Los Alamos NATIONAL LABORATORY

Aerosol sources, optics and mixed-phase chemistry in the coastal marine environment

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June 22, 2021

Managed by Triad National Security, LLC, for the U.S. Department of Energy's NNSA.
LA-UR-21-25845

Combustion Aerosol Sources in the Coastal and Marine Environment

- **Ship tracks impact radiation and cloud formation with uncertainties, but what about large urban centers?**
  - What is the role of combustion aerosols (include carbonaceous and absorbing aerosol) in the coastal marine environment?
  - Significant urban sources from NW LA/LB basin (~12–24 hrs)
  - What are the seasonally dominant aerosol processes?
- **Previous work: carbonaceous aerosol dominates mass**
  - Sources: Combustion > biomass burning > marine
  - SOA formation/Ozone chemistry (Aug-Sept)

		combustion	biomass burning	marine Origin
FTIR	Concentration ($\mu g\ m^{-3}$)	3.0 (62 %)	0.88 (18 %)	0.97 (20 %)
	O/C	0.46	0.48	1.04
	Composition			
AMS	Concentration ($\mu g\ m^{-3}$)	2.5 (61 %)	1.1 (26 %)	0.51 (13 %)
	(m/z 44)/OM	26 %	5 %	2.6 %
	(m/z 60)/OM	0.1 %	0.1 %	0.6 %
	(m/z 44)/(m/z 43)	7.5	0.6	1.5
	(m/z 44)/(m/z 57)	146	2.7	3.6

Ship tracks in the Pacific off the coast of N. America. NASA GOES WEST
Glassmeier et al., Science, 2021.

Concentrations and composition of PMF factors identified from FTIR and AMS measurements. Mass fractions in parentheses. *S. Liu et al., ACP 2011.*

Functional groups:

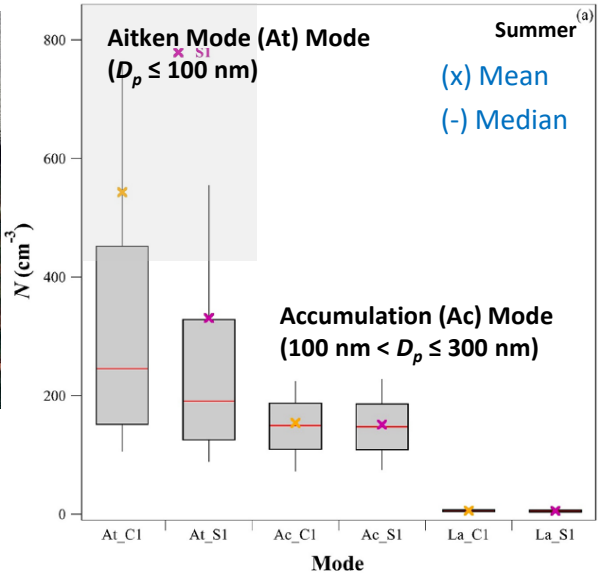
Alkane
Carboxylic Acid
Hydroxyl
Amine
Non-Acid Carbonyl

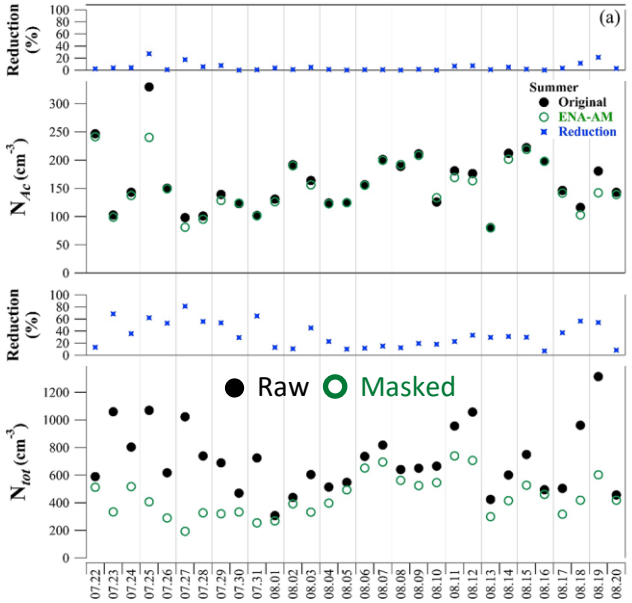
Aerosols in the Eastern North Atlantic (ENA)

Well-mixed in the summer (at the surface and in the boundary layer)

- Deployed a supplementary aerosol site (S1) during the ACE-ENA Intensive Operating Period
- Novel mathematical algorithm to identify local aerosol sources that largely impact Aitken diameter
- Improved ability for aerosol process studies within the marine boundary layer

Submicron Number Concentrations
AOS (C1) and Supplementary Site (S1)





Gallo, F. et al. Identifying a regional aerosol baseline in the Eastern North Atlantic using collocated measurements and a mathematical algorithm to mask high-submicron-number-concentration aerosol events. Atmospheric Chemistry and Physics, 2020. https://doi.org/10.5194/acp-20-7553-2020

Aerosols in the Eastern North Atlantic (ENA)

Multiday long-range transport events impact year-round CCN budgets

- 3 Aerosol regimes identified during 9 events in 2017 using a series of mathematical algorithms using Aerosol Observing System (AOS) chemical, physical and optical properties
- NOAA HYSPLIT back trajectories and NASA CALIPSO aerosol products validate the method
- Increased absorbing aerosol (Black Carbon) and Accumulation mode with decreased Single Scattering Albedo in comparison to regional background periods (SSA: 0.95-0.97)

	CALIPSO Classification	Event Dates	Median N_{Ac} (cm^{-3})	Mean SSA @ 529 nm	Mean BC ($ng\ m^{-3}$)	ΔN_{tot}	ΔN_{At}	ΔN_{Ac}
Cluster 1: Elevated Background	Dust and Marine	Mar 12 - 15 Nov 26 - 28 Dec 7 - 10	314 284 220	0.93 0.91 0.91	103 167 108	> +105%	> +75%	+150% - +200%
Cluster 2: Enhanced Accumulation	Continental Mixed Aerosol	Jan 7 - 11 Apr 20 - 22 May 21 - 22 Oct 11 - 13	370 434 607 544	0.87 0.94 0.92 0.89	247 111 161 199	> +90%	+20% to +75%	> +200%
Cluster 3: Enhanced CCN	Biomass Burning / Smoke	Aug 26 - 29 Sept 9 - 13	326 288	0.94 0.93	220 186	< +25%	< -25%	< +130%

Aerosol regime identification algorithms for the Aerosol Observing System (AOS)

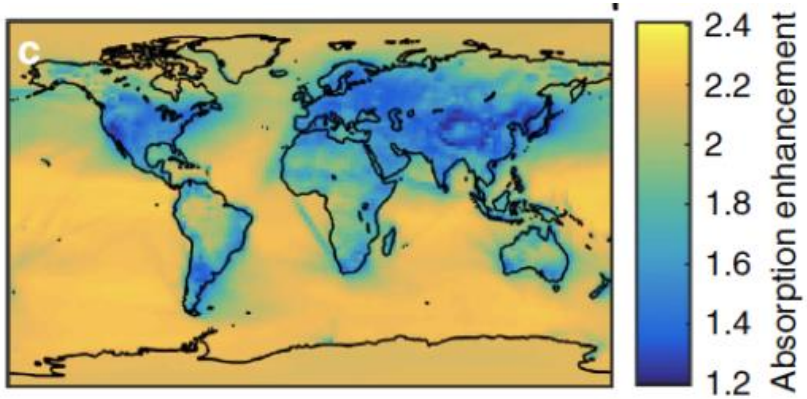
Aiken, Gallo et al.
Poster Session 5
Thurs at 2-3pm

LA-UR-21-25845

GALLO, F. ET AL. LONG-RANGE TRANSPORT AEROSOL IN THE EASTERN NORTH ATLANTIC: THREE MULTIDAY EVENT REGIMES INFLUENCE CLOUD CONDENSATION NUCLEI. IN PREP FOR ATMOSPHERIC CHEMISTRY AND PHYSICS, 2021.

Are large modeled black carbon absorption enhancements above oceans accurate?

- **EPCAPE's Coastal environment with ship, large urban center (LA/LB) and fire emissions are ideal to test this.**



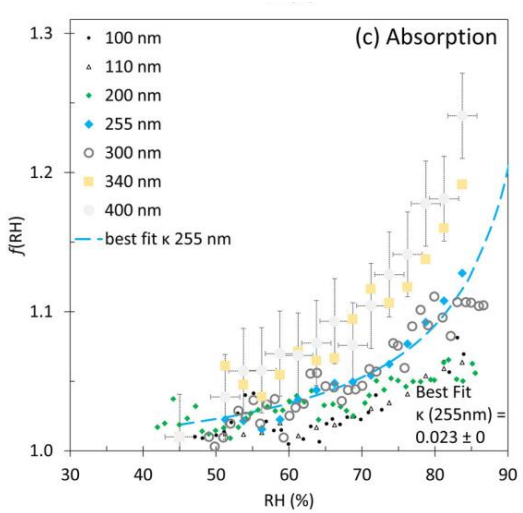
Simulated BC absorption enhancement by humidity GISS model. Fierce et al., Nature Communications, 2016.

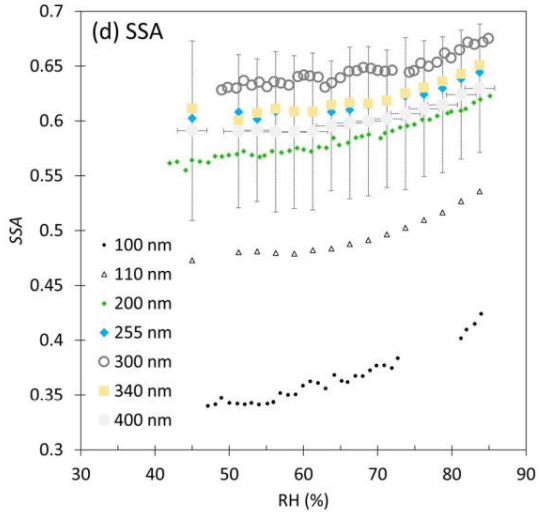
Science Questions

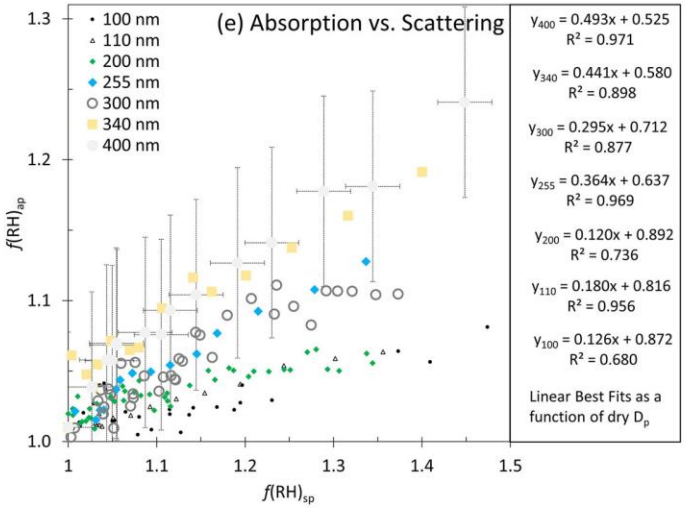
- How do marine inorganic (NaCl) and organic (DMS) sources affect optical properties of absorbing aerosol from combustion? What is their mixing state with absorbing species, e.g. Black and Brown Carbon?
- Does the marine environment enhance carbonaceous aerosols' ability to uptake water and affect cloud formation?
- What are the affects of aging and mixing on optics, chemistry and CCN?

SQ1: How do marine inorganic (NaCl) and organic (DMS) sources affect optical properties of absorbing aerosol?

- Submicron aerosol scattering and extinction vs humidity
  - SSA reduction & absorption enhancement of $(NH_4)_2SO_4$-nigrosin mixture with RH
  - Empirical absorption versus scattering enhancement function for use in models





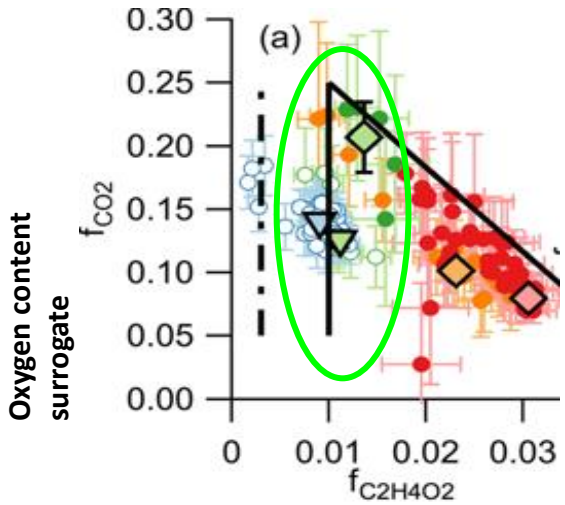


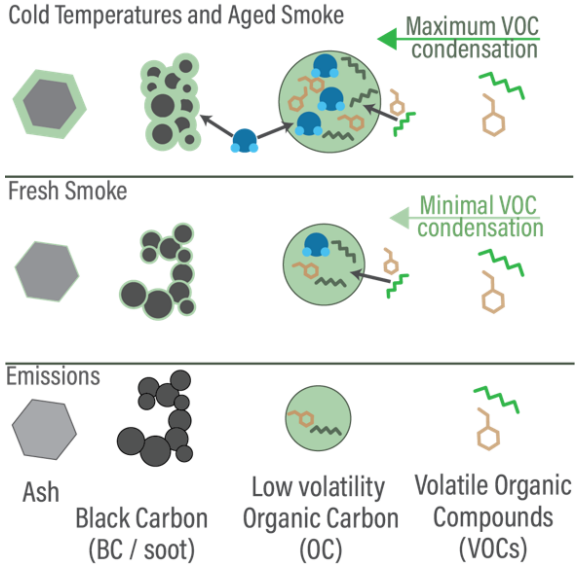
Carrico, C. M., et al. Relative Humidity-Controlled Single Scattering Albedometer (H-CAPS-PMSSA): Design, Data-Analysis and Validation. *Aerosol Science & Technology*, 2021. https://doi.org/10.1080/02786826.2021.1895430

SQ2: How does marine aerosol internally mix with Black and Brown Carbon?

- Wildfire absorbing aerosol chemistry
- Woodbury, AZ wildfire, aged <1 day
- Highest Oxygen content at plume edges
- Thick BC coatings enhance absorption

Soot surface chemistry and aging in biomass burning
Secondary organic aerosol (SOA) condensation and growth





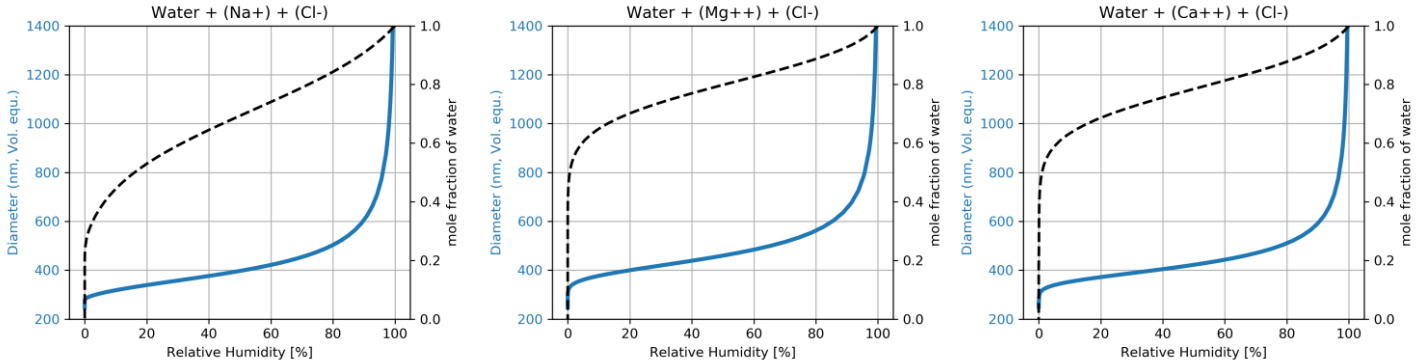
LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT

Los Alamos NATIONAL LABORATORY

J. E. Lee, M. K. Dubey, A. C. Aiken, P. Chylek and C. M. Carrico, "Optical and chemical analysis of absorption enhancement by mixed carbonaceous aerosols in the 2019 Woodbury, AZ fire plume." *Journal of Geophysical Research, Atmosphere* https://doi.org/10.1029/2020JD032399

SQ3: Does the marine environment enhance carbonaceous aerosol's ability to uptake water and affect cloud formation?

- Simulated water uptake of 250 nm dry diameter sea salt mixtures

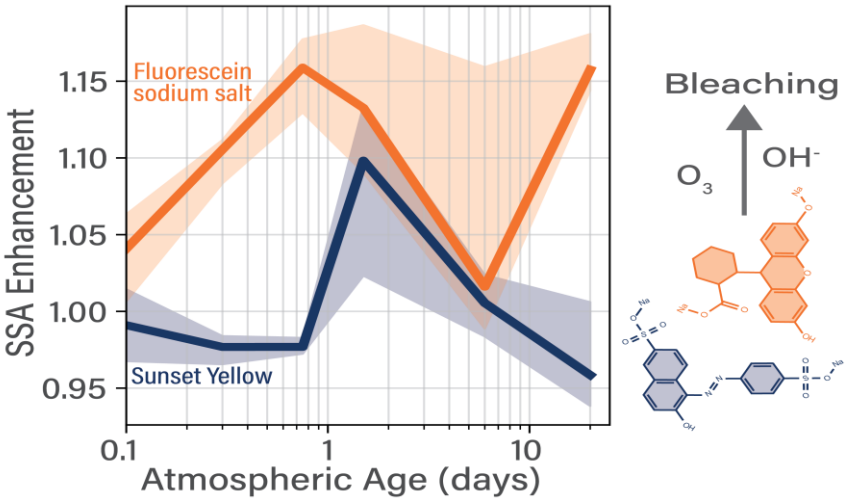


mixture	Growth Factor at 85% RH
$MgCl_2$	2.42
$CaCl_2$	2.36
NaCl	2.37
$(NH_4)_2SO_4$	2.22

- Hygroscopic growth of BC mixtures are simulated and will be compared with laboratory experimental results using the H-CAPS-PM_{SSA}

SQ3: What are the affects of aging and mixing on optics, chemistry and CCN?

- Preliminary studies with an oxidation flow reactor of the brown carbon mimics.
- Both mimics have a peak at ~1 day of OH equivalent aging.
- Evidence of darkening and bleaching for both with diverging responses for longer exposure periods.
  - Further study with repeated experiments and online chemical analysis using aerosol mass spectrometry.
  - Gorkowski et al., in prep, 2021.



Dubey, Gorkowski et al.
Poster Session 1, Tues at 2-3pm

Update: LANL-Guest-AOS Container

- **Deployment to TRACER in 2022 for TRACER-Carbonaceous Aerosol Thrust**
  - Housing for 2 SP-AMS's and humidified CAPS systems with UC-Davis
  - Rack-mountable and open floor space
- **Currently being reconfigured**
  - Replicate of New generation AOS (Uin et al., 2019)
  - Shared aerosol inlet (1000 LPM turbulent flow) and separate trace gas inlet at ~10 m a.g.l.
  - Flexible rack and instrument layout
  - HVAC controlled

ASR Atmospheric System Research

LA-UR-21-25845

Center for Aerosol-gas Forensics Experiments CAFÉ

- **3 Staff:** (Dubey, Aiken, Benedict)
- **2-3 Postdocs:** (Gallo*, Gorkowski, Lee)
- **2-3 Undergraduate/Graduate Students**

ASR Atmospheric System Research

ARM

LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT

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