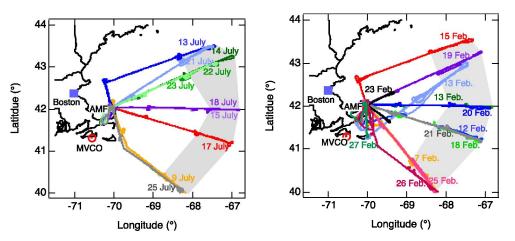


Figure 2. AMF location and flight patterns used in the summertime (G-1 and King Air; left) and wintertime (G-1 only; right) during TCAP. Gray shading marks the locations of the maritime columns that varied from flight to flight.

2.0 Notable Events or Highlights

The TCAP deployment was marked by two significant meteorological events. The first was Hurricane Sandy, which passed over the AMF site on 29 October 2012. During passage of the storm, the surface pressure at the AMF site dropped below 99.5 kilopascals (kPa). Sandy knocked out power over much of Cape Cod and the AMF systems were down from 30 October through 1 November 2012. The second significant event, which occurred during the winter IOP, was one of the largest blizzards in the history of New England. This storm dropped over a foot of snow on 8 February 2013 and led to widespread power outages over much of Cape Cod, resulting in a relatively long down period for the AMF and MAOS that lasted from 8 through 15 February. While the AMF and MAOS did have an onsite generator, the systems were forced to shut down when the fuel was expended and the instruments did not come back online until 15 February. The blizzard also led to a break in aircraft operations due to the closure of the Hyannis airport. In each case, however, the AMF and AAF were up and functioning as soon as was reasonably possible.

3.0 Lessons Learned

While TCAP was an unqualified success, there are a number of logistical lessons that can be learned from the deployment that could be applicable for future campaigns.

- There were issues associated with AMF instruments due to the relatively quick turnaround time between TCAP and the Ganges Valley Aerosol Experiment (GVAX; the AMF deployment immediately before TCAP). This led to a delay in getting a number of instruments operational until after the official TCAP start date. This issue has been addressed by ARM and a new paradigm of operations includes downtime between deployments.
- Greater coordination is needed in the setup of the AMF and MAOS and the timing of the AAF deployment. By their nature, long-term surface deployments such as the AMF have a different sense

of urgency than short-term, but intense, aircraft deployments. In the future, the AMF site should be set up well before the start of the AAF IOP to ensure that all surface-based instrument systems are operational at the start of the aircraft IOPs.

• Instrument mentors should closely follow the output from their instruments, especially at the start of the study. During TCAP, issues with the surface radiometric instruments were found by members of the science team. The problems were successfully addressed, but identification of the problems took longer than is ideal.

4.0 Results

TCAP provides one of the first opportunities to look at the annual cycle of a wide range of key aerosol properties near the coast of North America. Other networks, such as AERONET, provide some information about the columnar aerosol properties, but only for a small number of parameters in clear-sky conditions. An analysis of the seasonal cycle of the aerosol optical properties shows that there is a great deal of annual variability, making it difficult to identify the season from the aerosol optical properties alone, as shown in Figure 3 (and as presented by [*Titos et al.* 2014]). When the data is used to construct probability density functions (PDFs) of aerosol optical properties, seasonal differences become apparent, with the wintertime having a smaller single-scattering albedo (ω_0) than is found during the summer. The deployment of the MAOS allows us to look into this difference more carefully to better understand its root causes.

The Aerosol Chemical Speciation Monitor (ACSM) deployed with the MAOS provides a quantitative measure of the aerosol mass loading and the chemical speciation for non-refractory aerosol. In the summertime the aerosol mass was dominated by organic aerosol (nearly 75% of the total mass) and relatively small amounts of sulfate and ammonium (Figure 4). Data from the airborne miniSPLAT (Single-Particle Laser Ablation Time-of-Flight mass spectrometer) indicates that the majority of the summertime aerosol consists of particles that are a mixture of organic compounds and various amounts of sulfate [*Berg et al.* 2015]. The total mass loading is found to be smaller in winter, and the relative amount of organics is decreased. The change in the organic mass loading (and mass fraction) is most likely associated with the absence of biogenic emissions in wintertime and reduced production of secondary organic aerosol (SOA).

Given the proximity of the AMF to the shore, one would anticipate that a large amount of sea salt aerosol would be observed. Using data from the MAOS Particle Into Liquid Sampler (PILS—the only instrument in the MAOS that can measure sea salt concentrations), a strong seasonal cycle in the mass loading of sea salt aerosol was noted. The summertime data show relatively small, but nearly equal, amounts of sodium and chloride (Figure 5). This ratio is not indicative of sea-salt aerosol, and likely represents sodium and chloride aerosol from other sources with some minor contribution to the mass loading of each associated with sea salt. The mass loading of sodium and chloride is much larger in the wintertime, and the ratio of the two elements is consistent with sea salt. Likewise, the miniSPLAT measured larger amounts of sea-salt aerosol during the wintertime than during the summer IOP (not shown). These differences are likely driven by relatively weaker winds that occur in the summertime and differences in the thermodynamic structure of the atmosphere, which featured a much deeper mixed layer in winter associated with the advection of cold air over the relatively warm waters of the Atlantic Ocean.

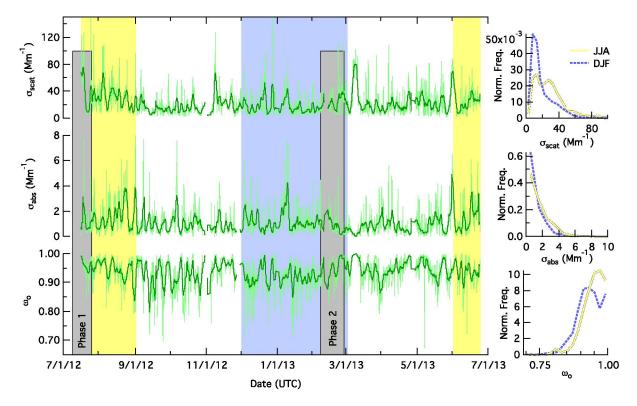


Figure 3. Time series of aerosol light scattering (σ_{scat} ; top), aerosol light absorption (σ_{abs} ; middle), and ω_0 (bottom) at a wavelength of 550 nm measured at the AMF. Right panels show PDFs for summer (June, July, August; yellow) and winter (December, January, February; blue). Gray boxes indicated AMF and MAOS IOPs; blue and yellow boxes indicated periods used to generate PDFs.

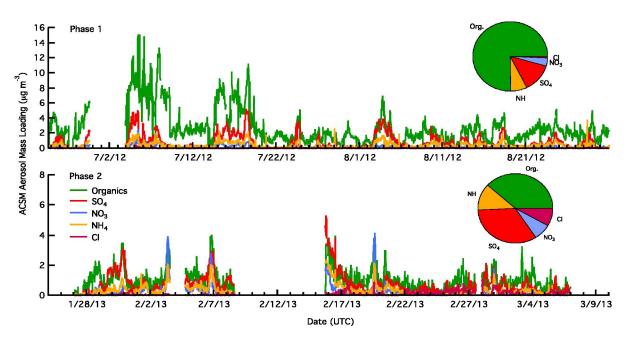


Figure 4. Time series of aerosol chemical composition derived from the ACSM deployed with the MAOS. Pie charts indicate mass fraction derived for each IOP.

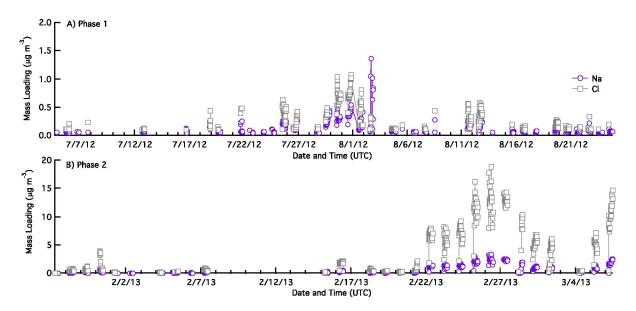


Figure 5. Time series of sodium (purple) and chloride (gray) measured using the PILS deployed with the MAOS during the summer (top) and winter (bottom) IOPs.

In addition to new insights into the seasonal cycle of aerosol, TCAP produced several other important scientific findings. For brevity, they are not described in detail in this document, but rather specific highlights and references to the peer-reviewed literature are included below.

- *Presence of aerosol layers aloft*: Using a combination of data from the second-generation NASA High Spectral Resolution Lidar (HSLR-2) and the in situ data from the G-1, frequent aerosol layers aloft were identified (occurring on 4 of 6 nearly cloud-free flights during the summer IOP). These layers were found to contribute up to 60% of the column aerosol optical depth and were marked with increased amounts of biomass burning and nitrate aerosol as measured by the miniSPLAT. A detailed description of the layers and their impact can be found in *Berg et al.* [2015].
- *Differences in the aerosol properties in the Cape Cod and Maritime Columns*: Data from the two columns were analyzed to document differences in the aerosol chemical composition and optical properties for both seasons. No systematic differences between the two columns were found, which highlights the complicated flow structure and air mass history over eastern North America. On a day-by-day basis, however, there were appreciable differences in the aerosol properties within the two columns [*Berg et al.*,2015].
- Aerosol properties at the AMF site. A number of studies have examined the aerosol optical properties at the AMF site in some detail. Work by *Kassianov et al.* [2013] highlighted the importance of accounting for the temporal variability of the AOD when computing aerosol radiative forcing, which is particularly important to consider for satellite data that is temporally sparse. *Titos et al.* [2014] used data from the entire year-long AMF deployment to investigate relationships in the aerosol optical properties and the aerosol source region.
- *New data processing methods: Kassianov et al.* [2015] showed the importance of considering particle light absorption (even for weakly absorbing particles) when determining particle size distributions and conducting optical closure studies using data from the ARM G-1. New techniques have also been

developed to estimate the aerosol density and real refractive index from AMF measurements of light scattering and particle size distributions [*Kassianov et al.* 2014].

• Deployment of new instruments: TCAP was the first scientific deployment of three key instruments that have been documented in a series of separate manuscripts. This suite of new instrumentation provides a powerful package of remote-sensing and in situ instruments to better understand the optical properties of the aerosol and new retrievals of aerosol microphysical properties as well as detailed measurements of the aerosol chemical composition and mixing state. The new instruments include the Spectrometer for Sky-Scanning Sun-Tracking Atmospheric Research [4STAR; Dunagan et al. 2013; Shinozuka et al. 2013], the NASA HSRL-2 [Müller et al. 2014], and the miniSPLAT single-particle mass spectrometer [Zelenvuk et al. 2015]. Data from the 4STAR have been used to determine the columnar spectrally resolved AOD [Shinozuka et al. 2013], and to determine the columnar concentration of a number of trace gases [Segal-Rosenheimer et al. 2014]. As part of the analysis of TCAP data, the HSRL-2 has been used to determine the height-resolved contribution to AOD [Berg et al. 2015] and aerosol microphysical properties [Müller et al. 2014]. TCAP was also the first operational deployment of the scanning ARM cloud radar (SACR) with the AMF, providing an unprecedented view of the clouds over Cape Cod that have been used in several studies [Berg et al. 2015; Kollias et al. 2014; Lamer et al. 2014]. An example of cumulus and stratus sampled during TCAP are shown in Figure 6 (after Berg et al. [2015]).

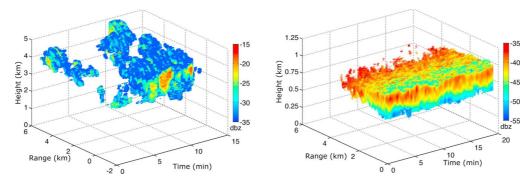


Figure 6. Examples of 3-D gridded radar reflectivity fields from the Ka-band SACR during TCAP for a cumulus field sampled on 27 July 2012 (left) and a stratus sampled on 19 November 2012 (right).

5.0 Public Outreach

In collaboration with the National Park Service, there was a large public outreach effort associated with TCAP. This included the hiring of a special park ranger to specifically focus on the educational opportunities provided by the deployment of the AMF¹. Other outreach included online articles in *Physics Today*² and physics.org³ (articles in local papers, interviews with the local National Public Radio station⁴,

¹<u>https://www.nps.gov/resources/2016.htm?id=B57B49EB-155D-451F-67CF018A1E01E868</u>.

² http://scitation.aip.org/content/aip/magazine/physicstoday/news/10.1063/PT.4.0447

³ <u>http://phys.org/news/2013-03-mobile-aerosol-deployed-cape-cod.html</u>

⁴ <u>http://capeandislands.org/post/cape-based-project-aims-strengthen-climate-models#stream/0</u>

a seminar given as part of the Marine Biology Laboratory Seminar Series in Woods Hole, Massachusetts, and articles in the Cape Cod⁵ and Cape Cod National Seashore papers.

6.0 TCAP Journal Articles/Manuscripts

This list only includes manuscripts that have already been published as of May 2016. Those in the peerreview process have not been included.

Berg, LK, JD Fast, JC Barnard, SP Burton, B Cairns, D Chand, JM Comstock, S Dunagan, RA Ferrare, CJ Flynn, JW Hair, CA Hostetler, J Hubbe, A Jefferson, R Johnson, EI Kassianov, CD Kluzek, P Kollias, K Lamer, K Lantz, F Mei, MA Miller, J Michalsky, I Ortega, M Pekour, RR Rogers, PB Russell, J Redemann, AJ Sedlacek, M Segal-Rosenheimer, B Schmid, JE Shilling, Y. Shinozuka, SR Springston, JM Tomlinson, M Tyrrell, JM Wilson, R Volkamer, A Zelenyuk, and CM Berkowitz. 2015. "The Two-Column Aerosol Project: Phase I—Overview and impact of elevated aerosol layers on aerosol optical depth. "Journal of Geophysical Research–Atmospheres 121(1): 336-361, doi:10.1002/2015JD023848.

Dunagan, SE, R.Johnson, J Zavaleta, PB Russell, B Schmid, C Flynn, J Redemann, Y Shinozuka, J Livingston, and M Segal-Rosenhaimer. 2013. "Spectrometer for sky-scanning sun-tracking atmospheric research (4STAR): Instrument technology." *Remote Sensing* 5(8): 3872-3895, <u>doi:10.3390/rs5083872</u>.

Müller, D, CA Hostetler, RA Ferrare, SP Burton, E Chemyakin, A Kolgotin, JW Hair, AL Cook, DB Harper, RR Rogers, RW Hare, CS Cleckner, MD Obland, J Tomlinson, LK Berg, and B Schmid, 2014. "Airborne multi wavelength High Spectral Resolution Lidar (HSLR-2) observations during TCAP 2012: Vertical profiles of optical and microphysical properties of a smoke/urban haze plume over the northeastern coast of the US." *Atmospheric Measurement Techniques* 7: 3487-3496, doi:10.5194/amt-7-3487-2014.

Kassianov, E, J Barnard, M Pekour, LK Berg, J Michalsky, K Lantz, and G Hodges. 2013. "Do diurnal aerosol changes affect daily averaged radiative forcing?" *Geophysical Research Letters* 40(12): 3265-3269, <u>doi:10.1002/grl.50567</u>.

Kassianov, E, J Barnard, M Pekour, LK Berg, J Shilling, C Flynn, F Mei, and A Jefferson. 2014. "Simultaneous retrieval of effective refractive index and density from size distribution and light scattering data: Weakly absorbing aerosol." *Atmospheric Measurement Techniques* 7: 3247-3261, <u>doi:10.5194/amt-7-3247-2014</u>.

Kassianov, E, LK Berg, M Pekour, J Barnard, D Chand, C Flynn, M Ovchinnikov, A Sedlacek, B Schmid, J Shilling, J Tomlinson, and J Fast. 2015. "Airborne aerosol in situ measurements during TCAP: A closure study of total scattering." *Atmosphere* 6(8): 1069-1101, <u>doi:10.3390/atmos6081069</u>.

Kollias, P, I Jo, P Borque, A Tatarevic, K Lamer, N Bharadwau, K Widener, K Johnson, EE Clothiaux. 2014. "Scanning ARM cloud radars. Part II: Data quality control and processing." *Journal of Atmospheric and Oceanic Technology* 31: 583-598, doi:10.1175/JTECH-D-13-00045.1.

⁵ http://www.capecodtimes.com/article/20120720/NEWS/207200337

Lamer, K, A Tatarevic, I Jo, and P Kollias. 2014. "Evaluation of gridded scanning ARM cloud radar reflectivity observations and vertical Doppler velocity retrievals." *Atmospheric Measurement Techniques* 7: 1089-1103, doi:10.5194/amt-7-1089-2014.

Ortega, I, LK Berg, RA Ferrare, JW Hair, CA Hostetler, and R Volkamer. 2016. "Elevated aerosol layers modify the O_2 - O_2 absorption measured by ground-based MAX-DOAS." *Journal of Quantitative Spectroscopy and Radiative Transfer* 176: 34-49, <u>doi:10.1016/j.jqsrt.2016.02.021</u>.

Segal-Rosenheimer, M. PB Russell, B Schmid, J Redemann, JM Livingston, CJ Flynn, RR Johnson, SE Dunagan, Y Shinozuka, J Herman, A Cede, N Abuhassan, JM Comstock, JM Hubbe, A Zelenyuk, and J Wilson. 2014. "Tracking elevated pollution layers with a newly developed hyperspectral sun/sky spectrometer (4STAR): Results from the TCAP 2012 and 2013 campaigns." *Journal of Geophysical Research–Atmospheres* 119(5) 2611-2628, doi:10.1002/2013JD020884.

Shinozuka, Y, RR Johnson, CJ Flynn, PB Russell, B Schmid, J Redemann, SE Dunagan, CD Kluzek, JM Hubbe, M. Segal-Rosenheimer, JM Livingston, TF Eck, R Wagener, L Gregory, D Chand, LK Berg, RR Rogers, RA Ferrare, JW Hair, CA Hostetler, and SP Burton. 2013. "Hyperspectral aerosol optical depths from TCAP flights." *Journal of Geophysical Research–Atmospheres* 118(21): 12180-12194, doi:10.1002/2013JD020596.

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Zelenyuk, A, D Imre, J Wilson, Z Zhang, J Wang, and L Mueller. 2015. "Airborne single-particle mass spectrometers (SPLAT II & miniSPLAT) and new software for data visualization and analysis in a geo-spatial context." *Journal of the American Society of Mass Spectrometry* 26(2): 257-270, doi:10.1007/s13361-014-1043-4.

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Kollias, P, I Jo, P Borque, A Tatarevic, K Lamer, N Bharadwau, K Widener, K Johnson, EE Clothiaux. 2014. "Scanning ARM cloud radars. Part II: Data quality control and processing." *Journal of Atmospheric and Oceanic Technology* 31: 583-598, doi:10.1175/JTECH-D-13-00045.1.

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Shinozuka, Y, et al. 2013. "Hyperspectral aerosol optical depths from TCAP flights." *Journal of Geophysical Research* 118: 1-15, doi:10.1002/2013jd020596.

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Zelenyuk, A, D Imre, J Wilson, Z Zhang, J Wang, and K Mueller. 2015. "Airborne single-particle mass spectrometers (SPLAT II & miniSPLAT) and new software for data visualization and analysis in a geo-spatial context." *Journal of the American Society of Mass Spectrometry* 26(2): 257-270, doi:10.1007/s13361-014-1043-4.



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