Entrainment and aerosol effects on marine boundary-layer clouds: An investigation using ACE-ENA data from HOLODEC, G1, Pico and ACTOS
Final Report on Project Activities

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Abstract
Marine boundary-layer clouds cover large regions of the globe and are known to strongly influence radiative balances. The microphysical properties and persistence of these clouds are tightly coupled with cloud-top entrainment and aerosol properties within both the boundary layer and the overlying free troposphere. This work addressed the microphysical response to entrainment and aerosol properties in marine stratocumulus clouds, using data from the Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) project. Specifically, emphasis was placed on 1. extensive in situ measurements taken with the Atmospheric Radiation Measurement (ARM) G1 aircraft using the Holographic Detector for Clouds (Holodec) instrument; 2. airborne measurements obtained with the helicopter-borne Airborne Cloud Turbulence Observation System (ACTOS) operated by the Leibniz Institute for Tropospheric Research (TROPOS) during the first phase of ACE-ENA; and 3. above-boundary-layer, mountain-top measurements taken at the Pico Mountain Observatory (OMP) by the research group of the co-investigator and scientists from TROPOS during the first phase of ACE-ENA. The work is aligned with the topic "Warm Boundary-Layer Atmospheric Processes." Key results include the following. Data from the Holographic Detector for Clouds (Holodec) were reprocessed, refined, and validated through a careful instrument intercomparison, resulting in a high-quality dataset available on the ARM archive for ACE-ENA. High-resolution ACTOS measurements of thermodynamics, microphysical, and turbulence properties were analyzed to explore the behavior of the entrainment velocity at cloud top. Analysis of the vertical variability of cloud droplet size distribution shape suggests that mixing is more inhomogeneous near cloud top, and more homogeneous deeper into the cloud. Analysis of aerosol measurements from OMP explored the mixing state and cloud condensation nucleus properties, as well as the implications for radiative forcing. Finally, a machine learning algorithm was developed for identifying characteristic cloud droplet size distributions, and was employed to explore microphysical regimes in stratocumulus clouds observed during ACE-ENA. The characteristic size distributions are narrow, and only when spatially averaged do they produce the broad "gamma" distributions typically assumed in models. This implies that precipitation development should account for variability and correlations in the distribution shape as well as the number concentration of cloud droplets.

Overview

This document summarizes activities supported by the DOE Atmospheric System Research program under grant number DE-SC0020053 (beginning August 15, 2020).

Personnel from the groups of PI's Mazzoleni and Shaw at Michigan Technological University include:

- Nithin Allwayin (advisor R. Shaw) – will complete PhD in Summer 2024.
- Shreya Joshi (advisor C. Mazzoleni) – will complete PhD in Summer 2024.
- Susan Mathai (advisor C. Mazzoleni) – completed PhD in Fall 2023.
- Megan Morgenstern (advisor C. Mazzoleni) – completed MS in Spring 2022.
- Jae Min Yeom, postdoctoral scientist (advisor R. Shaw) March 2020 through present.

Collaborators on the project include:

- Dr. Swarup China, Pacific Northwest National Laboratory
- Dr. Neel Desai and Dr. Yangang Liu, Brookhaven National Laboratory
- Dr. Andre Ehrlich and Prof. Manfred Wendisch, Leipzig University
- Prof. Paolo Fialho and Diamantino Henriques, Azores University
- Dr. Virendra Ghate, Argonne National Laboratory
- Dr. Susanne Glienke, Pacific Northwest National Laboratory
- Dr. Silvia Henning, Dr. Holger Siebert, and Dr. Birgit Wehner, Leibniz Institute for Tropospheric Research (TROPOS)
- Prof. Pavlos Kollias, Stony Brook University
- Ed Luke, Brookhaven National Laboratory
- Prof. Szymon Malinowski, University of Warsaw
- Prof. Juan-Pedro Mellado, Polytechnic University of Catalonia
- Prof. Seong-Soo Yum, Yonsei University

Our main activities and achievements during this time period are detailed in the Publications listed next and available on the OSTI website, and briefly summarized below.

Publications

Allwayin, N., M. L. Larsen, A. G. Shaw, R. A. Shaw. Automated identification of characteristic droplet size distributions in stratocumulus clouds utilizing a data clustering algorithm. *Artificial Intelligence for the Earth Systems*, **1**, https://doi.org/10.1175/AIES-D-22-0003.1 (2022).

Allwayin, N., M. L. Larsen, S. Glienke, and R. A. Shaw: Locally narrow droplet size distributions are ubiquitous in stratocumulus clouds. Science, in review (first revision returned).

Cheng, Z., M. Morgenstern, B. Zhang, M. Fraund, N. N. Lata, R. Brimberry, M. A. Marcus, L. Mazzoleni, P. Fialho, S. Henning, B. Wehner, C. Mazzoleni, and S. China. Particle phase-state variability in the North Atlantic free troposphere during summertime is determined by atmospheric transport patterns and sources. *Atmospheric Chemistry and Physics*, **22**, 9033-9057 (2022).

Cheng, Z., M. Morgenstern, S. Henning, B. Zhang, G. C. Roberts, M. Fraund, M. A. Marcus, N. N. Lata, P. Fialho, L. Mazzoleni, B. Wehner, C. Mazzoleni, S. China. Cloud condensation nuclei activity of internally mixed particle populations at a remote marine free troposphere site in the North Atlantic Ocean. *Science of the Total Environment*, **904**, 166865 (2023).

Desai, N., Y. Liu, S. Glienke, R. A. Shaw, C. Lu, J. Wang, and S. Gao. Vertical variation of turbulent entrainment mixing processes in marine stratocumulus clouds using high-resolution digital holography. *Journal of Geophysical Research*, **126**, e2020JD033527 (2021).

Glienke, S., N. Allwayin, F. Mei, J. M. Comstock, R. A. Shaw, J. Tomlinson, and B. Schmid. Performance of optical sensors for cloud measurements deployed by the ARM Aerial Facility during ACE-ENA. *Applied Optics*, **62**, 5282-5293 (2023).

La, I. Y., S. S. Yum, J. M. Yeom, and R. A. Shaw. Influence of entrainment on centimeter-scale cloud microphysics in marine stratocumulus clouds observed during CSET. *Journal of the Atmospheric Sciences*, **79**, 2935-2948 (2022).

Morgenstern M., Z. Cheng, N. N. Lata, S. Mathai, R. P. Reveggino, P. Fialho, D. Henriques, S. Schum, A. Baccarini, L. Mazzoleni, S. China, B. Zhang, S. Joshi, and C. Mazzoleni. Optical and single-particle properties of North Atlantic free tropospheric aerosols and implications for aerosol direct radiative forcing. In preparation for *Environmental Science: Atmospheres*.

Siebert, H., K. Szodry, U. Egerer, B. Wehner, S. Henning, K. Chevalier, J. Lückerath, O. Welz, K. Weinhold, F. Lauermann, M. Gottschalk, A. Ehrlich, M. Wendisch, P. Fialho, G. Roberts, N. Allwayin, S. Schum, R. A. Shaw, C. Mazzoleni, L. Mazzoleni, J. L. Nowak, S. P. Malinowski, K. Karpinska, W. Kumala, D. Czyzewska, E. P. Luke, P. Kollias, R. Wood, and J. P. Mellado. Observations of aerosol, cloud, turbulence, and radiation properties at the top of the marine boundary layer over the Eastern North Atlantic Ocean: The ACORES campaign. *Bulletin of the American Meteorological Society*, **102**, E123–E147 (2021).

Wang, J., R. Wood, M. P. Jensen, J. C. Chiu, Y. Liu, K. Lamer, N. Desai, S. E. Giangrande, D. A. Knopf, P. Kollias, A. Laskin, X. Liu, C. Lu, D. Mechem, F. Mei, M. Starzec, J. Tomlinson, Y. Wang, S. S. Yum, G. Zheng, A. C. Aiken, E. B. Azevedo, Y. Blanchard, S. China, X. Dong, F. Gallo, S. Gao, V. P. Ghate, S. Glienke, L. Goldberger, J. C. Hardin, C. Kuang, E. P. Luke, A. A. Matthews, M. A. Miller, R. Moffet, M. Pekour, B. Schmid, A. J. Sedlacek, R. A. Shaw, J. E. Shilling, A. Sullivan, K. Suski, D. P. Veghte, R. Weber, M. Wyant, J. Yeom, M. Zawadowicz, and Z. Zhang. Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA). *Bulletin of the American Meteorological Society*, **103**, E619-E641 (2022).

Yeom, J. M., S. S. Yum, R. A. Shaw, I. La, J. Wang, C. Lu, Y. Liu, F. Mei, B. Schmid, and A. Matthews. Vertical variations of cloud microphysical relationships in marine stratocumulus clouds observed during the ACE-ENA campaign. *Journal of Geophysical Research*, **126**, doi:10.1029/2021JD034700 (2021).

Yeom, J., K.-E. Szodry, H. Siebert, A. Ehrlich, J. P. Mellado, R. A. Shaw, and S. S. Yum. High-resolution measurements of microphysics and entrainment in marine stratocumulus clouds. *Quarterly Journal of the Royal Meteorological Society*, https://doi.org/10.1002/qj.4586 (2023).

Summary of Project Activities

Improved Processing of Holodec Data from ACE-ENA

A foundational part of the work accomplished involved refined processing of the Holodec data from ACE-ENA to address noise in the large-particle counts. This was accomplished by training a convolution neural network to correctly recognize noise artifacts versus real droplets, and then applying it to the full dataset. The updated size distribution files were submitted to the ARM archive so that the highest-quality data are available to the full scientific community. Figures 1 and 2 below illustrate the improvements for one of the flights. The work was conducted in collaboration with Dr. Susanne Glienke from PNNL.

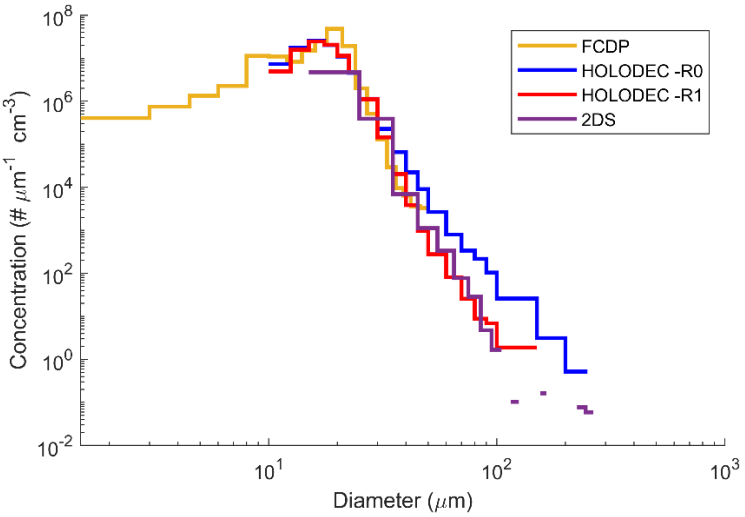

Figure 1. Droplet size distributions from the FCDP, the 2DS, and for the initial (R0) and improved (R1) processing of Holodec data.

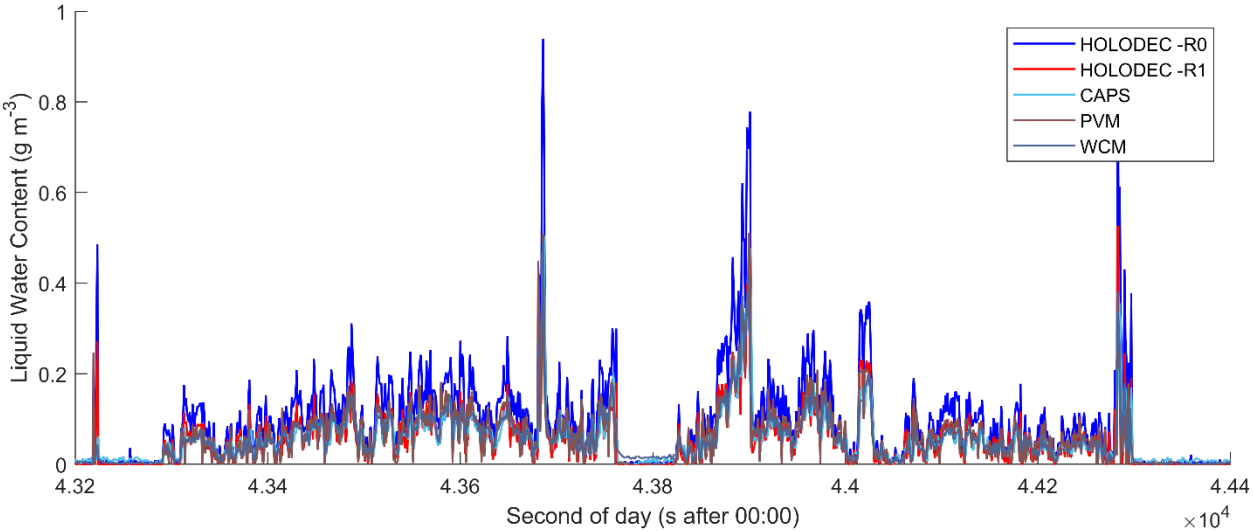

Figure 2. Liquid water content time series for the CAPS, PVM, and WCM, as well as for the initial (R0) and improved (R1) processing of Holodec data.

Cloud microphysics and its relation to entrainment at the marine boundary layer top over the Northern Atlantic Ocean

High-resolution measurements from the Airborne Cloud-Turbulence Observation System (ACTOS) during the Azores Stratocumulus Measurements of Radiation, Turbulence and Aerosols (ACORES) campaign (Siebert et al. 2021) were analyzed for an investigation of the vertical profiles of microphysical properties and entrainment velocity (W_e) in marine stratocumulus clouds. The vertical profiles show the transition from the cloudy layer to free troposphere with nearly linear profiles of total water mixing ratio, liquid water potential temperature and virtual potential temperature, but the thickness of entrainment interfacial layer varies significantly. Sharp transitions of cloud microphysical and optical properties within a single horizontal flight leg are found in one stratocumulus cloud system. They seem to be related to the local environmental conditions, such as the wind shear and turbulent dissipation rate. W_e values estimated by three methods show consistent tendencies in general and are clearly related to the local environmental conditions, such as vertical shear of the horizontal wind and turbulence intensity. However, the magnitudes of W_e values differ by up to two orders of magnitude depending on the methods, which suggests that the estimation of W_e from in situ measurements is still a challenge. Analysis of the microphysical response to entrainment suggests that inhomogeneous mixing occurs dominantly. On the other hand, the analysis results for the clouds under more humid conditions indicate a higher likelihood of homogeneous mixing.

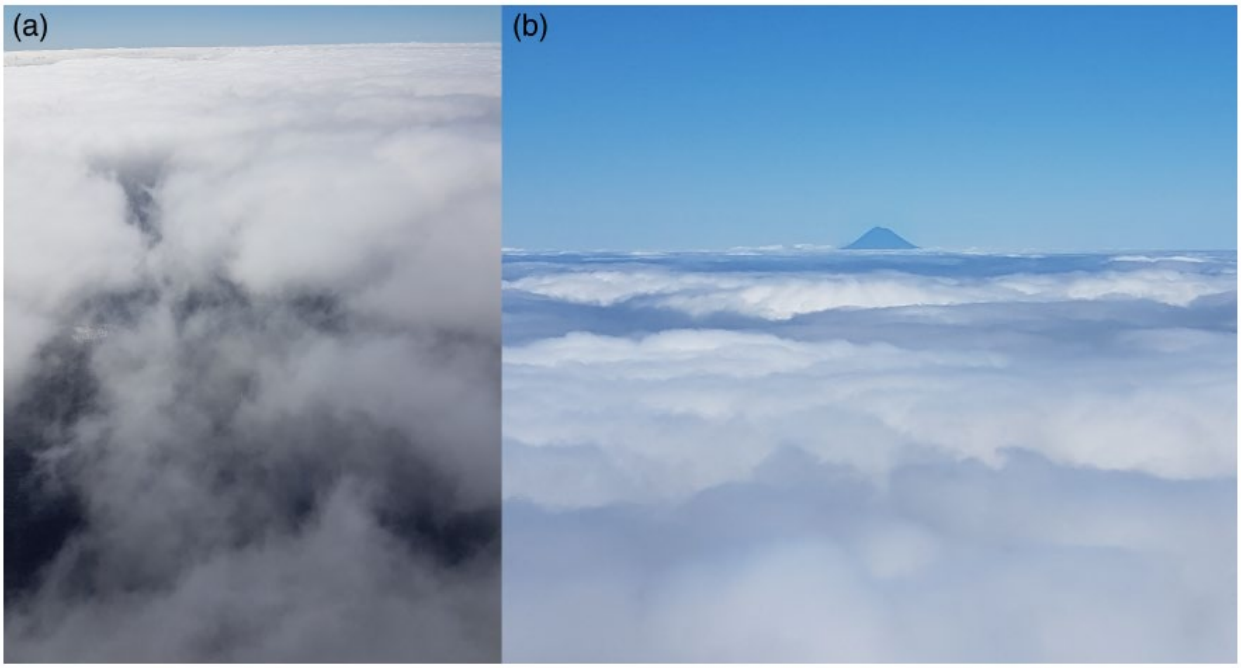

Figure 3. Photographs of the stratocumulus cloud top taken during the flight on 9 July 2017. The right photograph shows Mount Pico in the distance. Photo credit: R. A. Shaw, Michigan Technological University. From Yeom et al. (2023).

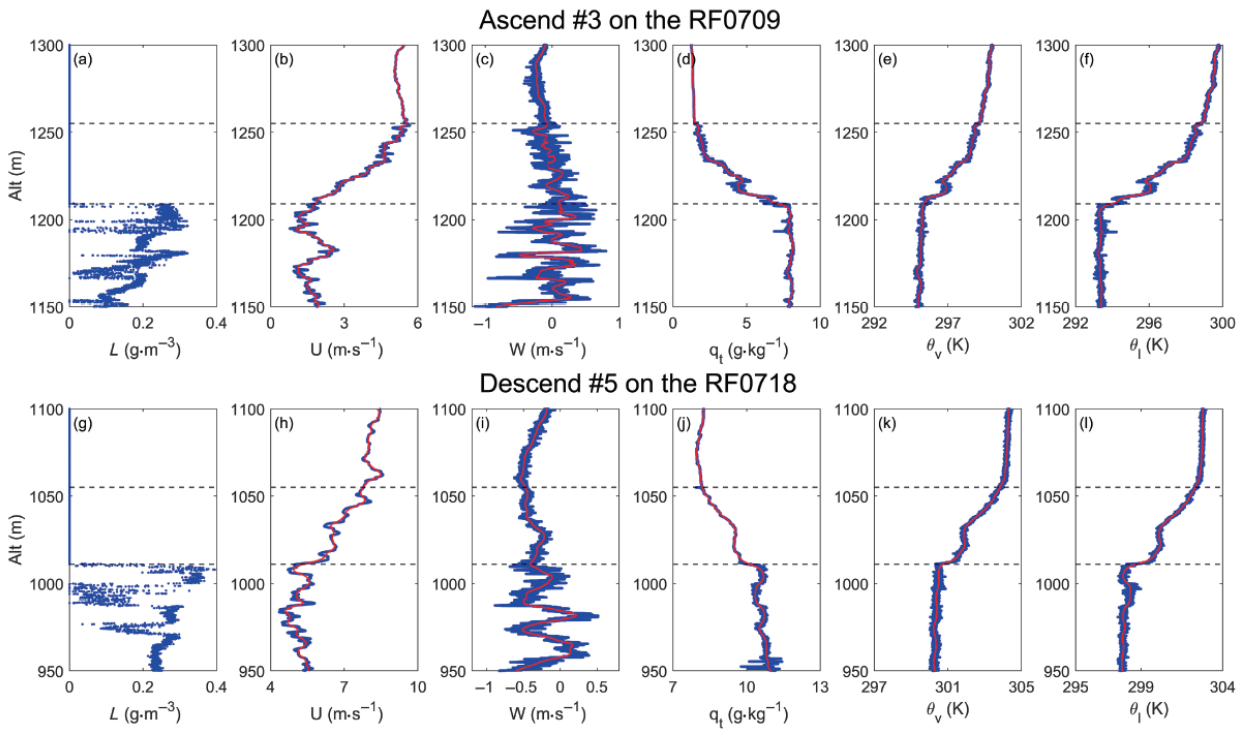

Figure 4. Two examples of the vertical profiles for liquid water content (L), horizontal wind velocity (U), vertical velocity (W), total water mixing ratio (q_t), virtual potential temperature (θ_v), and liquid water potential temperature (θ_l) (blue: raw, and red: filtered data with cut-off frequency=0.1Hz) observed during (a–f) an ascending porpoise profile on the 9 July, 2017 flight and (g–l) the descending porpoise profile on the 18 July flight. Dashed lines indicate the cloud top and the top of the entrainment interfacial layer. From Yeom et al. (2023).

Vertical variation of cloud microphysical relationships in stratocumulus clouds measured during the ACE-ENA campaign

We examined the vertical variations of cloud microphysical relationships and their implications to cloud microphysical processes in marine stratocumulus clouds using in-situ aircraft observations during the Aerosol and Cloud Experiments in Eastern North Atlantic (ACE-ENA) field campaign (Wang et al. 2022). A new diagram with a coordinate system based on cloud droplet liquid water content (L_c) and phase relaxation time scale is proposed to investigate mixing mechanisms (see Figure 5 below). This new diagram analysis shows that the inhomogeneous mixing trait is dominant near the cloud top, but homogeneous mixing trait is stronger at lower altitudes. The relevant scale parameters (i.e., transition length scale and transition scale number) also indicate a high likelihood of inhomogeneous mixing. The relationship between L_c and standard deviation of droplet radius (σ_r) clearly shows a vertical transition: the correlation between L_c and σ_r is positive at lower cloud altitudes, but it becomes negative as altitude increases. Such a vertical transition is consistent with the vertical circulation mixing, modulating the cloud microphysical relationships to suggest homogeneous mixing at a significant depth from the cloud top.

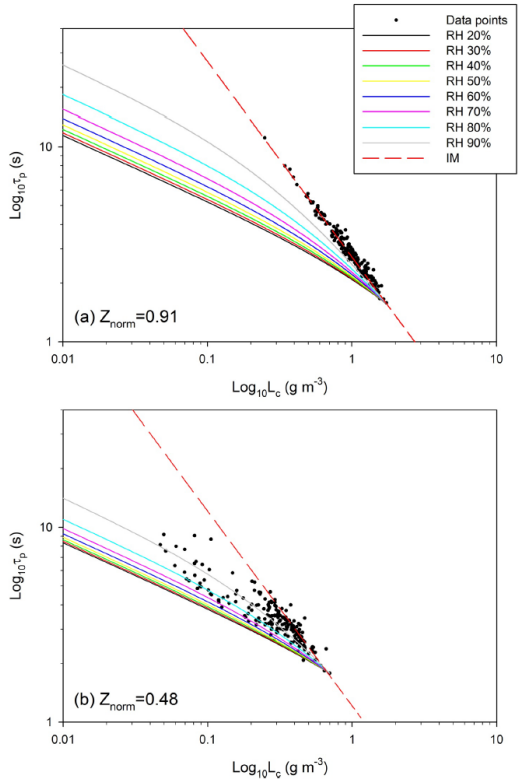

Figure 5. An example of a new type of mixing diagram, where L_c is cloud droplet liquid water content and τ_p is the phase relaxation time. The dashed line indicates the expected behavior for extreme inhomogeneous mixing with varying mixing fraction, and the solid lines indicate the expected behavior for homogeneous mixing with varying mixing fraction, for eight different relative humidity values assumed for the entrained air. The data are from a horizontal flight leg through stratocumulus clouds sampled during ACE-ENA by the G1 aircraft on 18 July 2017. From Yeom et al. (2021).

CCN activity of free tropospheric aerosols at the Observatory of Mt. Pico (2225 m a.s.l.) in the Azores

In the study by Cheng et al. (2023) we investigated the physicochemical properties and the cloud condensation nucleus activity of aerosols transported to the Azores in the free troposphere. Aerosols samples were collected at the Observatory of Mount Pico (OMP) during the ACE-ENA summer 2017 campaign. During the same period, we also operated a cloud condensation nucleus counter. The PNNL computer-controlled scanning electron microscope with an energy-dispersive X-ray spectrometer (CCSEM-EDX) was used to determine the chemical composition, the mixing state, and the hygroscopicity parameter, *k*, of individual particles. The FLEXible PARTicle Lagrangian particle dispersion model (FLEXPART) was used to determine the transport path to the site. The most abundant particles species were carbonaceous and salts. Most particles were internally mixed as expected for long range transported aerosols. The hygroscopicity of the particles were found to be in line with values typically measured over the north Atlantic. More hygroscopic particles were found when salt particles were more abundant, for example, during recirculation episodes above the North Atlantic as determined by FLEXPART. The *k* values calculated from the CCSEM-EDX overlapped with the lower range of *k* measured by the cloud condensation nucleus counter below 0.15 % supersaturation (Figure 6 below). The results suggest that single particle analysis can be used to predict the hygroscopicity of aerosols.

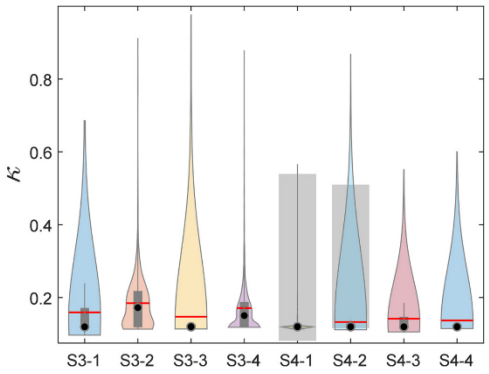

Figure 6. Violin plot of the hygroscopicity parameter, *k*, for particles analyzed using the CCSEM-EDX for different samples (Si-j) (red lines indicate means, black dots medians, small gray rectangles interquartile ranges, gray vertical lines 95 % confidence intervals). The rectangular gray shaded areas for S4-1 and S4-2 represent the range of *k* values derived from the cloud condensation nucleus counter. From Cheng et al. (2023).

Phase state and mixing state of free tropospheric aerosols at the Observatory of Mt. Pico (2225 m a.s.l.) in the Azores

In a study by Cheng et al. (2022) we studied the phase state of particles transported to OMP over three summers (2014, 2015, and 2017), the last one overlapping with the ACE-ENA campaign. The CCSEM-EDX was used in combination with near-edge X-ray absorption fine structure spectroscopy to determine the phase state and the chemical composition of individual particles. As in the previous study (Cheng et al., 2023), most particles were found to be carbonaceous or salt in nature. The presence of inorganic inclusions resulted in particles mostly in the liquid phase; however, the viscosity of the particles depended on transport paths and times. A wide range of viscosity values were found for particles transported in wildfire plumes; example of tilted view images used for the analysis with respective aspect ratio values (horizontal to vertical size ratios) are shown in Figure 7 below.

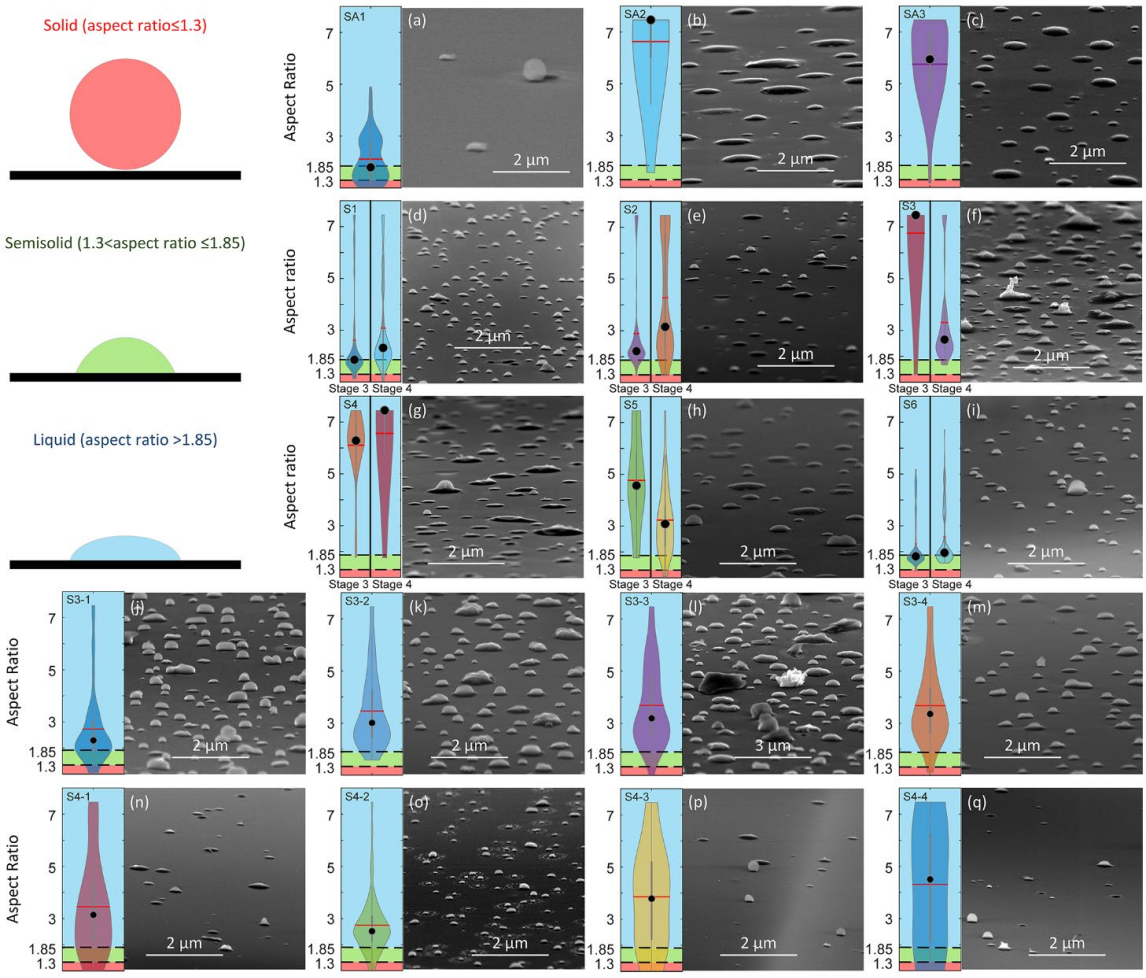

Figure 7. Examples of tilted view scanning electron microscopy images used to determine the aspect ratio that particles took at impaction on the substrate at the time of sampling. Particles with lower aspect ratio are considered solid, while particles with higher aspect ratio are less viscous. Violin plots of the aspect ratio are also reported. 2014 (a–c), 2015 (d–i), and 2017 (j–q). The shaded regions correspond to the different phase states (red: solid state; green: semisolid state; blue: liquid state). Figure from Cheng et al. (2022).

Radiative forcing of free tropospheric aerosols at the Observatory of Mt. Pico (2225 m a.s.l.) in the Azores

In a study by Morgenstern et al. (in preparation), we analyzed the physicochemical and optical properties of well-aged aerosols which traveled in the free troposphere to OMP in the summer of 2015. The physicochemical properties of single particles were analyzed using the same CCSEM-EDX system mentioned earlier. The in-situ aerosol optical properties were measured with a nephelometer (scattering and back scattering coefficients) and an aethalometer (absorption coefficients). The aerosol optical depth was measured using a portable sunphotometer. The properties of the particle were used to estimate the top of the atmosphere radiative forcing of free tropospheric aerosols via the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model. In Figure 8 below, we compare two sampling periods (S5 and S7) for which we had available all the data necessary for the calculations. The plot shows the top of the atmosphere radiative forcing as a function of the cloud optical depth, assuming that clouds were underneath the aerosol layer (OMP lays typically above the stratocumulus clouds). The colored bands represent extreme radiative forcing values when varying the single scatter albedo, asymmetry parameter, and aerosol optical depth by the respective measurement uncertainties. As expected, the magnitude and sign of the forcing depends on the cloud optical depth. For sample S5, for the clear sky case (corresponding to a cloud optical thickness of zero), the aerosols have a large cooling effect of almost –15 Wm^{-2}; while for the optically thick cloud case, the aerosols have a warming effect that plateaus around 3 Wm^{-2}. For sample S7, the forcing has a narrower range around zero, with a slight cooling effect near –0.5 Wm^{-2} for clear sky or optically thin clouds, and a small warming effect that plateaus around 0.2 Wm^{-2} for optically thick clouds.

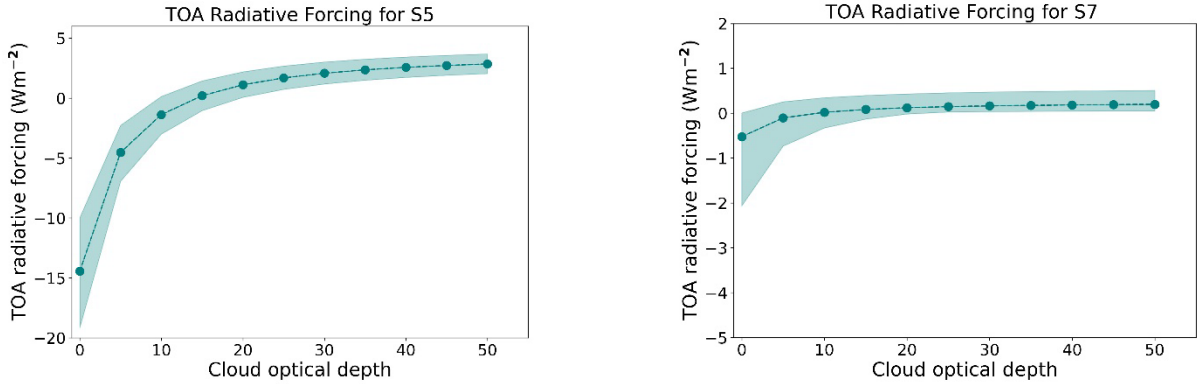

Figure 8. Top-of-the-atmosphere (TOA) radiative forcing of aerosols during the collection periods of two samples (S5, left and S7, right) calculated from SBDART as a function of the cloud optical depth simulating conditions going from clear sky to very optically thick cloud deck underneath the aerosol layer. Figure from Morgenstern et al. (in preparation).

Droplet Size Distributions are Locally Narrow in Stratocumulus Clouds

Marine stratocumulus clouds are the "global reflectors," sharply contrasting with the underlying dark ocean surface and exerting a net cooling on Earth's climate. The magnitude of this cooling remains uncertain, in part due to the averaged representation of microphysical processes such as the droplet-to-drizzle transition in global climate models (GCMs). Current GCMs parameterize cloud droplet size distributions as broad, cloud-averaged gammas. Using digital holographic measurements of discrete stratocumulus cloud volumes obtained during ACE-ENA, we find cloud drop size distributions to be narrower at the centimeter scale, never resembling the cloud average. These local distributions tend to form pockets of similar-looking cloud regions each characterized by a size distribution shape that is diluted to varying degrees. These observations open the way for novel modeling representations of microphysical processes.

The research used an algorithm developed as part of the research to search and classify characteristic size distributions within a cloud. The approach combines hypothesis testing, specifically, the Kolmogorov–Smirnov (KS) test, and a widely used class of machine learning algorithms for identifying clusters of samples with similar properties: density-based spatial clustering of applications with noise (DBSCAN) is used as the specific example for illustration. The two-sample KS test does not presume any specific distribution, is parameter free, and avoids biases from binning. Importantly, the number of clusters is not an input parameter of the DBSCAN-type algorithms but is independently determined in an unsupervised fashion. As implemented, it works on an abstract space from the KS test results, and hence spatial correlation is not required for a cluster. The method is explored using data obtained from the Holographic Detector for Clouds (Holodec) deployed during the Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) field campaign. The algorithm identifies evidence of the existence of clusters of nearly identical local size distributions. It is found that cloud segments have as few as one and as many as seven characteristic size distributions. To validate the algorithm's robustness, it is tested on a synthetic dataset and successfully identifies the predefined distributions at plausible noise levels. The algorithm is general and is expected to be useful in other applications, such as remote sensing of cloud and rain properties.

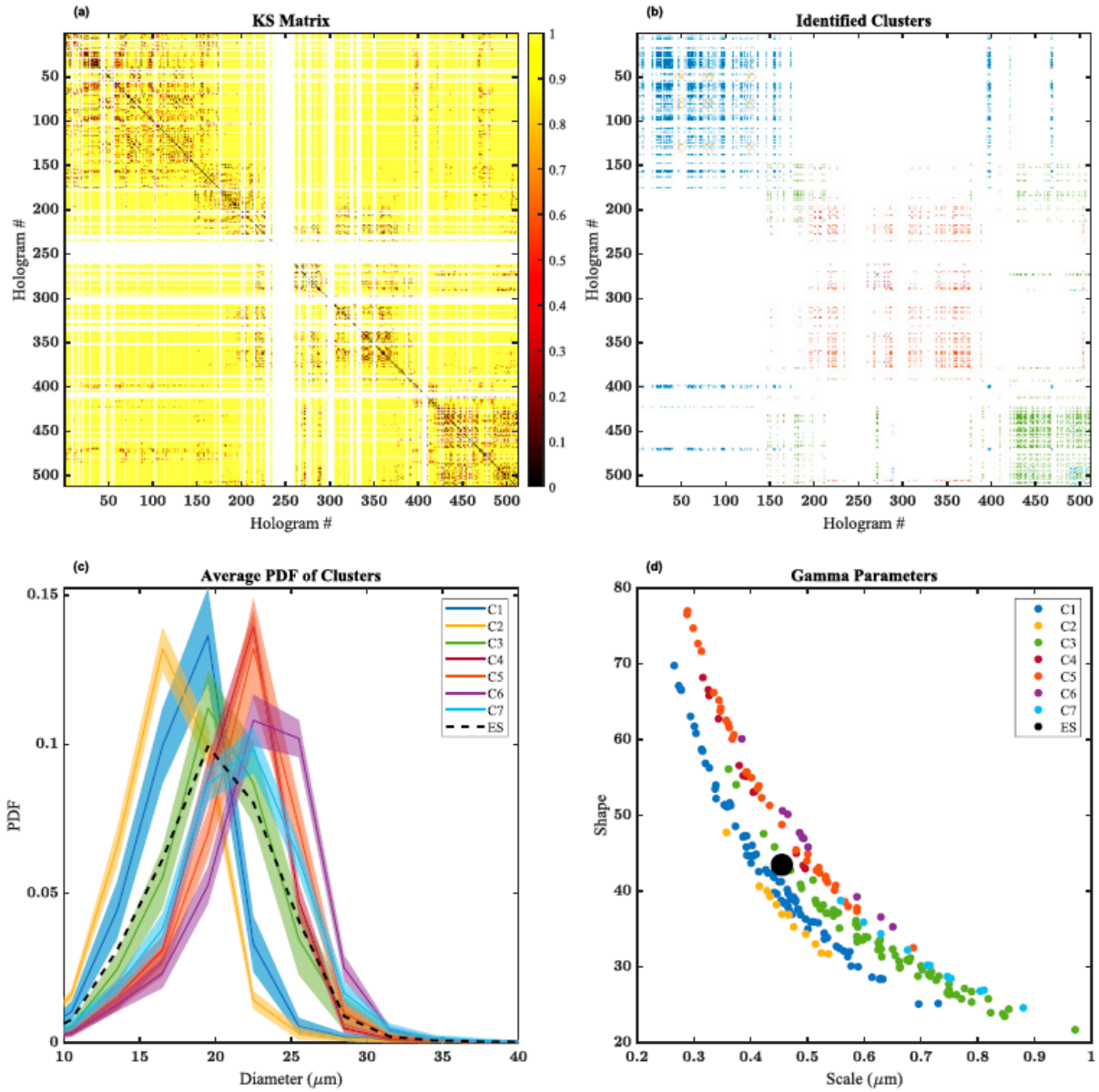

Figure 9. Results from a level flight through stratocumulus clouds during ACE-ENA: (a) KS matrix showing regions of similar size distribution type. (b) Clusters identified by the machine learning algorithm, depicted by different colors. (c) Average size distributions of the different clusters. The shaded portion represents 1 standard deviation. The dashed black line shows the PDF for the entire segment of holograms above the cutoff. (d) The fitted shape and scale parameters of the modified gamma distribution for the holograms in different clusters. The large black dot gives the shape and scale parameter for the entire segment. From Allwayin et al. (2022).

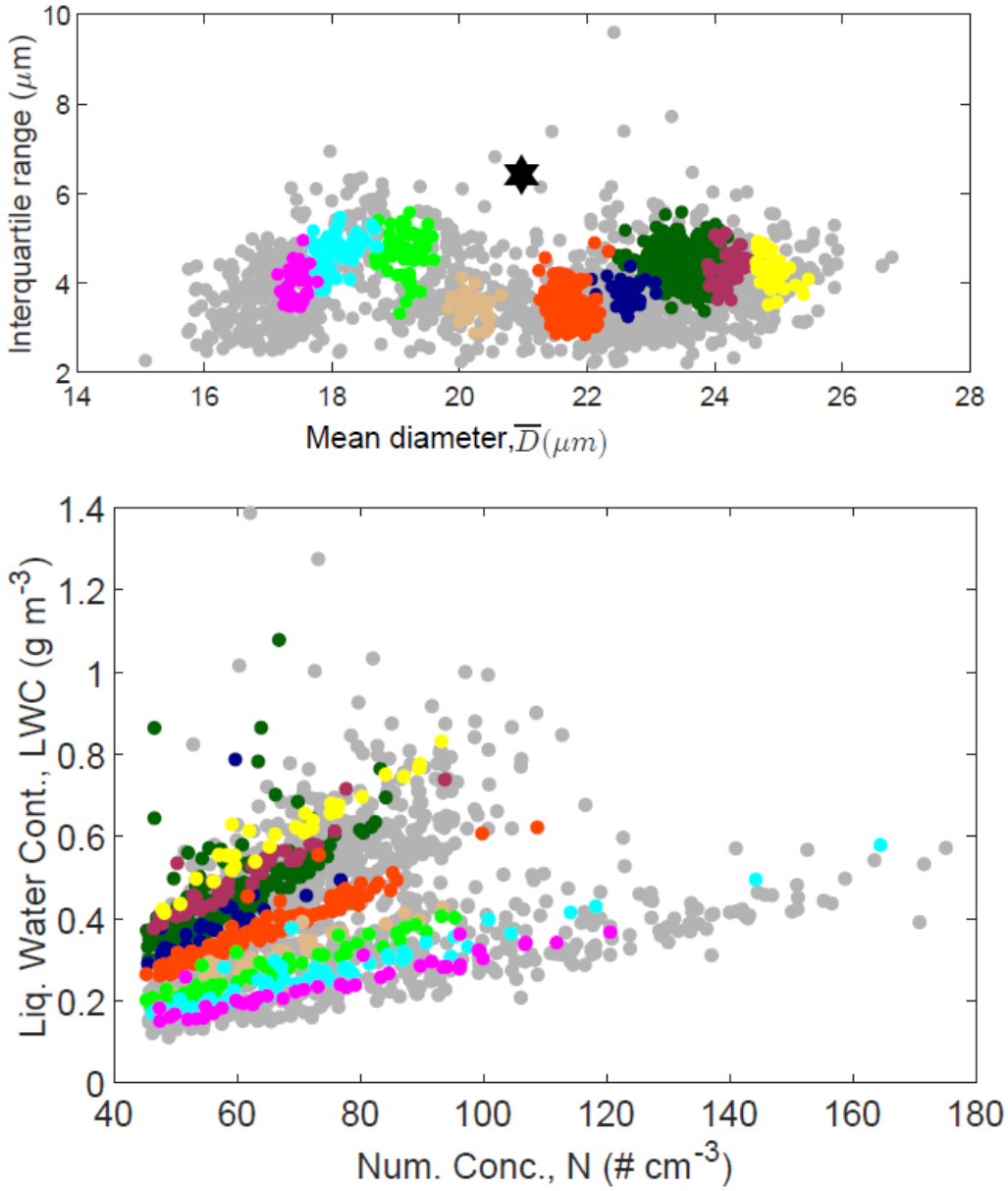

Figure 10. Top panel: A measure of the width of local distribution, the interquartile range, versus the mean diameter. Individual holograms are shown as circles, with clusters denoted by the colors. The black star denotes the segment average and the gray circles indicate the unclassified holograms. All holograms are from a 60-km flight segment at 850 m on July 18, 2017. Bottom panel: Cloud liquid water content versus cloud droplet concentration, suggesting that microphysical relationships for individual clusters of similar holograms can reflect cloud histories. Cloud liquid water content and number concentrations for different clusters have a linear relationship with slope depending on the cluster type. As the distribution shape and correspondingly the mean diameter remains constant for holograms of a cluster, the linear reduction in liquid water content with number concentration suggests a predominance of dilution and complete evaporation of droplets during the mixing process with dry environmental air (inhomogeneous mixing). From Allwayin et al. (2023).