

Aerosol and Cloud Experiments in Eastern North Atlantic (ACE-ENA) Field Campaign Report

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Executive Summary

With their extensive coverage, low clouds greatly impact global climate. Presently, low clouds are poorly represented in global climate models (GCMs), and the response of low clouds to changes in atmospheric greenhouse gases and aerosols remains the major source of uncertainty in climate simulations. The poor representations of low clouds in GCMs are in part due to inadequate observations of their microphysical and macrophysical structures, radiative effects, and the associated aerosol distribution and budget in regions where the aerosol impact is the greatest. The Eastern North Atlantic (ENA) is a region of persistent but diverse subtropical marine boundary layer (MBL) clouds, whose albedo and precipitation are highly susceptible to perturbations in aerosol properties. Boundary-layer aerosol in the ENA region is influenced by a variety of sources, leading to strong variations in cloud condensation nuclei (CCN) concentration and aerosol optical properties.

A permanent ENA site was established by the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility on Graciosa Island in the Azores, providing invaluable information on MBL aerosol and low clouds. At the same time, the vertical structures and horizontal variabilities of aerosol, trace gases, cloud, drizzle, and atmospheric thermodynamics are critically needed for understanding and quantifying the budget of MBL aerosol, the radiative properties, precipitation efficiency, and lifecycle of MBL clouds, and the cloud response to aerosol perturbations. Much of this data can be obtained only through aircraft-based measurements. In addition, the interconnected aerosol and cloud processes are best investigated by a study involving simultaneous in situ aerosol, cloud, and thermodynamics measurements. Furthermore, in situ measurements are also necessary for validating and improving ground-based retrieval algorithms at the ENA site.

The Aerosol and Cloud Experiments in Eastern North Atlantic (ACE-ENA) project was motivated by the need for comprehensive in situ characterizations of boundary-layer structure, and associated vertical distributions and horizontal variabilities of low clouds and aerosol over the Azores. The ARM Aerial Facility (AAF) Gulfstream-1 (G-1) aircraft was deployed during two intensive measurement periods (IOPs). The first deployment took place from June 21 to July 20, 2017, and the second one took place from January 15 to February 18, 2018. Flights were carried out in the Azores, near the ARM ENA site on Graciosa Island. Deployments during both seasons allow for examination of key aerosol and cloud processes under a variety of representative meteorological and cloud conditions. The science themes for the deployments include: (1) Budget of MBL CCN and its seasonal variation; (2) Effects of aerosol on cloud and precipitation; (3) Cloud microphysical and macrophysical structures, and entrainment mixing; (4) Advancing retrievals of turbulence, cloud, and drizzle; and (5) Model evaluation and processes studies.

A key advantage of the deployments is the strong synergy between the measurements onboard the G-1 and the routine measurements at the ENA site, including state-of-the-art profiling and scanning radars. The 3D cloud structures provided by the scanning radars will put the detailed in situ measurements into mesoscale and cloud lifecycle contexts. On the other hand, high quality, in situ measurements will enable validation and improvements of ground-based retrieval algorithms at the ENA site, leading to high-quality and statistically robust data sets from the routine measurements.

Acronyms and Abbreviations

2D-S	2-dimensional stereo probe
3D	three-dimensional
AAF	ARM Aerial Facility
ACAPEX	ARM Cloud Precipitation Experiment
ARM	Atmospheric Radiation Measurement
CAPS	cloud, aerosol, and precipitation spectrometer
CAS	cloud and aerosol spectrometer
CCD	charge-coupled device
CCN	cloud condensation nuclei
CPC	condensation particle counter
CVI	counter-flow virtual impactor
DOE	U.S. Department of Energy
ENA	Eastern North Atlantic
FCDP	fast cloud droplet probe
FIMS	fast integrated mobility spectrometer
FT	free troposphere
G-1	Gulfstream-1 aircraft
GCM	global climate model
GLOMAP	Global Model of Aerosol Processes
HOLODEC	holographic detector for clouds
HR-ToF-AMS	high-resolution time-of-flight aerosol mass spectrometer
HVPS-3	high-volume precipitation spectrometer version 3
IOP	intensive operational period
IPCC	Intergovernmental Panel on Climate Change
LES	large-eddy simulation
MBL	marine boundary layer
PCASP	passive cavity aerosol spectrometer
PILS	particle-into-liquid sampler
PSAP	particle soot absorption photometer
PTR-MS	proton transfer reaction-mass spectrometer
RH	relative humidity
SP2	single-particle soot photometer
TCAP	Two-Column Aerosol Project
TRAC	time-resolved aerosol collector
UCPC	ultrafine condensation particle counter
UHSAS	ultra-high-sensitivity aerosol spectrometer
VOC	volatile organic compound

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1.0 Background

1.1 Motivation

The responses of low cloud systems to changes in atmospheric greenhouse gases and aerosols are major sources of uncertainty that limit our ability to predict future climate (Bony and Dufresne 2005; Lohmann and Feichter 2005; Wyant et al. 2006). Climate models disagree substantially in the magnitude of cloud feedback for the regimes of subtropical marine low clouds (Bony and Dufresne 2005; Bony et al. 2006), and suffer from the so-called “too few, too bright” problem (Allan et al. 2007; Nam et al. 2012; Webb et al. 2013). The “too few” problem, an underestimate in cloud amount, allows more solar radiation to reach the surface. The “too bright” problem, an overestimate in cloud albedo due, for example, to an overestimate in the amount of liquid water within the cloud, causes more sunlight to be reflected. These inaccurate cloud properties lead to an apparently realistic radiation budget due to compensating errors, but overly reflective clouds may result in a significant underestimate in the strength of a negative cloud feedback process involving increased cloud liquid water with warming (Stephens 2010). Additionally, the interdependence between cloud macrophysical, microphysical, and radiative properties strongly links to the stages of warm cloud and precipitation evolution (Suzuki et al. 2009), and could be modified by ambient aerosols (Albrecht 1989; Pincus and Baker 1994; Lohmann and Feichter 2005; Feingold et al. 2010). Remote marine low cloud systems have large spatial coverage and are particularly susceptible to perturbations in aerosols associated with anthropogenic emissions because of their relatively low optical thickness and background aerosol concentrations (e.g., see Figure 1; Twomey 1977; Carslaw et al. 2013). Indeed, recent studies find that a large fraction of the global aerosol indirect forcing can be attributed to changes in marine low clouds (Kooperman et al. 2013), despite their relatively long distance from most anthropogenic sources. There remain large uncertainties in the magnitude of the global aerosol radiative forcing (Lohmann and Feichter 2005; Isaksen et al. 2009; IPCC 2013). Major contributions to this uncertainty derive from poor understanding of the cloud responses to aerosol changes (Rosenfeld et al. 2014b, 2014a) and the natural aerosol state that is being perturbed by anthropogenic emissions (Carslaw et al. 2013). These prompt the need for both long-term and comprehensive observations to understand key aerosol and cloud properties, and their controlling processes for better representations in climate models.

The ACE-ENA campaign was motivated by the need for comprehensive in situ characterizations of boundary-layer and lower free troposphere structure, and associated vertical distributions and horizontal variabilities of low clouds and aerosol over the Azores. The overarching scientific objective is to understand key processes that drive the properties and interactions of aerosol and cloud under a variety of representative meteorological and cloud conditions. An important consideration for this deployment is to provide high-quality, in situ measurements for validating and improving ground-based retrieval algorithms at the ENA site. This will lead to high-quality, long-term data sets from the routine ground-based remote sensing, which allow greater statistical reliability in the observed properties and relationships among aerosols, clouds, and precipitation than is possible with the aircraft measurements alone. The in situ data and improved algorithms will enable better use of the routine measurements for model evaluation.

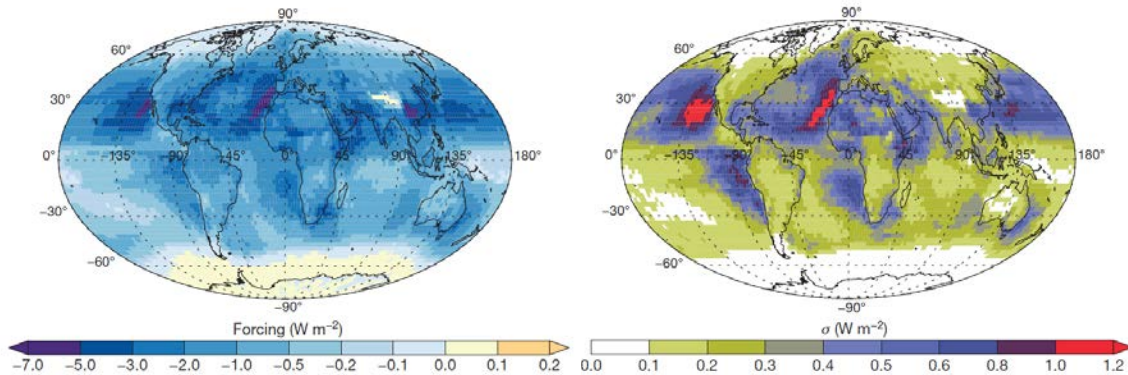


Figure 1. (a) Annual mean aerosol first indirect forcing, and (b) uncertainty in simulated first indirect forcing due to uncertainty in model (Global Model of Aerosol Processes [GLOMAP]) parameters. Note the large aerosol indirect forcing and uncertainty in the ENA. Adapted from Carslaw et al. (2013).

1.2 Deployment Strategies

Simultaneous characterizations of meteorological parameters, trace gases, aerosol, cloud, and drizzle fields were carried out onboard the ARM Aerial Facility Gulfstream-1 (G-1) aircraft near the ENA site. The campaign consisted of two intensive operational periods (IOPs), one from June 21 to July 20, 2017 during early summer, and the second one from January 15 to February 18, 2018 during the winter. Deployments during both seasons allow for examination of key aerosol and cloud processes under a variety of representative meteorological and cloud conditions, and for different aerosol sources and pathways. For example, during the summer months, the Azores are ideally located to sample overcast stratocumulus, and the transition to a broken trade cumulus regime; the winter frequently experiences maritime frontal clouds.

A key advantage of the deployments is the strong synergy between the in situ measurements onboard the G-1 and the ongoing measurements at the ENA site, including state-of-the-art profiling and scanning radars. The 3D cloud structures provided by the scanning radars put the detailed in situ measurements into mesoscale and cloud lifecycle contexts. On the other hand, high-quality, in situ measurements enable validation and improvements of ground-based retrieval algorithms at the ENA site, leading to high-quality, statistically robust data sets from the routine measurements. The ACE-ENA deployments, combined with the ongoing measurements at the ENA site, will have a long-lasting impact on the research and modeling of clouds and aerosols in remote marine environment.

1.3 Instruments and Measurements Onboard G-1

A list of the instruments and measurements is included in Table 1. Measurements onboard the G-1 included the standard meteorological, turbulence, and radiation (both up and downwelling) quantities, including measurements of sensible and latent heat fluxes. The size distribution of cloud droplets, drizzle, and rain drops were characterized using a combination of fast cloud droplet probe (FCDP), 2-dimensional stereo probe (2D-S), and high-volume precipitation spectrometer version 3 (HVPS-3). Liquid water content was measured using both a multi-element water content system (WCM-2000) and a Gerber (PVM-100a) probe. We also deployed a cloud, aerosol, and precipitation spectrometer (CAPS), which

provides redundant measurements of cloud droplet and drizzle drop size spectra and liquid water content. A novel holographic detector for clouds (HOLODEC) was deployed to sample an ensemble of hydrometeors in a localized volume ($\sim 20 \text{ cm}^3$) by digitally reconstructing interference patterns recorded by a charge-coupled device (CCD) camera (Fugal and Shaw, 2009). Under typical stratocumulus conditions each sampled region of cloud results in a statistically robust estimate of the cloud droplet size distribution, without the loss of information inherent in averaging over long distances.

Trace gas monitors that measure carbon monoxide (CO) and ozone (O₃) helped differentiate various air masses and identify the influences from anthropogenic emissions. A proton transfer reaction-mass spectrometer (PTR-MS) characterized important trace gas volatile organic compounds (VOCs), including key aerosol precursors and relevant reaction products. Comprehensive characterizations of aerosol included particle number concentration, size distribution, optical properties, and chemical composition. A fast integrated mobility spectrometer (FIMS), passive cavity aerosol spectrometer (PCASP), and cloud and aerosol spectrometer (CAS, part of the CAPS) provided size distribution from 10 nm to coarse size particles. A condensation particle counter (CPC) (for sizes $> 10 \text{ nm}$) and an ultrafine condensation particle counter (UCPC) (for sizes $> 3 \text{ nm}$) quantified total aerosol number concentrations. A single-particle soot photometer (SP2) was used to measure refractory black carbon concentrations, a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) characterized bulk aerosol composition and size, and a particle-into-liquid sampler (PILS) coupled to ion chromatography characterized sub-micrometer water-soluble aerosol chemical composition (i.e., inorganics, organic acids, amines). A time-resolved aerosol collector (TRAC) sampler was deployed to collect atmospheric particles for multiple off-line post-campaign laboratory analyses (Laskin et al. 2012; Moffet et al. 2013). Optical properties for aerosol absorption and scattering were measured by a particle soot absorption photometer (PSAP) and nephelometer, respectively. A dual-column CCN counter was used to quantify cloud condensation nuclei concentrations.

Aerosol size distribution measured by the FIMS and total number concentration measurements alternated between ambient samples and those processed by a thermal denuder, which allows volatility-based separation by exploiting the higher volatility of organics and sulfate versus sea salt and refractory black carbon (Clarke et al. 2013), which remain in the aerosol phase at the denuder temperature of 350 °C. We also used both an isokinetic inlet and a counter-flow virtual impactor (CVI) inlet to sample aerosols, similar to the previous G-1 deployments (e.g. the Two-Column Aerosol Project [TCAP] and the ARM Cloud Precipitation Experiment [ACAPEX]). When used to sample cloud droplets, the CVI allows determination of particle composition and size spectrum of cloud droplet residuals by the HR-ToF-AMS, PILS, SP2, and FIMS.

The measurements of meteorological parameters, cloud microphysics, and aerosol number concentration and size distribution were carried out at a frequency of 1 Hz or higher. Cloud droplet spectrum were measured by the FCDP at a frequency of 10 Hz. In addition, both FCDP and UHSAS have the “particle by particle” sampling capability, therefore allowing measurements at even higher frequency. The Gerber probe was operated at a frequency of 100 Hz. These high-resolution cloud microphysics measurements, combined with the unique data set provided by the HOLODEC, are critical for understanding and quantifying the microphysical impact of the entrainment mixing processes.

Table 1. A list of instrumentation onboard the G-1 aircraft in addition to the standard G-1 atmospheric state and aircraft state package.

Instrument	Measurement	Facility/Potential Contact
Aircraft integrated meteorological measurement system (AIMMS-20)	5-port air motion sensing: true airspeed, altitude, angle of attack, side slip, temperature, and relative humidity (RH).	AAF
Gust probe	5-port air motion sensing: true air speed, angle of attack, side slip	AAF
Multifilter radiometer (MFR)	Upwelling shortwave radiation global, 415, 500, 615, 673, 870, 940, 1625 nm spectral channels	AAF
Sunshine pyranometer, unshaded and shaded	Broadband upwelling and downwelling shortwave radiation global, broadband downwelling shortwave radiation global and diffuse	AAF
Fast cloud droplet probe (FCDP)	Cloud particles size distribution 2 to 50 μm	AAF
2-dimensional stereo probe (2D-S)	Cloud particles size distribution 10 to 3,000 μm	AAF
High-volume precipitation spectrometer version 3 (HVPS-3)	Cloud particles size distribution 150 to 19,600 μm	AAF
Holographic detector for clouds (HOLODEC)	Cloud particle size distribution 6 to 1,000 μm	AAF
Multi-element water content system (WCM-2000)	Liquid, total, and ice water content	AAF
Particle volume monitor-100A (PVM-100A) (Gerber probe)	Cloud liquid water content	AAF
Cloud, aerosol, and precipitation spectrometer (CAPS)	Aerosol particle and cloud hydrometeor size distributions from 0.51 to 50 μm , precipitation size distributions from 25 μm to 1550 μm , and liquid water content from 0.01 to 3 g/m^3	AAF
Trace gas instrument system for CO and O ₃	Concentrations of CO and O ₃	AAF
Proton reaction mass spectrometer (PTR-MS)	Volatile organic compounds (VOCs)	PNNL (John Shilling)
Fast integrated mobility spectrometer (FIMS)	Aerosol size distribution, 0.01 to 0.5 μm at 1 Hz	WUSTL (Jian Wang)
Passive cavity aerosol spectrometer-100X (PCASP)	Size distribution, 0.1 to 3 μm	AAF
Condensation Particle Counter (CPC)	Total aerosol concentration >0.010 μm	AAF
Ultrafine condensation particle counter (CPC)	Total aerosol concentration >0.004 μm	AAF
Dual-column cloud condensation nuclei counter (CCN)	CCN concentrations at two specified supersaturations	AAF
3-wavelength particle soot/absorption photometer (PSAP)	Aerosol absorption coefficient at 462, 523, 648 nm	AAF
Integrating nephelometer, 3-wavelengths, Model 3563	Aerosol scattering coefficient at 450, 550, 700 nm	AAF

Instrument	Measurement	Facility/Potential Contact
Single-particle soot photometer (SP2)	Soot spectrometry	AAF
High-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS)	Non-refractory aerosol composition	PNNL (John Shilling)
Particle-into-liquid sampler	Water-soluble aerosol composition	Colorado State University (Amy Sullivan)
TRAC sampling system	Particle collection on substrates	PNNL/EMSL (Alex Laskin)
Counter-flow virtual impactor inlet (CVI)	Sampling of cloud droplet residuals	AAF
Thermal denuder	Sampling of non-volatile component of aerosol particles	WUSTL (Jian Wang)

1.4 Flight Plans

The basic flight patterns included spirals to obtain vertical profiles of aerosol and clouds, and legs at multiple altitudes, including below cloud, inside cloud, at the cloud top, and in the free troposphere. Each leg was several tens of kilometers in length to capture the mesoscale variabilities of aerosol and cloud fields. The legs were flown both perpendicular to and along the wind direction. These measurements provided detailed characterization of boundary-layer and lower free troposphere structure, and associated vertical distributions and horizontal variations of low clouds and aerosols in the Azores under representative meteorological and cloud conditions. The G-1 was stationed at the Lajes airport on the island of Terceira, which is about 90 km from the ENA site.



Figure 2. ACE-ENA team members and AAF staff participated in the second IOP.

2.0 Notable Events or Highlights

During ACE-ENA, aerosol, cloud, and precipitation under a variety of conditions were sampled (Table 2). Low-level clouds dominated during most of the flights. Higher cloud fraction was observed during IOP2 (winter season) compared to that during the IOP1 (summer season). A large fraction of the clouds sampled were precipitating (Figure 3).

Table 2. Conditions sampled during ACE-ENA flights.

Conditions Sampled	IOP1, Flight number	IOP2, Flight number
Aerosol (Mostly clear)	2, 6, 11	17
Thin Stratus Clouds	1, 3, 4, 5, 7, 9, 11, 15	6, 9, 13, 15
Solid Stratocumulus	10, 12, 16	8
Multi-Layer Stratocumulus	13, 14	3, 7, 11
Drizzling Stratocumulus/Cumulus	8, 17, 18, 19, 20	1, 2, 4, 5, 12, 14, 16, 18, 19

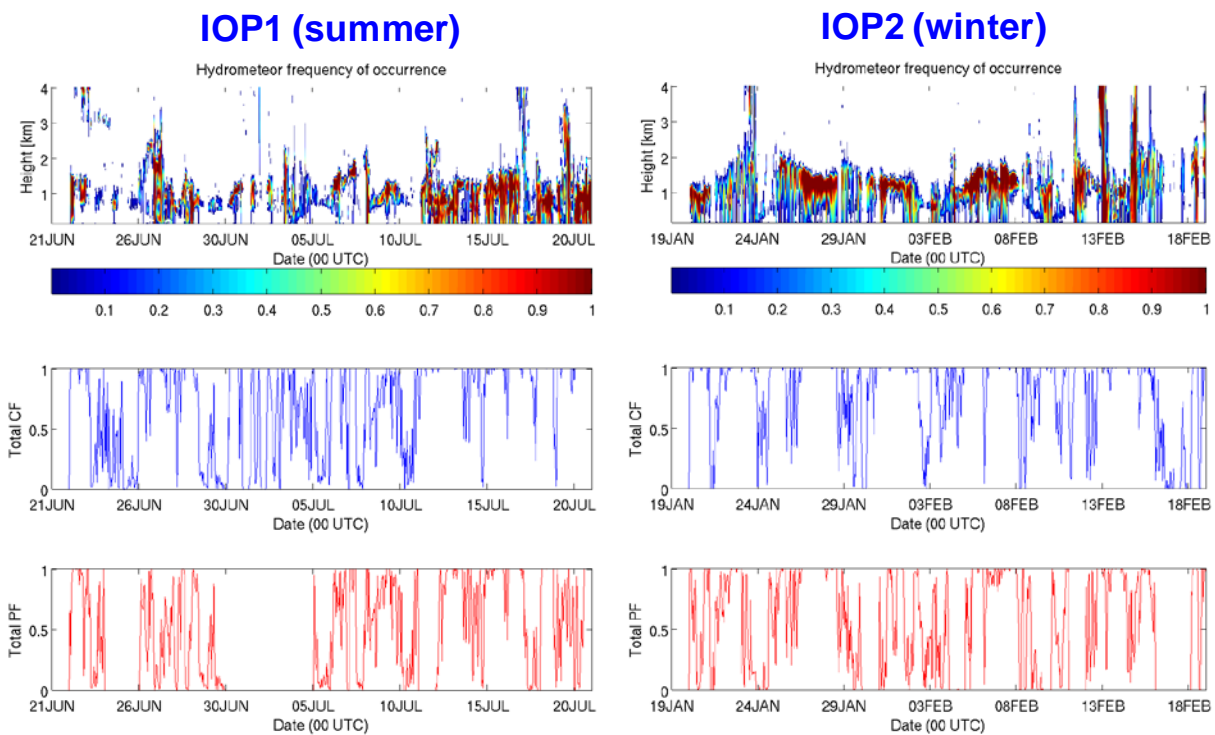


Figure 3. Hydrometeor frequency (top), total cloud fraction (middle), and total precipitation fraction (bottom) for the periods of first IOP (left) and second IOP (right).

3.0 Results

A total of 20 research flights, a total of 77 flight hours, were carried out during the first IOP of ACE-ENA. During the second IOP, the G-1 flew 19 research flights, a total of 76.2 hours. Most of the flights were near Graciosa Island and coordinated with measurements at the ENA site. These flights provided comprehensive in situ characterizations of boundary-layer and lower free troposphere (FT) structure, and associated vertical distributions and horizontal variabilities of low clouds and aerosol over the Azores, which are needed to understand key processes that drive the properties and interactions of aerosol and cloud under a variety of representative meteorological and cloud conditions. These flights also provided high-quality, in situ measurements for validating and improving ground-based retrieval algorithms at the ENA site. This will lead to high-quality, long-term data sets from the routine ground-based remote sensing, which allow greater statistical reliability in the observed properties and relationships among aerosols, clouds, and precipitation than is possible with the aircraft measurements alone. The in situ data and improved algorithms will enable better use of the routine measurements for model evaluation. The measurements onboard the G-1 and those at the ENA site will allow us to address the following scientific questions and objectives, which are organized into five themes:

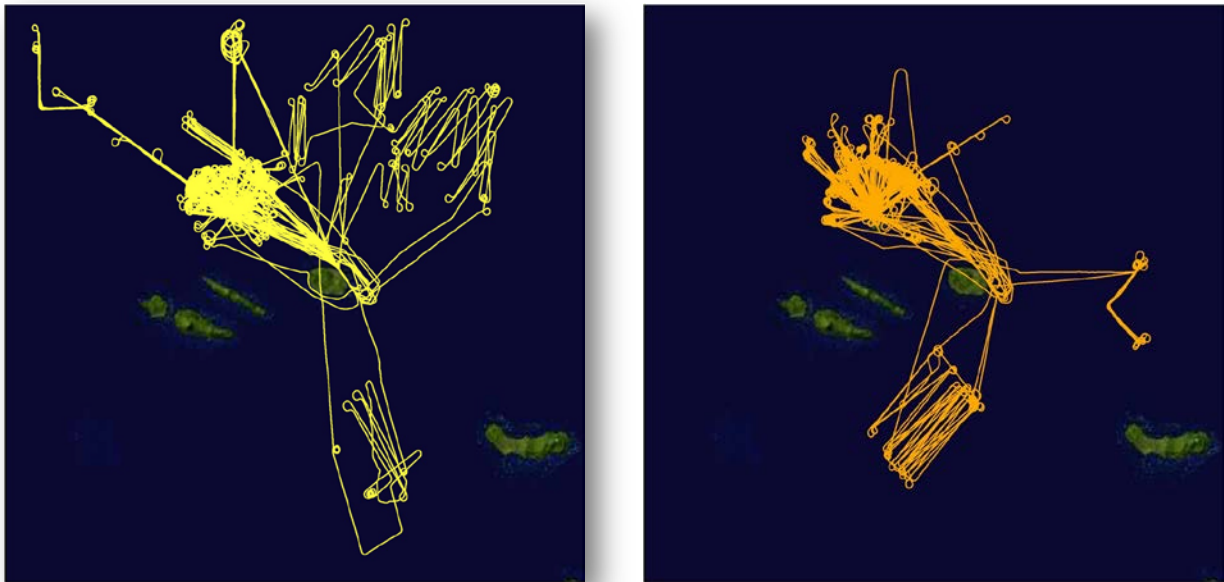


Figure 4. Flight tracks for 20 flights during the first IOP (left) and 19 flights during the second IOP (right). The G-1 aircraft was stationed at the Lajes airport on the island of Terceira, which is about 90 km from the ENA site. Most of the sampling was carried out near the ENA site on the island of Graciosa.

Budget of MBL CCN and its Seasonal Variation

- What are the contributions from different sources, including sea spray aerosol, long-range transport, and new particle formation? What are the seasonal variations of the characteristics and contributions of various sources?

- How does removal of CCN by droplet coalescence control the CCN population in the MBL for representative cloud regimes?

Effects of Aerosol on Clouds and Precipitation

- How can ground-based lidar and CCN measurements be used to better infer CCN concentration at cloud base?
- How does the CCN budget affect cloud microphysics? Do high CCN concentrations lead to increased cloud droplet concentrations and suppressed precipitation? Is precipitation susceptibility to CCN weaker in the deep open cells observed in the Azores?

Cloud Microphysical and Macrophysical Structures, and Entrainment Mixing

- What are the mesoscale variabilities of cloud microphysics and vertical velocity and how do they influence drizzle mesoscale organization and rates? What are the thermodynamic and spatial characteristics of cold pools and how do they relate to the properties and mesoscale organizations of cloud and drizzle?
- What are the relationships between the entrainment rate, thermodynamic stability, wind shear above cloud top, and coupling structure below cloud base? What is the prevalent entrainment mixing mechanism, and what are the controlling factors? What are the microphysical effects of entrainment mixing?

Advancing Retrievals of Turbulence, Cloud, and Drizzle

- Validating and quantifying the uncertainties in turbulence, cloud and drizzle microphysical properties retrieved from vertically pointing observations.
- Validating and improving 3D cloud and drizzle retrievals from scanning radars.

Model Evaluation and Processes Studies

- Comparison of the airborne data with predictions of global models using “nudged” or “specified” meteorology and local simulations with large-eddy simulation (LES) and WRF-Chem models.
- Examining the CCN budget terms and processes driving the vertical structure and mesoscale variation of aerosol, cloud, and drizzle fields using validated/constrained GCM and LES model simulations.

4.0 ACE-ENA Publications

4.1 Journal Articles/Manuscripts

Zheng, GJ, Y Wang, AC Aiken, F Gallo, MP Jensen, P Kollias, CG Kuang, E Luke, S Springston, J Uin, R Wood, and J Wang. 2018. “Marine boundary layer aerosol in the eastern North Atlantic: seasonal variations and key controlling processes.” *Atmospheric Chemistry and Physics* 18(23): 17615–17635, [10.5194/acp-18-17615-2018](https://doi.org/10.5194/acp-18-17615-2018)

4.2 Meeting Abstracts/Presentations/Posters

Wang, J, et al. 2017. “Aerosol, cloud, and precipitation interactions in Eastern North Atlantic.” American Geophysical Union falling meeting.

Wood, R, et al. 2018. “Aerosol, cloud, and precipitation interactions over the Eastern North Atlantic.” American Meteorological Society cloud physics meeting.

Wood, R, et al. 2018. “Aerosol, cloud, and precipitation interactions over the Eastern North Atlantic.” American Geophysical Union falling meeting.

Zawadowicz, M. et al. 2018. “Aerosol and cloud chemistry during the ACE-ENA campaign.” American Geophysical Union falling meeting.

Wang, Y, et al. 2018. “Cold air outbreak leads to new particle formation over the Eastern North Atlantic.” American Geophysical Union falling meeting.

Veghte, D et al. 2018. “Composition and Properties of Particles Sampled at Different Altitudes in the North Atlantic,” American Geophysical Union falling meeting.

Miller, M. et al. 2018. “Relating Drizzle Evaporation and Sub-Cloud Decoupling: Observations and Simulations during ACE-ENA.” American Geophysical Union falling meeting.

Yum, SS, et al. 2018. “Vertical Variation of Cloud Microphysical Relationships and Their Implication on Entrainment and Mixing Processes in Stratocumulus Clouds Measured during the ACE-ENA Campaign.” American Geophysical Union falling meeting.

Zheng, GJ, et al. 2018. “Long-range transported biomass burning aerosol observed in Eastern North Atlantic.” American Geophysical Union falling meeting.

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