Multi-campaign analysis of smoke properties and cloud interactions in the Southeast Atlantic using ORACLES, LASIC, and CLARIFY data with WRF-CAM5

Calvin Howes[1], Pablo Saide[1], Sharon Burton[2], Hugh Coe[3], Amie Dobracki[4], Marta Fenn[5], Richard Ferrare[2], Steffen Freitag[4], Johnathan Hair[2], Steven Howell[4], Siddhant Gupta[6], Uin Janek[7], Mary Kacarab[8], Chongai Kuang[7], L. Ruby Leung[9], Athanasios Nenes[10], Arthur Sedlacek[11], Kenneth Thornhill[2], Jenny Wong[12], Robert Wood[13], Huihui Wu[14], Yang Zhang[15], and Paquita Zuidema[16]

[1]University of California Los Angeles
[2]NASA Langley Research Center
[3]Department of Earth and Environmental Sciences
[4]University of Hawaii at Manoa
[5]SSAI
[6]University of Illinois at Urbana Champaign
[7]Brookhaven National Laboratory
[8]University of California Riverside
[9]Pacific Northwest National Laboratory
[10]Ecole Polytechnique Federale de Lausanne
[11]Brookhaven National Lab
[12]Mount Allison University
[13]University of Washington
[14]University of Manchester
[15]Northeastern University
[16]University of Miami

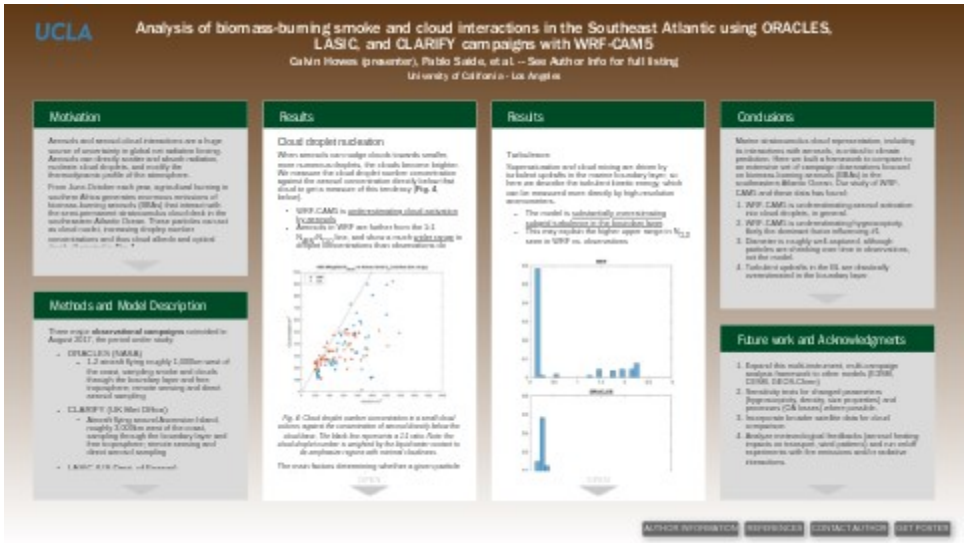
November 22, 2022

Abstract

The southeast Atlantic Ocean provides an excellent natural laboratory to study smoke-cloud interactions, a large driver of uncertainty in climate projections. The value of studying this in particular region is largely attributable to two factors—the expansive, bright, semi-permanent stratocumulus cloud deck and the fact that southern Africa is the largest source of biomass-burning aerosols in the world. We study this region using the WRF-Chem model with CAM5 aerosols and in situ observations from the ORACLES, LASIC, and CLARIFY field campaigns, all of which overlapped in August 2017. Across these campaigns, we compare aerosol, cloud, and thermodynamic variables to quantify model performance and expand upon observational findings of aerosol-cloud effects. Specifically, our approach is to analyze aerosol and cloud properties along flight tracks, picking out uniform legs within tropospheric smoke plumes and in the boundary layer. This unique approach allows us to sample the high spatiotemporal variability that can get lost to large-scale averaging. It also allows process-level comparison of local cloud responses to aerosol conditions, and measure model performance in those same processes. Along with better quantifying model predictive power, we find and justify updates to model parameters and processes to better emulate observations, notably aerosol

size parameters. Preliminary results suggest that WRF-CAM5 is activating a smaller percentage of aerosols into cloud droplets than shown in observations, which could lead to biased modeling of aerosol indirect radiative effects on a larger scale. We explore this effect further with CCN activation tendency, updraft, particle sizing, and composition analysis, as well as broader dynamics like entrainment and removal rates. Comparing the model with similar instrument suites across multiple colocated campaigns also allows us to quantify instrument uncertainty in ways that a focus on a single campaign cannot and gives further context to the model performance.

Analysis of biomass-burning smoke and cloud interactions in the Southeast Atlantic using ORACLES, LASIC, and CLARIFY campaigns with WRF-CAM5



Calvin Howes (presenter), Pablo Saide, et al. – See Author Info for full listing

University of California - Los Angeles

PRESENTED AT:

AGU FALL MEETING
New Orleans, LA & Online Everywhere
13–17 December 2021
Poster Gallery brought to you by WILEY

MOTIVATION

Aerosols and aerosol-cloud interactions are a huge source of uncertainty in global net radiative forcing. Aerosols can directly scatter and absorb radiation, nucleate cloud droplets, and modify the thermodynamic profile of the atmosphere.

From June-October each year, agricultural burning in southern Africa generates enormous emissions of biomass-burning aerosols (BBAs) that interact with the semi-permanent stratocumulus cloud deck in the southeastern Atlantic Ocean. These particles can act as cloud nuclei, increasing dropley number concentrations and thus cloud albedo and optical depth, illustrated in ***Fig. 1***.

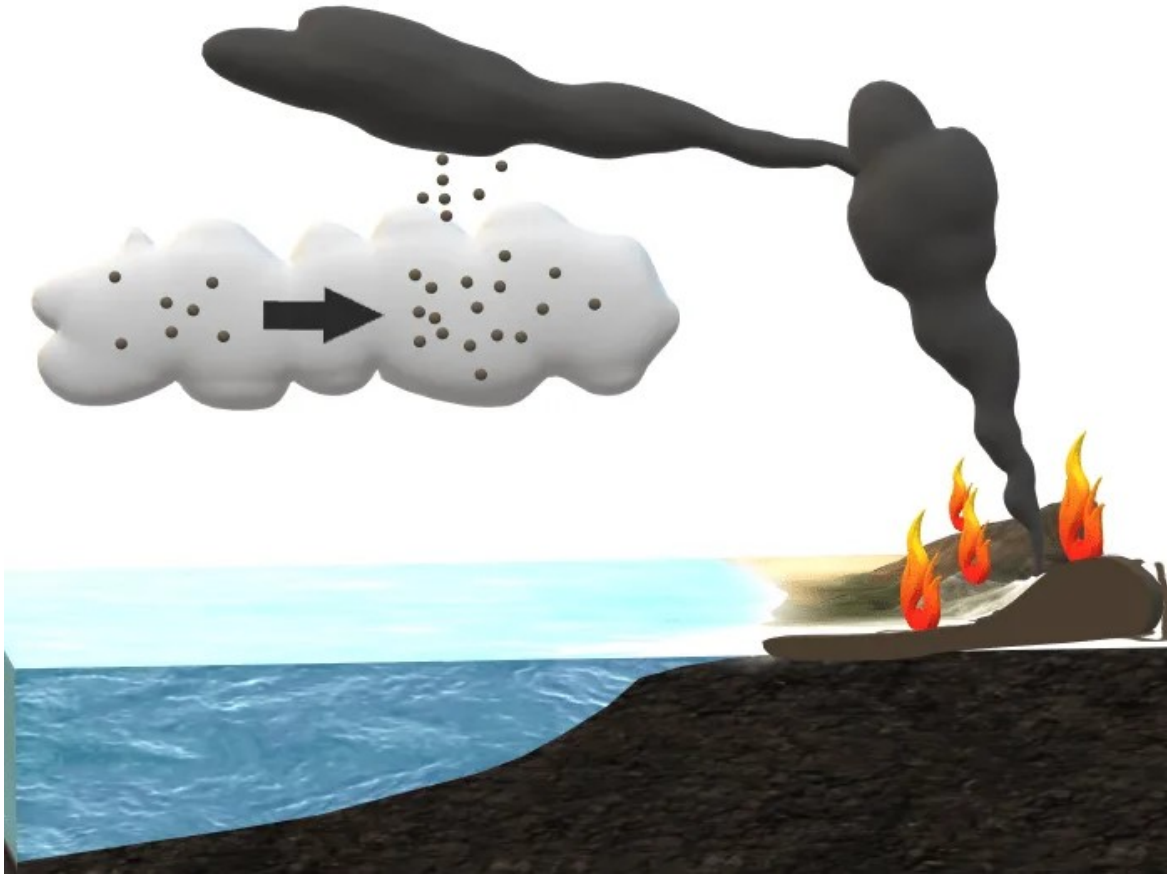


Fig. 1: Smoke from continental fires in southern Africa advects West, out over the marine stratocumulus cloud deck.

This leads to a substantial perturbation to the regional energy balance (Zuidema et al., 2016).

Models implement a very wide range of aerosol properties and processes, especially involving BBAs. The varied explicit and implicit assumptions about BBAs and their interactions are partially a result of a lack of *in situ* observations used to directly update models.

It is especially important for the study of aerosol-cloud impacts that models accurately represent particle size, number, hygroscopicity, chemical composition, aging, and cloud activation tendencies.

The existing multi-model intercomparison studies often focus on broad trends in a few parameters of interest across a large spatial area, such as cloud thickness or total aerosol optical depth (AOD), without closely examining the range of processes that underlie those differences (Shinozuka et al., 2020).

METHODS AND MODEL DESCRIPTION

Three major **observational campaigns** coincided in August 2017, the period under study:

- ORACLES (NASA)
  - 1-2 aircraft flying roughly 1,000km west of the coast, sampling smoke and clouds through the boundary layer and free troposphere; remote sensing and direct aerosol sampling
- CLARIFY (UK Met Office)
  - Aircraft flying around Ascension Island, roughly 3,000km west of the coast, sampling through the boundary layer and free troposphere; remote sensing and direct aerosol sampling
- LASIC (US Dept. of Energy)
  - Atmospheric Radiation Measurement (ARM) station on Ascension Island, 3km from coast; large remote sensing and direct sampling suite in the boundary layer, with radiosonde balloons.

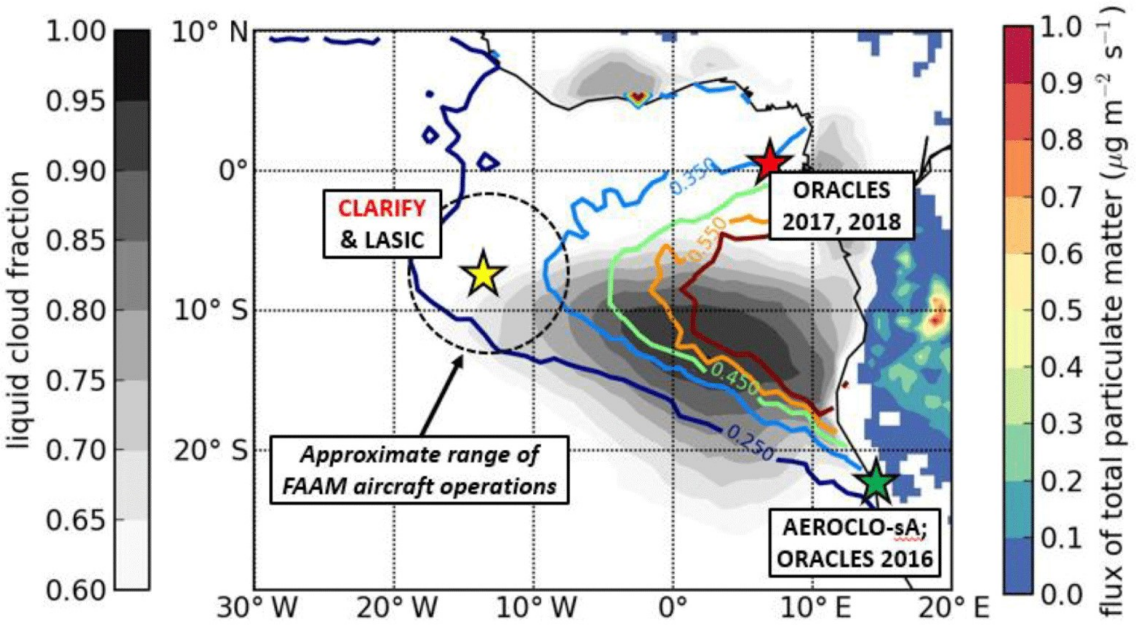


Fig. 2: Observation campaigns in southeastern Atlantic 2016-2018. Note the overlap of high cloud fraction and high PM flux. [Haywood et al., 2021]

Model configuration

- WRF-Chem with CAM5 aerosol and physics:
  - Intermediate resolution (36km) between purely regional and global scale models
  - 5hPa vertical layers (50 layers below 3km)
  - QFED2 Smoke emissions, NCEP FNL meteorology, MOZART trace gases

The model was restarted every 5 days from NCEP FNL with 2 days of spin-up for each initialization, aerosol initial conditions carried over from previous cycle. This **allows aerosol-meteorology feedbacks** to show up (e.g., aerosol heating causing transport and cloud changes).

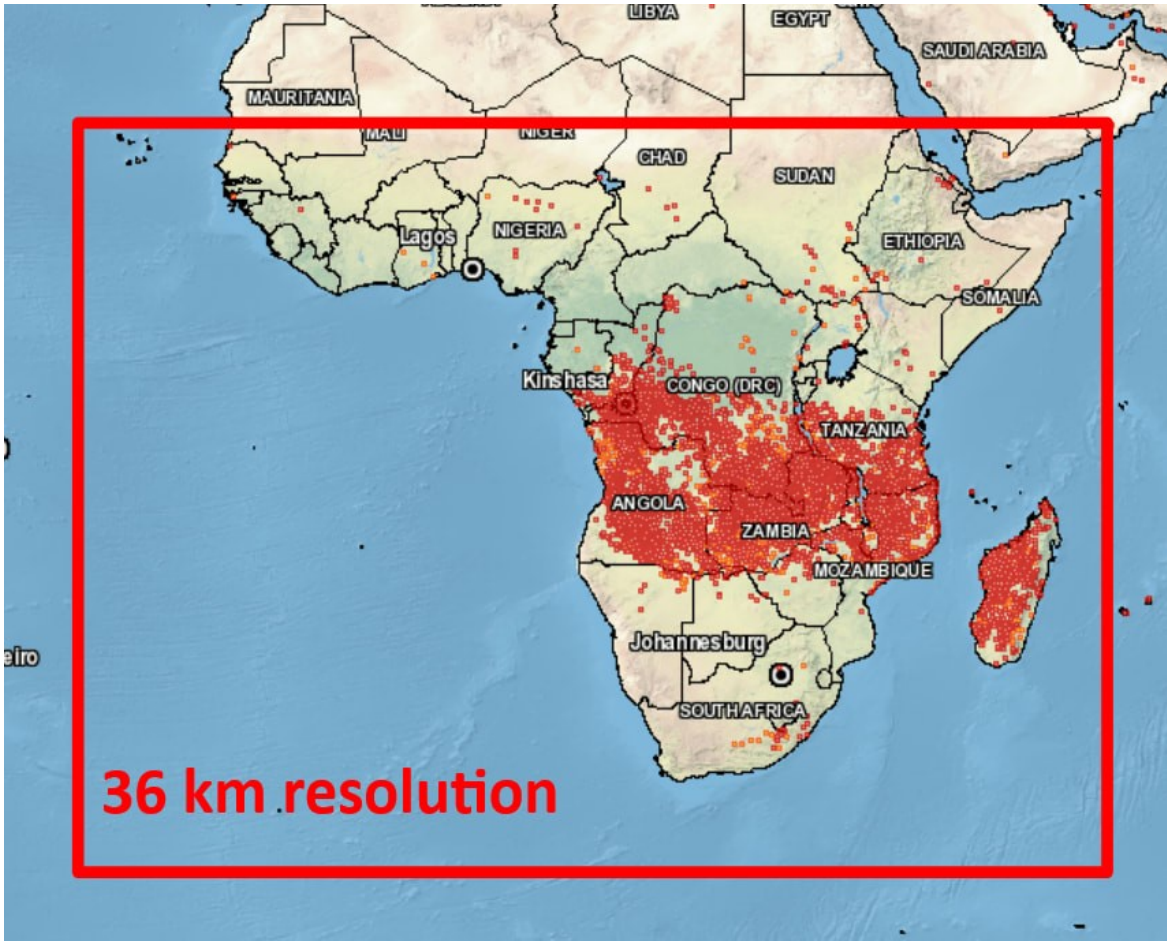


Fig. 3: WRF-CAM5 simulation region. Red dots are fire detections across the burning season.

RESULTS

Cloud droplet nucleation

When aerosols can nudge clouds towards smaller, more numerous droplets, the clouds become brighter. We measure the cloud droplet number concentration against the aerosol concentration directly below that cloud to get a measure of this tendency (**Fig. 4**, below).

- WRF-CAM5 is underestimating cloud activation by aerosols
- Aerosols in WRF are further from the 1:1 N_{AER}:N_{CLD} line, and show a much wider range in droplet concentrations than observations do

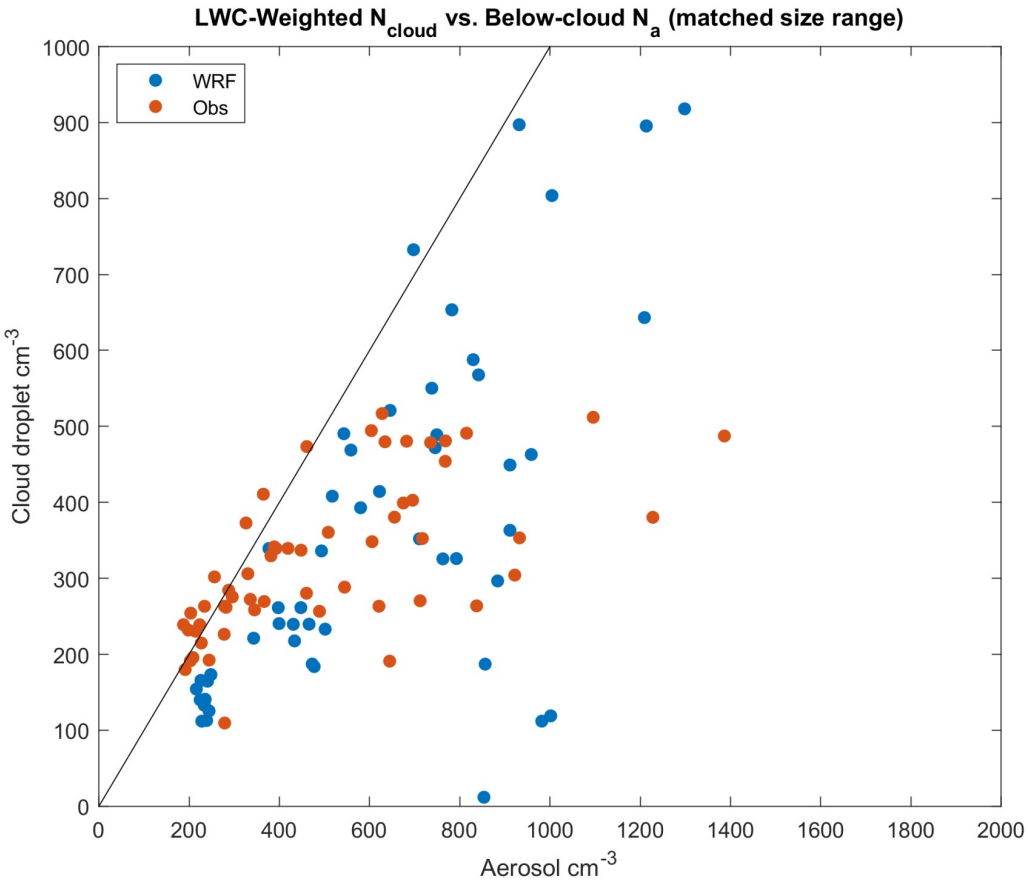


Fig. 4: Cloud droplet number concentration in a small cloud column, against the concentration of aerosol directly below the cloud base. The black line represents a 1:1 ratio. Note: the cloud droplet number is weighted by the liquid water content to de-emphasize regions with minimal cloudiness.

The main factors determining whether a given particle is able to activate are its diameter, its single-parameter hygroscopicity, and the ambient supersaturation (using turbulent kinetic energy as a measurable proxy).

Smoke Mean Diameter

- WRF-CAM5 is roughly accurate in estimating FT smoke diameter, though biased somewhat high.

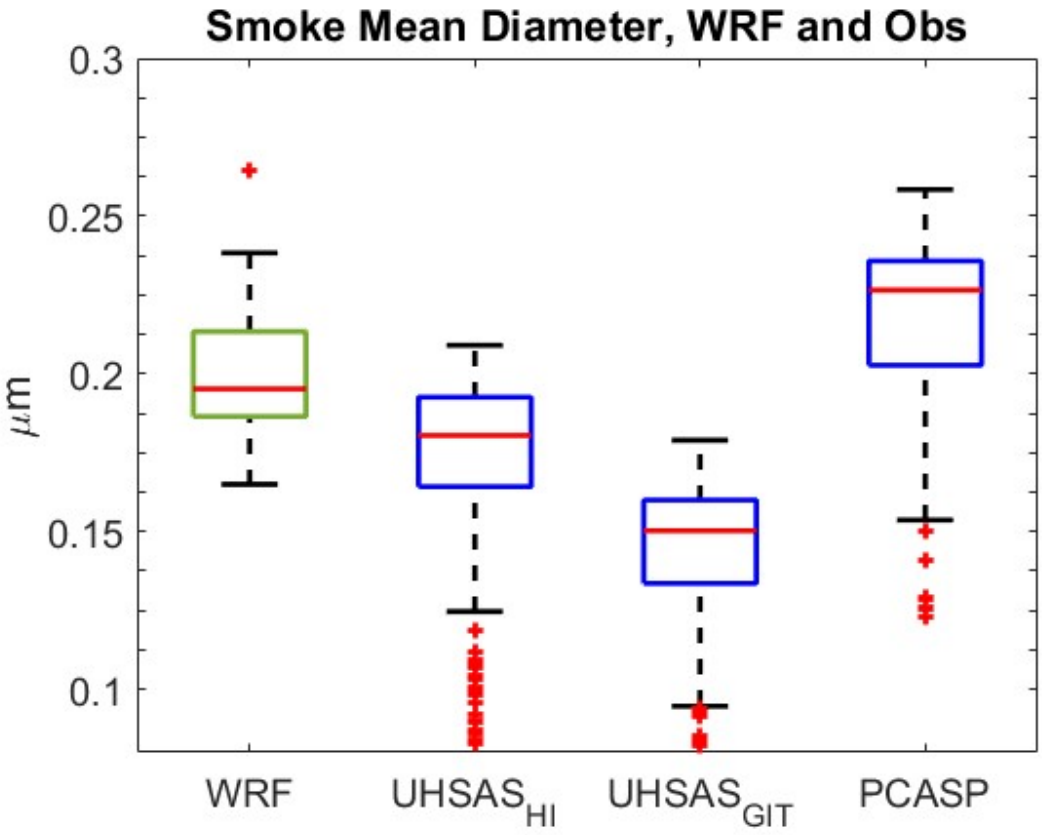


Fig. 5: Mean diameter of free-troposphereic smoke from the U. of Hawaii UHSAS, Georgia Inst. of Technology UHSAS, and U. North Dakota PCASP instruments. Red lines are medians, box edges are 25th/75th percentile, and whiskers are range, excluding outliers.

Larger particles will activate into cloud droplets more easily than smaller ones. Here, the U. Hawaii UHSAS is the most reliable report of geometric mean diameter as it has been corrected the most extensively. WRF is thus slightly overestimating average diameter, but significantly overlaps with UHSAS mean and range. The variability between instruments is possibly due to calibration differences or anisokinetic inlet setup.

- Observations in the BL show a persistent coarse mode and nucleation mode at low and medium smoke loading
- The lower free troposphere, which is often smoky, shows no major coarse mode
  - CN counters show there is no major nucleation mode in the FT

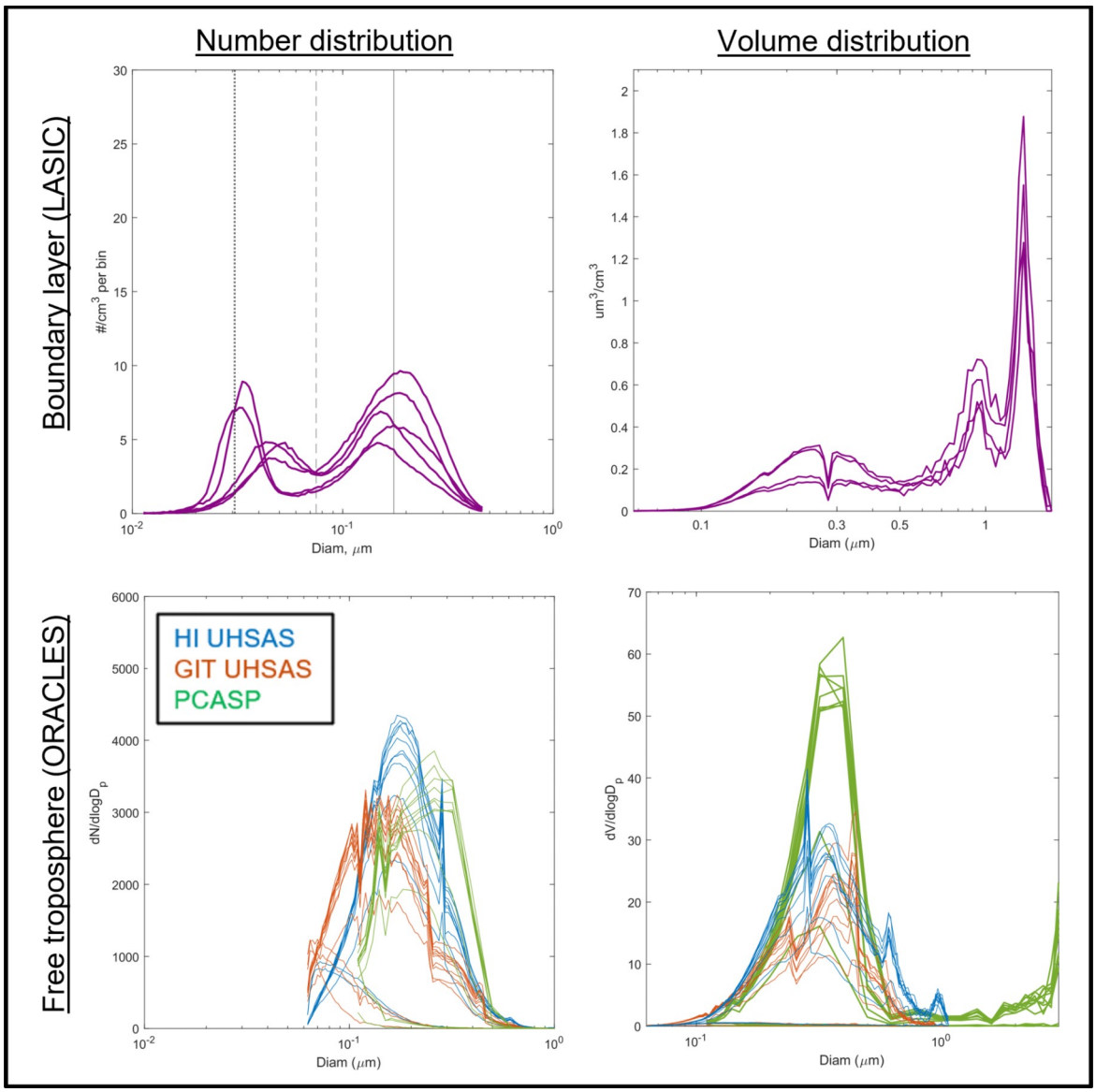


Fig. 6: Number and volume distributions in the BL, top (from LASIC) and FT, bottom (from ORACLES).

Hygroscopicity

- WRF-CAM5 underestimates hygroscopicity overall, which is likely leading to the cloud underactivation tendency above.
- WRF also shows a much narrower range of hygroscopicity than CCN estimates.

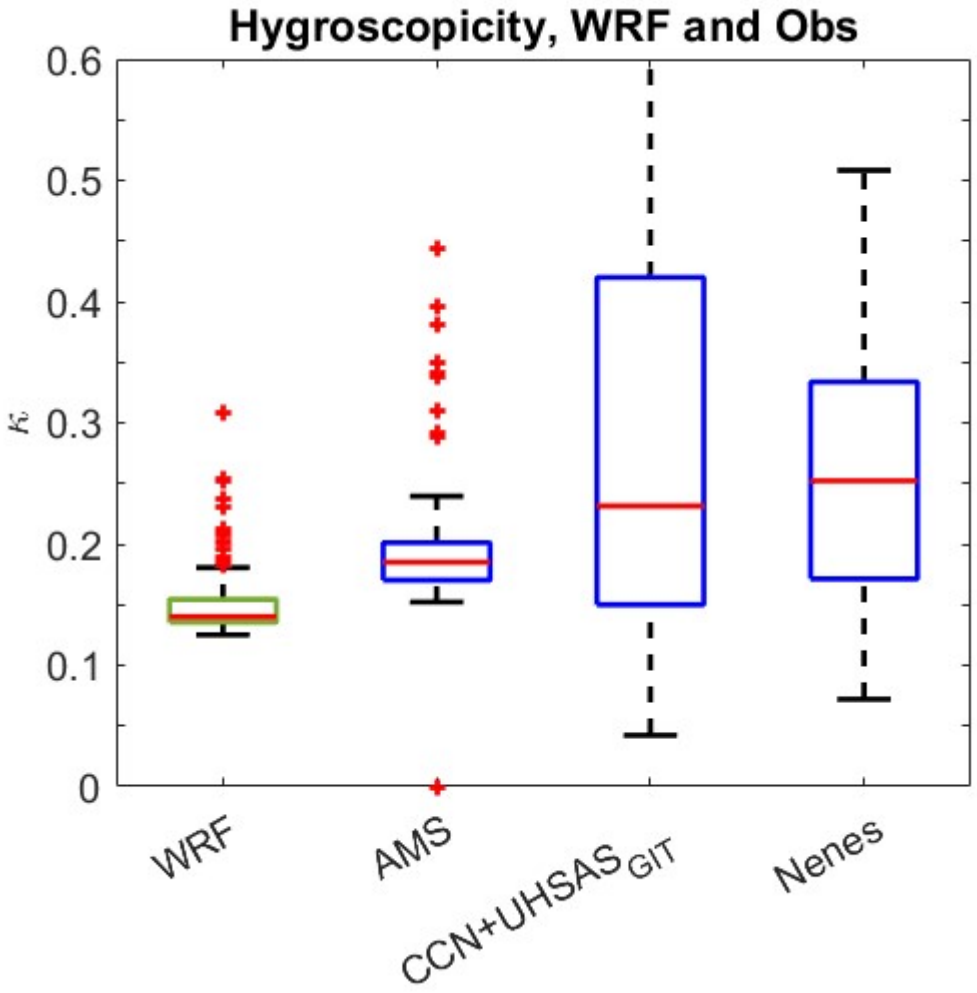


Fig. 7: Hygroscopicity from WRF, AMS+SP2 volume average, recalculated CCN+UHSAS$_{GIT}$ observations, and latest reported values form the Nenes (GIT) team.

The single hygroscopicity parameter is calculated by either a weighted volume average, as done with either the AMS and SP2 or in WRF, or from integrating the size distribution with the UHSAS and CCN counter and using a lookup table [Petters & Kreidenweiss, 2008].

WRF-CAM5 in this configuration also has a lower fixed hygroscopicity and lower density than suggested by experimental literature, and both of these are also likely to evolve as the particles age and oxidize over the course of several days [Dinar et al., 2006; Kuwata et al., 2011]. This is an under-studied topic for long-range biomass-burning smoke transport.

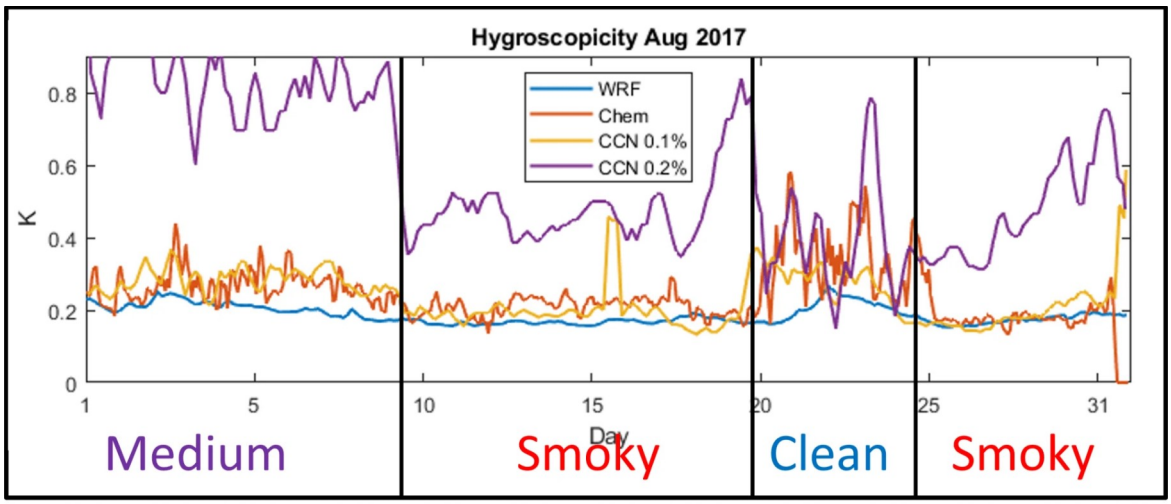


Fig. 8: Time series of hygroscopicity measured at Ascension Island (LASIC) in August 2017. Days are divided by smoke level, as clean, medium, or high (smoky).

- BL hygroscopicity in WRF has less variability than observations
- BL hygroscopicity may largely depend on mixing between modes, as there are significant coarse and nucleation modes.
- Between 0.1% and 0.2% supersaturation, K changes drastically, indicating different composition between 50-200nm. Within the accumulation mode, particles are not uniformly, internally mixed. This is an ongoing research topic.

Chemical composition

- WRF-CAM5 estimates an accurate composition in the smoky free troposphere, especially the volume fraction of OA and sulfate (**Fig. 9**, below). FT smoke is well-approximated by the accumulation mode alone.
- The model underestimates SO_4 in the boundary layer
- Note: This build of WRF-Chem has not implemented aerosol-form NH_4 and NO_3, although it is possible.

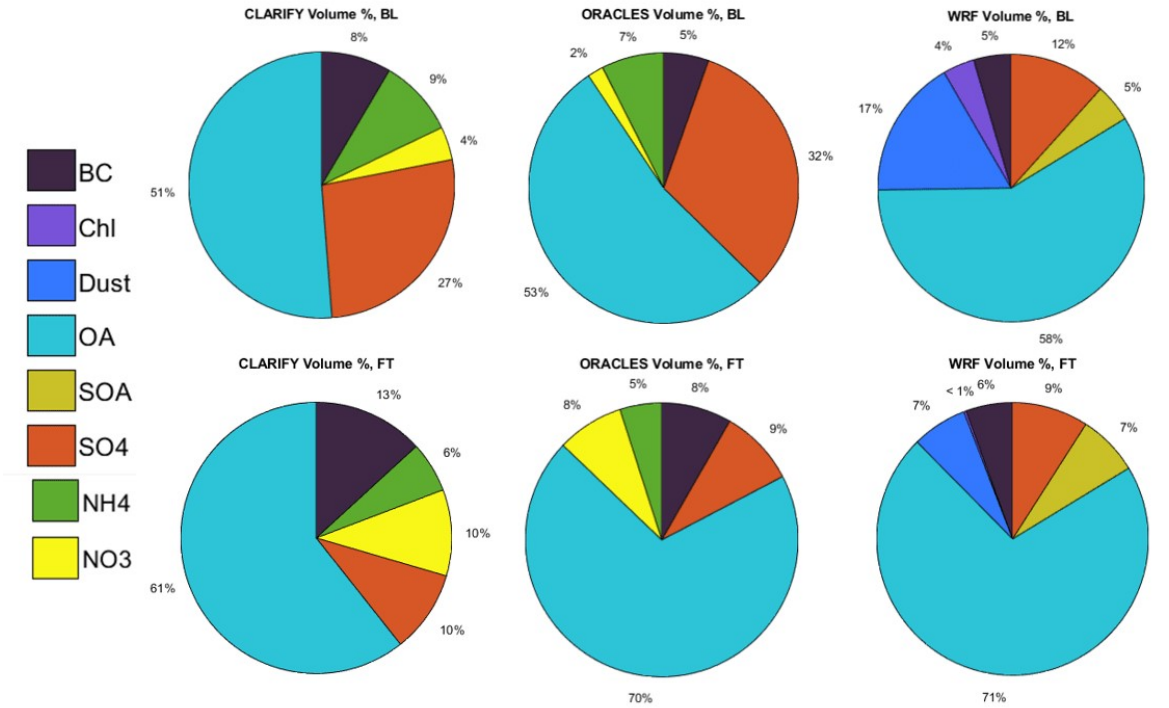


Fig. 9: Chemical composition from CLARIFY AMS (left), ORACLES AMS (center) and WRF-CAM5 (right). Volume averages across boundary layer flight legs (top) and the smoky lower free troposphere (bottom).

Chemical composition measurements from LASIC at Ascension Island show again that WRF is consistently underestimating the sulfate fraction in the boundary layer (Fig. 10, below).

- The relative enhancement of BL SO_4 during clean periods indicates that the background aerosol mass below 1μm is sulfate-dominated in the MBL.

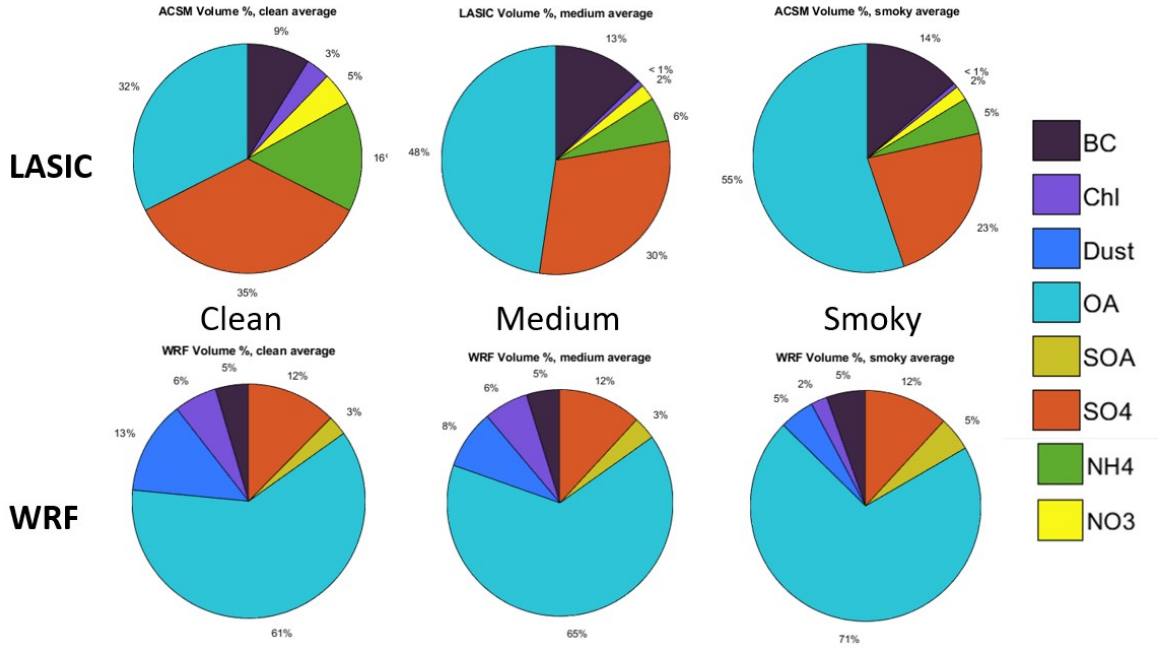


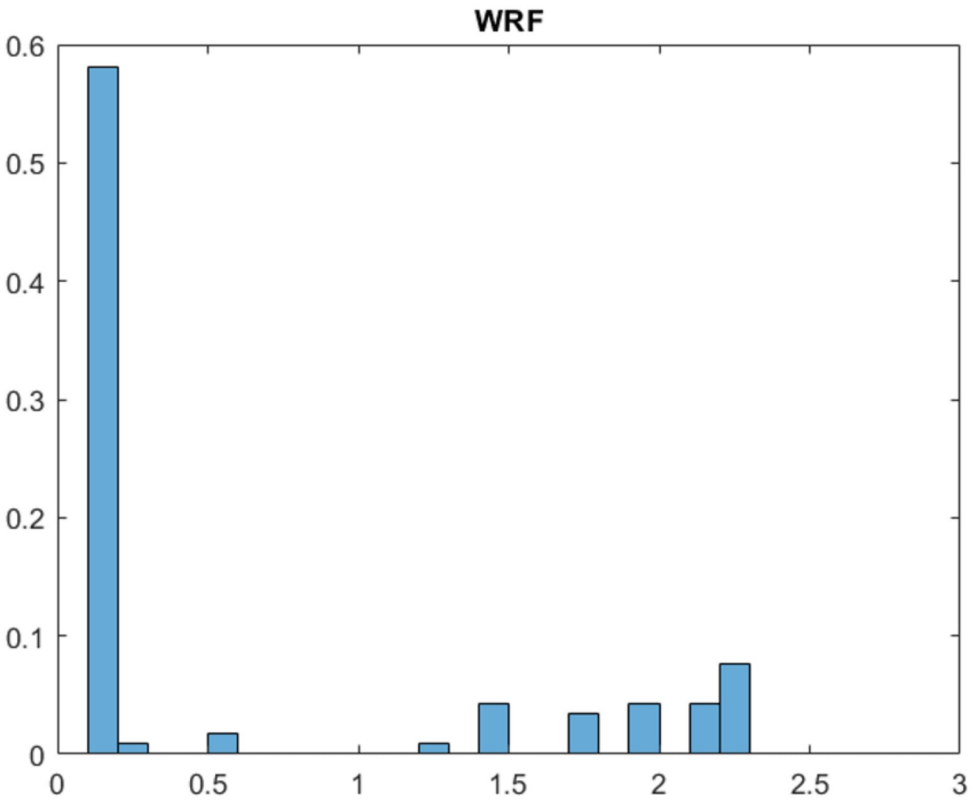
Fig. 10: Chemical composition at Ascension Island during smoky, medium-smoky, and clean periods in August 2017. Observations from the ARM station ACSM.

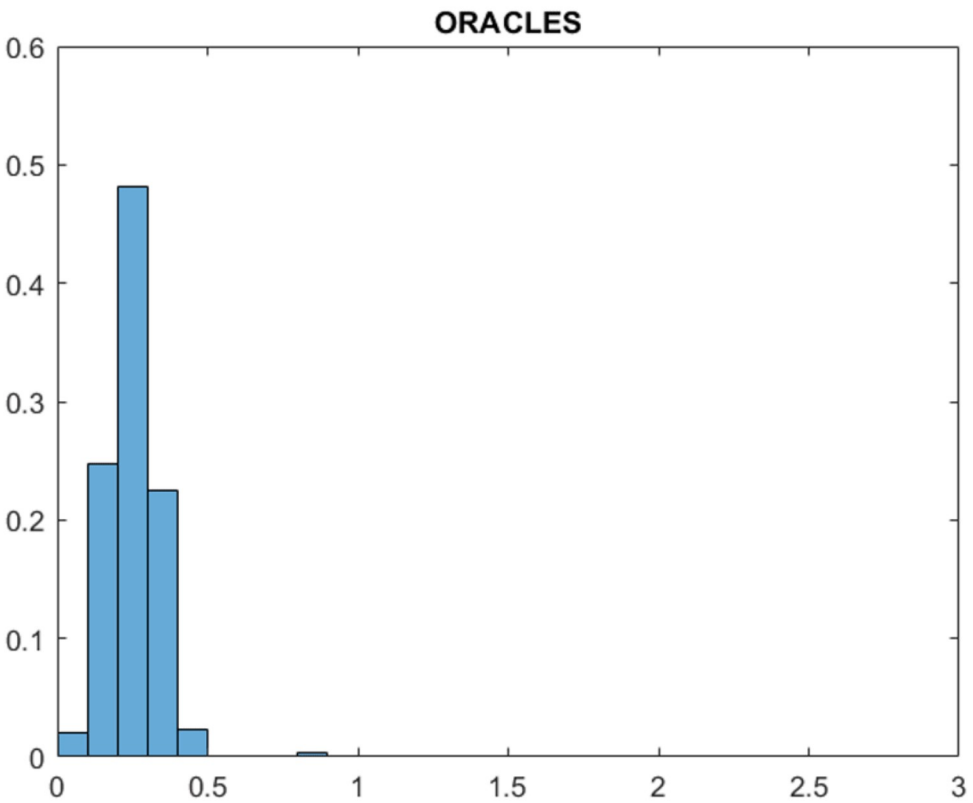
RESULTS

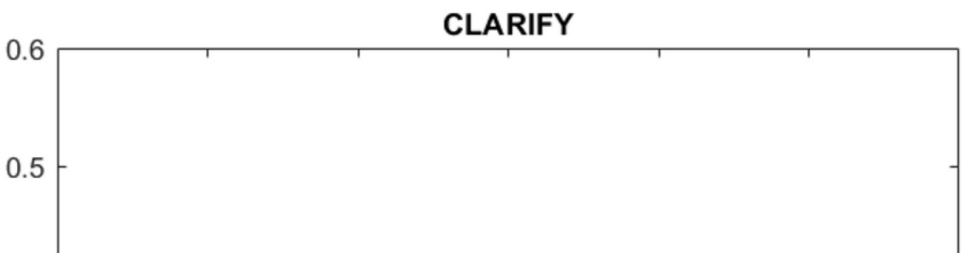
Turbulence

Supersaturation and cloud mixing are driven by turbulent updrafts in the marine boundary layer, so here we describe the turbulent kinetic energy, which can be measured more directly by high-resolution anemometers.

- The model is substantially overestimating subgrid turbulence in the boundary layer.
- This may explain the higher upper range in N_{CLD} seen in WRF vs. observations


WRF
0.6
0.5
0.4
0.3
0.2
0.1
0
0
0.5
1
1.5
2
2.5
3


ORACLES
0.6
0.5
0.4
0.3
0.2
0.1
0
0
0.5
1
1.5
2
2.5
3


CLARIFY
0.6
0.5

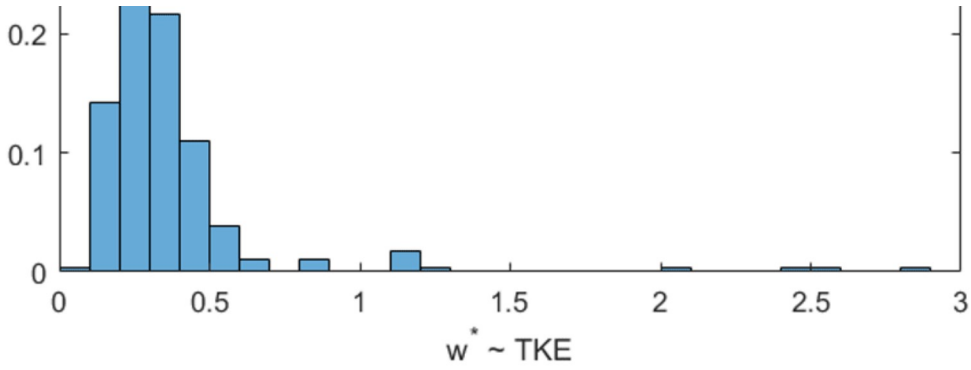


Fig. 11: Turbulent kinetic energy in the boundary layer between observations from aircraft aenemometers (top, middle) and WRF (bottom). Note the higher total scale of WRF.

Turbulent updrafts allow air parcels to cool and supersaturate with water vapor, and are a key to cloud activation. Cloud activation efficiency is low, despite this overestimation, thus this is likely not the dominant factor. The 'characteristic updraft' w* is derived from the standard deviation of the vertical wind distribution over short periods, sampled by anemometers on the campaign aircraft [Morales & Nenes, 2010].

Aging trends

Smoke particles can evolve over time in diameter, composition, optical properties, hygroscopicity, and more. Many models have limited ability to capture these trends.

- WRF has aerosols mostly growing with time, while observations show shrinkage (**Fig. 12)**
- OA is decreasing relative to SO_4 significantly with age. WRF does not capture this relative loss of OA (**Fig. 13)**

This divergence between WRF-CAM5 and observations could explain the population differences in diameter and the relatively static WRF hygroscopicity.

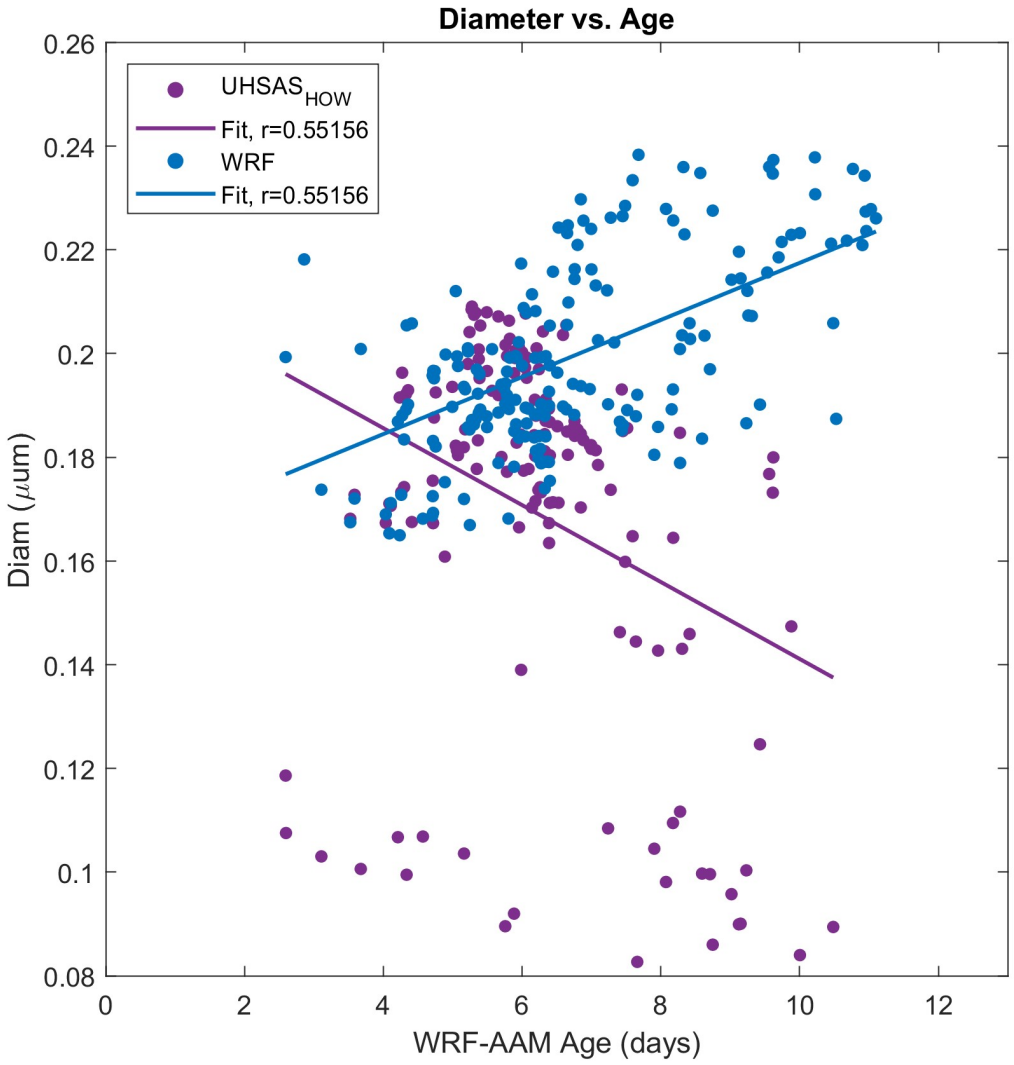


Fig. 12: Mean particle diameter aging trends in observations and WRF-CAM5. Age is calculated by tracers added to a WRF-AAM forecasting model build.

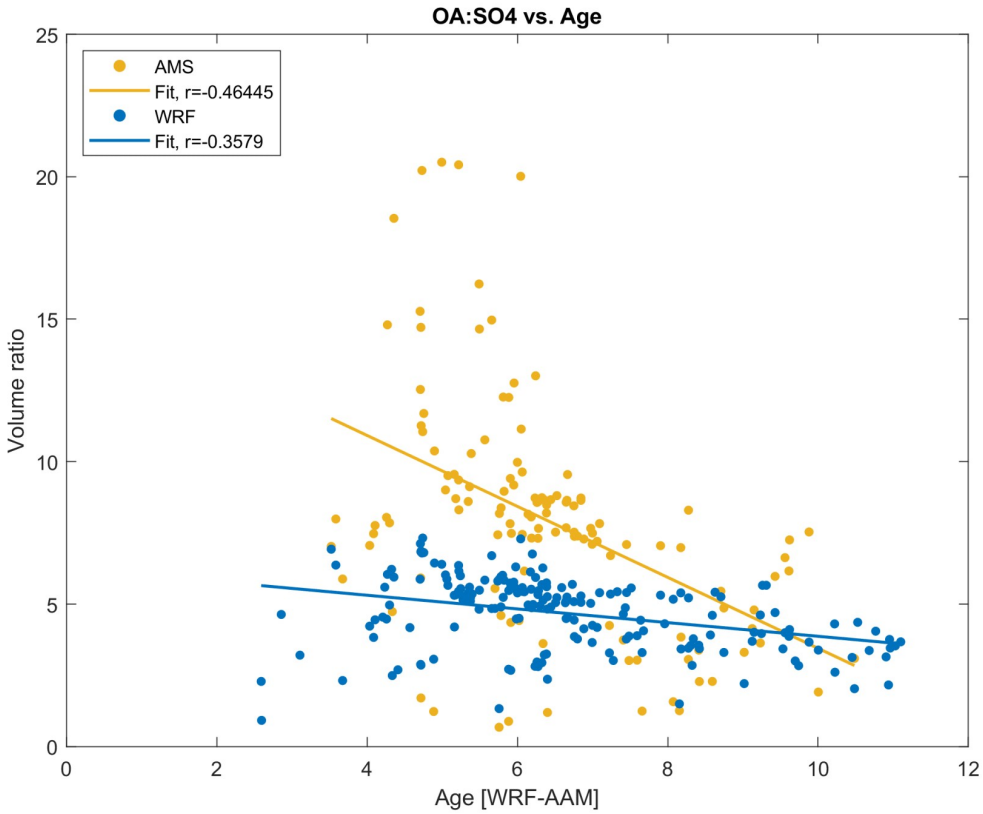


Fig. 13: Ratio of Organic Aerosol (OA) to SO_4 across smoke plume ages.

CONCLUSIONS

Marine stratocumulus cloud representation, including its interactions with aerosols, is critical to climate prediction. Here we built a framework to compare to an extensive set of campaign observations focused on biomass-burning aerosols (BBAs) in the southeastern Atlantic Ocean. Our study of WRF-CAM5 and these data has found:

1. WRF-CAM5 is underestimating aerosol activation into cloud droplets, in general.
2. WRF-CAM5 is underestimating hygroscopicity, likely the dominant factor influencing #1
3. Diameter is roughly well-captured, although particles are shrinking over time in observations, not the model.
4. Turbulent updrafts in the BL are drastically overestimated in the boundary layer.
5. Sulfate fraction in the boundary layer is far higher in observations than WRF-CAM5, despite smoke being well-represented in composition. This is likely an insufficient background generation process, rather than due to BBA emissions.

Understanding *why* models have a particular bias is necessary to constrain resulting aerosol-cloud-radiation interactions and make model improvements.

FUTURE WORK AND ACKNOWLEDGMENTS

1. Expand this multi-instrument, multi-campaign analysis framework to other models (E3SM, CESM, GEOS-Chem)
2. Sensitivity tests for changed parameters (hygroscopicity, density, size properties) and processes (OA losses) where possible.
3. Incorporate broader satellite data for cloud comparison
4. Analyze meteorological feedbacks (aerosol heating impacts on transport, wind patterns) and run on/off experiments with fire emissions and/or radiative interactions.

Acknowledgments

This work is supported by NASA ORACLES grant 80NSSC19K1463 and DOE LASIC grant DE-SC0018272. Special thanks to multiple ORACLES, LASIC, and CLARIFY data teams, and the University of California – Los Angeles Department of Atmospheric and Oceanic Sciences.

AUTHOR INFORMATION

Calvin Howes – Presenting Author

University of California – Los Angeles

Email: calvinhowes@ucla.edu

Pablo Saide - University of California – Los Angeles

Sharon P Burton – NASA Langley Research Center

Hugh Coe – University of Manchester

Amie Nicole Dobracki – University of Miami

Marta A Fenn – NASA Langley Research Center

Richard Anthony Ferrare - NASA Langley Research Center

Steffen Freitag - University of Hawaii

Siddhant Gupta – University of Oklahoma

Johnathan W Hair - NASA Langley Research Center

Steven G Howell – University of Hawaii

Janek Uin – Brookhaven National Lab

Mary Kacarab

Chongai Kuang – Brookhaven National Lab

L. Ruby Leung – Pacific Northwest National Laboratory

Sebastian Milinski – National Center for Atmospheric Research (NCAR)

Athanasios Nenes - Swiss Federal Institute of Technology Lausanne

Arthur J Sedlacek III – Brookhaven National Lab

Kenneth Lee Thornhill II - NASA Langley Research Center

Jenny Wong – Mount Allison University

Robert Wood – University of Washington

Huihui Wu – University of Manchester

Yang Zhang – Northeastern University

Paquita Zuidema – University of Miami

REFERENCES

Dinar, E., Mentel, T. F., & Rudich, Y. (2006). The density of humic acids and humic like substances (HULIS) from fresh and aged wood burning and pollution aerosol particles. *Atmos. Chem. Phys*, *6*, 5213–5224. Retrieved from www.atmos-chem-phys.net/6/5213/2006/

Haywood, J. M., Abel, S. J., Barrett, P. A., Bellouin, N., Blyth, A., Bower, K. N., … Zuidema, P. (2021). The CLoud-Aerosol-Radiation Interaction and Forcing: Year 2017 (CLARIFY-2017) measurement campaign. *Atmospheric Chemistry and Physics*, *21*(2), 1049–1084. https://doi.org/10.5194/ACP-21-1049-2021 (https://doi.org/10.5194/ACP-21-1049-2021)

Kuwata, M., Zorn, S. R., & Martin, S. T. (2011). Using Elemental Ratios to Predict the Density of Organic Material Composed of Carbon, Hydrogen, and Oxygen. *Environmental Science and Technology*, *46*(2), 787–794. https://doi.org/10.1021/ES202525Q

Morales, R., & Nenes, A. (2010). Characteristic updrafts for computing distribution-averaged cloud droplet number and stratocumulus cloud properties. *Journal of Geophysical Research: Atmospheres*, *115*(D18), 18220. https://doi.org/10.1029/2009JD013233

Petters, M. D., & Kreidenweis, S. M. (2008). A single parameter representation of hygroscopic growth and cloud condensation nucleus activity - Part 2: Including solubility. *Atmospheric Chemistry and Physics*, *8*(20), 6273–6279. https://doi.org/10.5194/acp-8-6273-2008

Shinozuka, Y., Saide, P. E., Ferrada, G. A., Burton, S. P., Ferrare, R., Doherty, S. J., … Zuidema, P. (2020). Modeling the smoky troposphere of the southeast Atlantic: a comparison to ORACLES airborne observations from September of 2016. *Atmospheric Chemistry and Physics*, *20*(19), 11491–11526. https://doi.org/10.5194/acp-20-11491-2020

Zuidema, P., Redemann, J., Haywood, J., Wood, R., Piketh, S., Hipondoka, M., & Formenti, P. (2016). Smoke and clouds above the southeast Atlantic: Upcoming field campaigns probe absorbing aerosol's impact on climate. *Bulletin of the American Meteorological Society*, *97*(7), 1131–1135. https://doi.org/10.1175/BAMS-D-15-00082.1