Evaluation of modeled aerosol-cloud interactions using data from the ORACLES and LASIC field campaigns

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Abstract

Aerosol-cloud interactions are both uncertain and important in global and regional climate models, and especially in the southeast Atlantic Ocean. This uncertainty in the region is largely due to two correlated 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 and LASIC field campaigns in August-October of 2016 through 2018. We compare aerosol and cloud properties to measure and improve model performance and expand upon observational findings of aerosol-cloud effects. Relevant comparison variables include aerosol number concentration, mean particle diameter and spread, CCN activation tendency, hygroscopicity, and cloud droplet number concentrations. Specifically, our approach is to analyze colocated model data along flight tracks to resolve aerosol-cloud interactions. Within and between single-day flights, there is high spatiotemporal variability that can get lost to large-scale averaging analyses. We have found that CCN is substantially under-represented in the model compared to observations. For a given aerosol number concentration, size, supersaturation and hygroscopicity, the model will consider fewer particles as CCN than observations indicate. We plan to explore this result further, diagnosing the model-observation differences more consistently and updating the model with more physically accurate values of aerosol size, concentration, or hygroscopicity based on observations. We will also intercompare multiple instrument platforms involved with the ORACLES and LASIC

campaigns. With improved small-scale aerosol-cloud interactions, this work also shows promise to substantially improve that representation in climate models.

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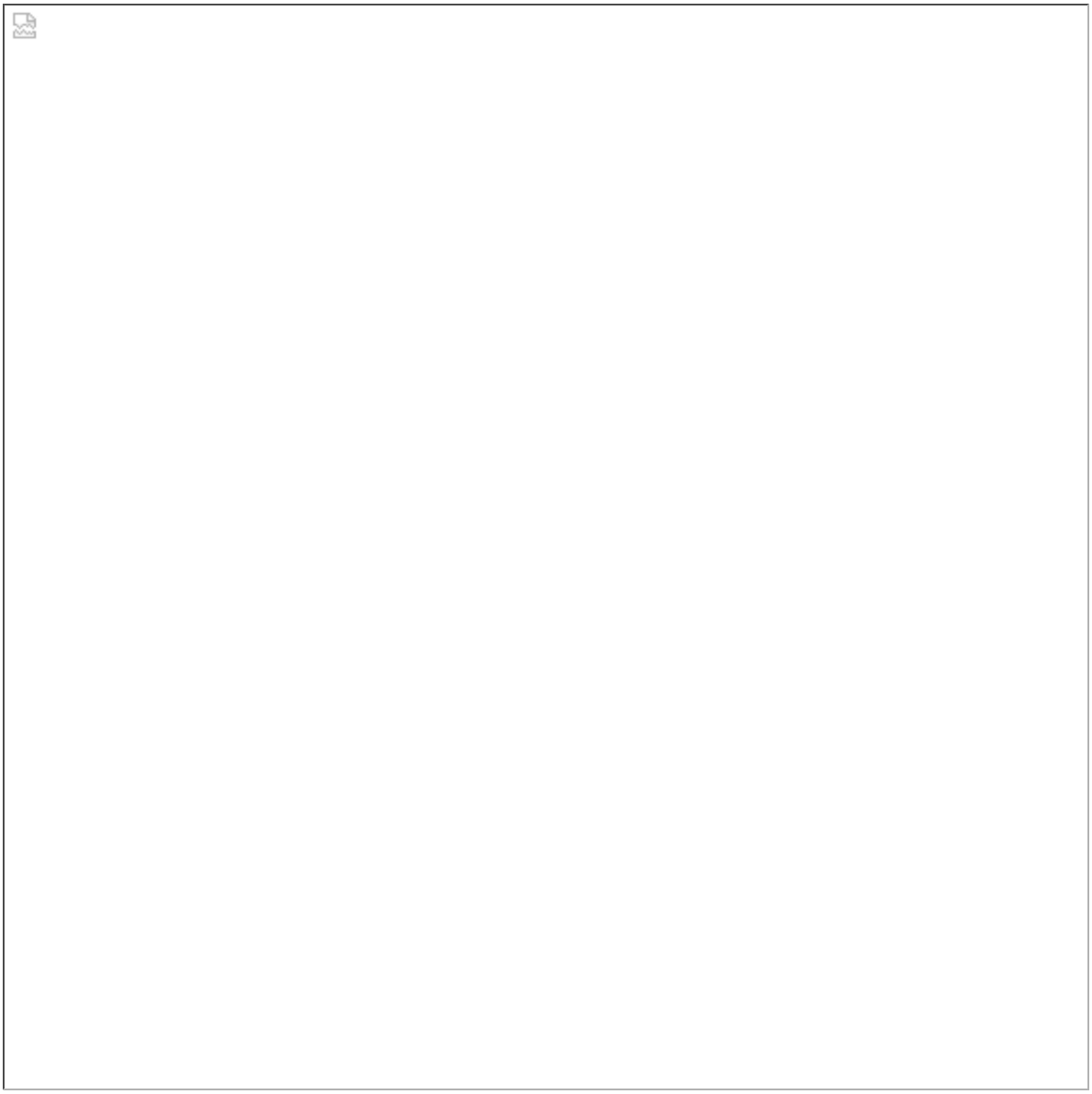

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INTRODUCTION

Southern Africa is the largest source of biomass-burning smoke in the world, and it advects westward over a persistent South Atlantic stratocumulus deck (Fig. 1). Aerosol-cloud-radiation interactions in the area contribute to significant uncertainty in global climate models (Fig. 2).

In particular, the ability for smoke aerosols to activate into cloud droplets is highly variable, and accurate representation depends upon accurate intensive aerosol variables. Aerosol chemical composition and size distribution, for example, can be substantially improved with observational constraints. The ORACLES and LASIC campaigns set out to better quantify these interactions and constrain model predictions [1,2]. This work uses those observations to test WRF-CAM5 performance.

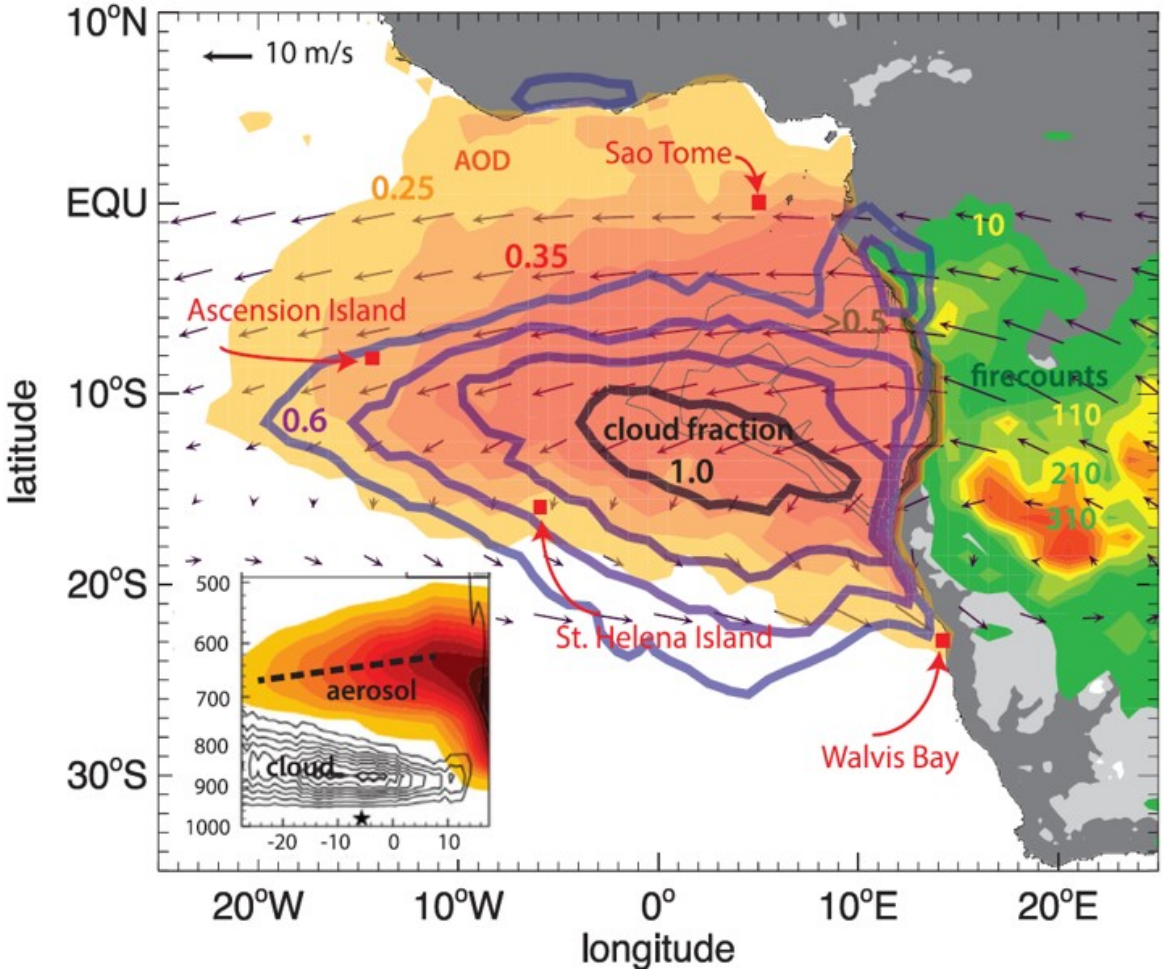


Fig 1. Smoke AOD and high cloud fraction overlap in the Southeast Atlantic. *Stier et al., 2013*

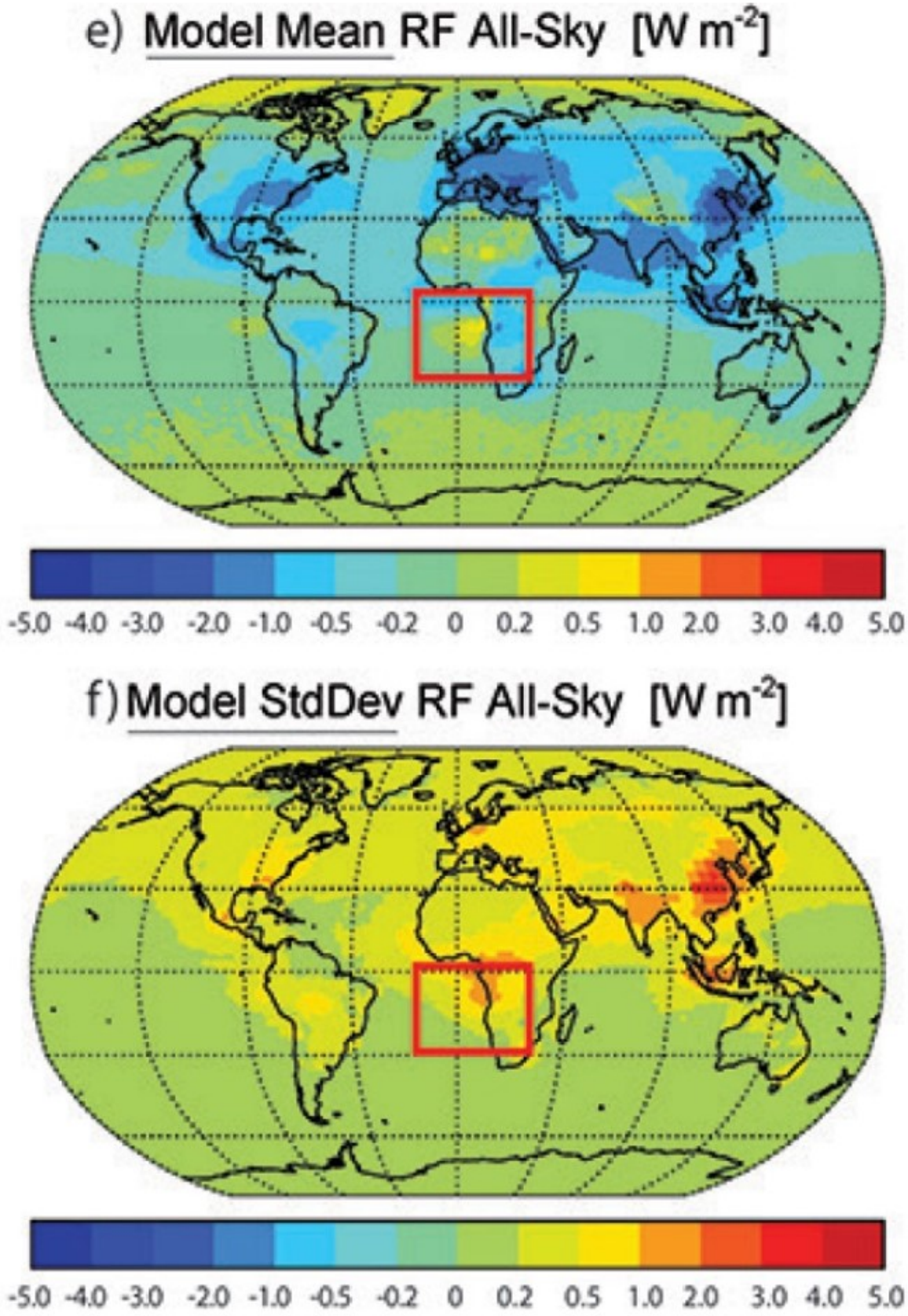


Fig 2. Aggregate model all-sky radiative forcing mean and standard deviation. Note the high value and high uncertainty in the Southeast Atlantic region.

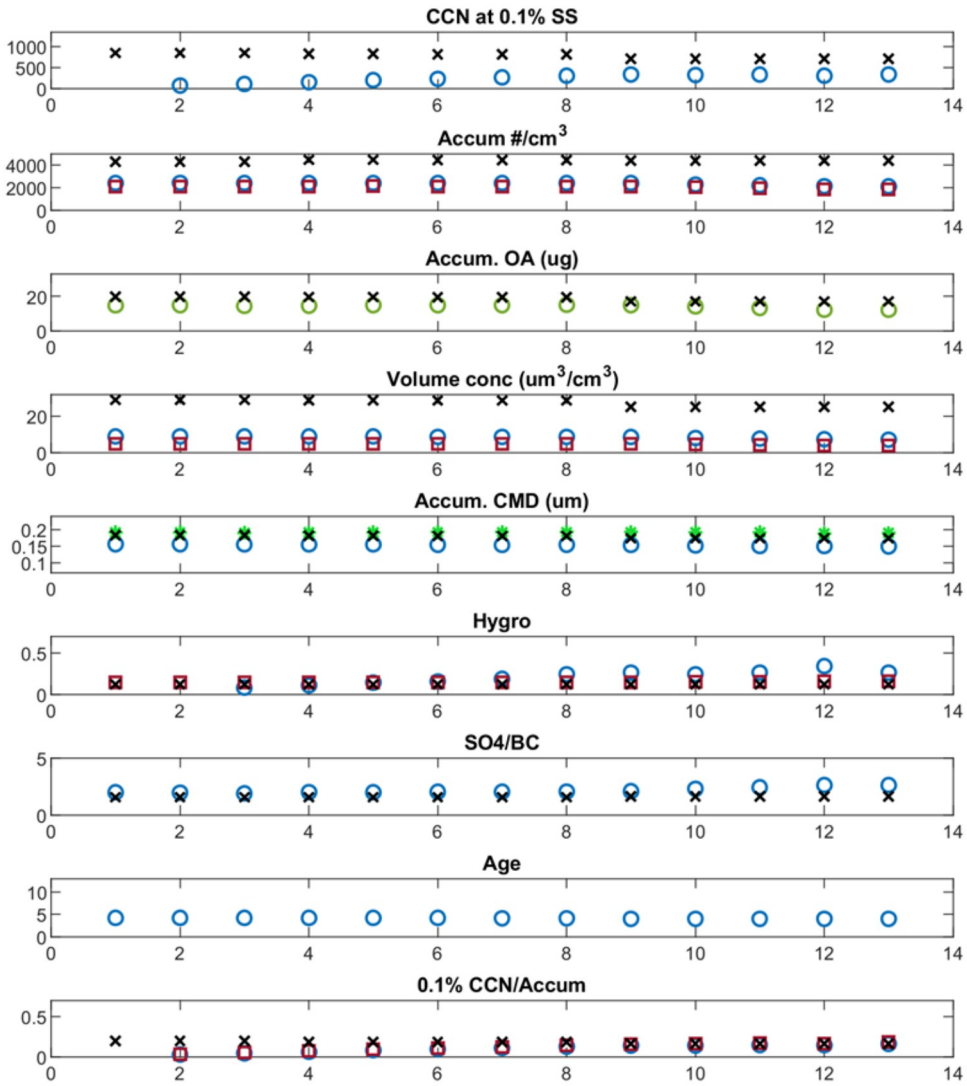
METHODS

- We used WRF-Chem with CAM5 aerosols, CAMS boundaries, 36x36km horizontal resolution, and 74 vertical layers with variable spacing. The model ran from July 15-Sept 1, 2017 in 7-day cycles with 2-day spin-up, reinitialized from Final Analysis NCEP and persistent aerosols between cycles. Age was calculated from a WRF-AAM forecasting model configuration, used successfully for the ORACLES campaign.

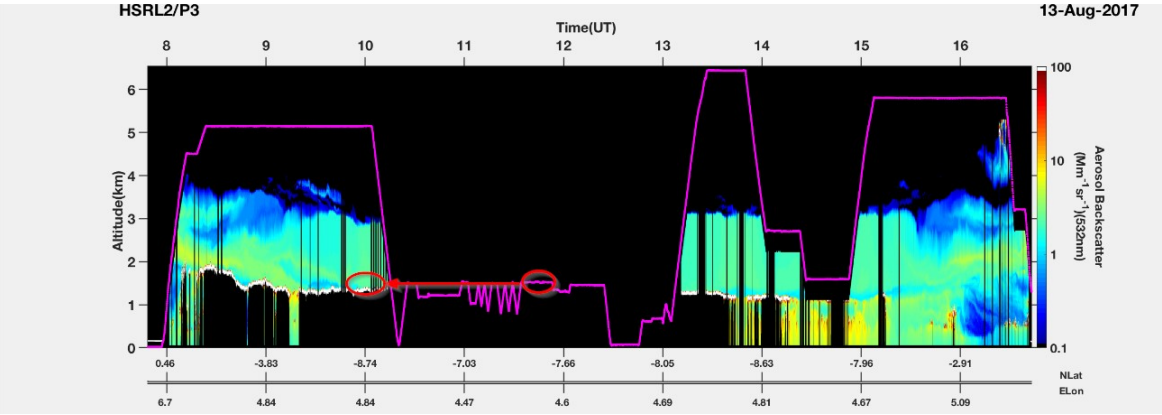
- Model data were compared with observations from a P-3 aircraft flying for the NASA ORACLES campaign and the LASIC ARM facility on Ascension island. The P-3 instrument suite included two UHSAS aerosol samplers, one aerosol mass spectrometer (AMS), and a CCN counter, among many other *in situ* and remote sensing instruments. The LASIC aerosol data from Ascension Island included a Scanning Mobility Particle Sizer (SMPS), used here, as well as an extensive *in situ* and remote sensing suite that will be used to expand these results [1,2].

We extracted small time windows of several minutes to an hour, called 'test cases' in which many instruments' data were available, where the aircraft was flying level in the lower troposphere, and where smoke properties remained relatively constant. This strategy allows us to analyze model performance on intensive properties without losing information to averaging done in large-scale studies centered on this campaign [3]. Fig. 3 shows a single test case in detail from the flight on 13th August 2017.

8-13


CCN at 0.1% SS
Accum #/cm3
Accum. OA (ug)
Volume conc (um3/cm3)
Accum. CMD (um)
Hygro
SO4/BC
Age
0.1% CCN/Accum

(a)


HSRL2/P3
13-Aug-2017
Time(UT)
Altitude(km)
Aerosol Backscatter (Mm-1 sr-1)(532nm)
NLat
ELon

(b)

Fig 3. (a) A demonstration of the comparison variables from WRF and various instruments. WRF is black x's, and various instruments are the colored circles (variously CCN counter, UHSAS, GIT-UHSAS, AMS, and Nenes derived K). Each point is data averaged over 60 seconds. (b) Backscattering ratio, showing the position of the plume. The right-most red circle is the flight transect segment used for the test case, and the left-side red circle highlights the plume position the aircraft flew back through. This test case was at roughly 1.3km altitude, and from 14:33 to 14:45 UTC.

The ongoing goal of this work is to measure consistency between this model and observations. This test case approach addresses that goal by answering model performance questions. For example, given a supersaturation, aerosol number, size distribution, and chemical composition, how many of those particles may be considered CCN?

To aid in answering that question and others, we derived three quantities. The first is the ratio of CCN number concentration to accumulation mode number concentration (a ratio referred to here as CCN/#). The CCN/# ratio represents the ability of a given particle to be activated as CCN, regardless of extensive variables like smoke plume concentration. For a smoke plume, this ability depends on both the size distribution and its hygroscopicity. This calculation assumes that the CCN and UHSAS number concentrations are unbiased.

The second derived quantity is hygroscopicity from the AMS, calculated as a volume-weighted average of the main aerosol chemical constituents. This assumes total internal mixing, as well as assuming some reasonable and fixed values for the hygroscopicity per species.

The third derived quantity is the count-mean diameter using volume from AMS and number concentration from UHSAS. This assumed that the AMS captured the great majority of the aerosol mass in its observed species and that UHSAS number concentration is unbiased.

SIZING AND INSTRUMENT ISSUES

CCN bug fix

We discovered a bug in the WRF-Chem cloud droplet activation code. This bug meant that diagnostic CCN number concentration was **not** being calculated from a particle diameter based on mass and number concentration, as it should be. Instead, the model was using a prescribed diameter for all model cells and times. We coded a fix that calculated diameter appropriately based on aerosol mass and number concentration. This resulted in a generally higher CCN concentration, bringing WRF closer to observations for most cases (Fig. 4). This fix was added to the WRF-Chem repository and has the potential to significantly impact any studies comparing WRF-Chem CCN concentrations to observations or other models.

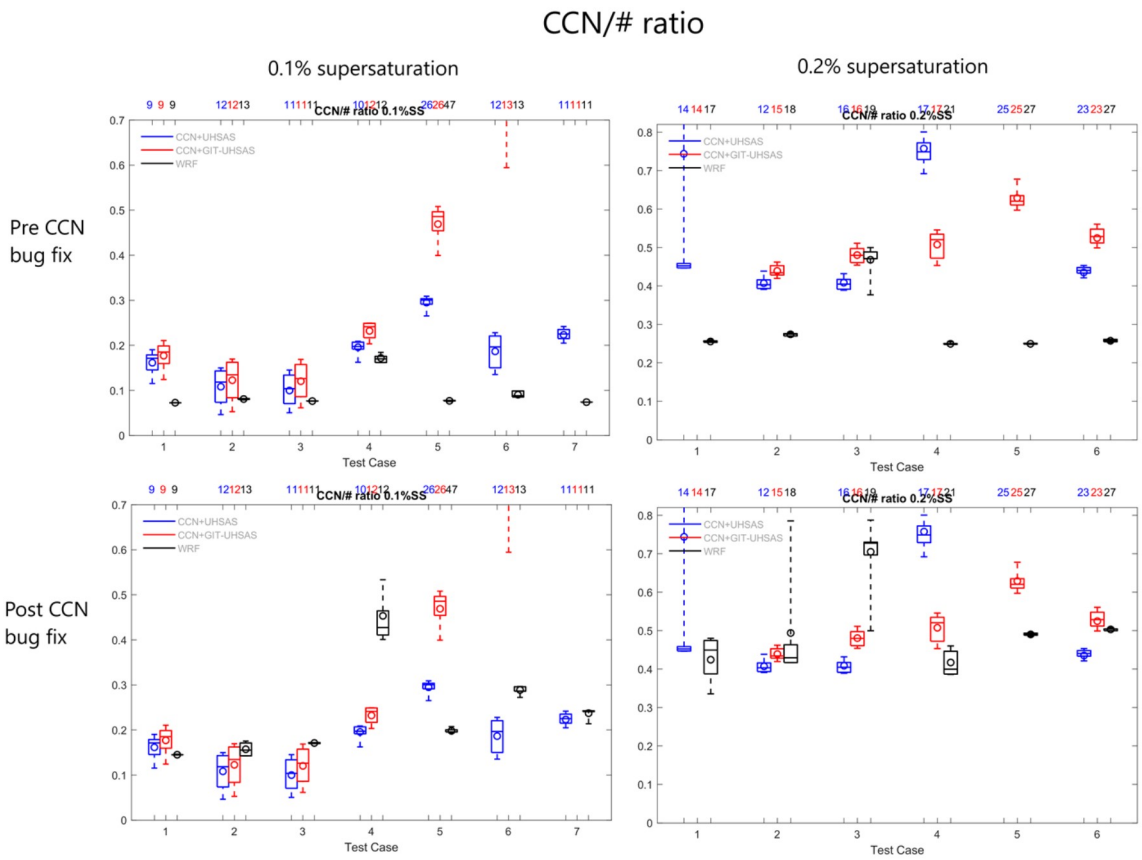


Fig. 4 CCN/# ratio before (top) and after (bottom) fixing the bug in WRF-Chem CCN calculation. Modeled ratio values become much closer to observations, especially at 0.2% supersaturation.

Adjustments to the modeled size distribution

Based on observations from ORACLES and LASIC, we adjusted WRF-CAM5's standard deviation, σ, from 1.8 to 1.5 for the accumulation mode. This increased number significantly above observations, as expected, but brought the model diameter closer to observations. All figures shown here are from the simulation with σ=1.5.

The two UHSAS instruments on the ORACLES flights both had systemic under-sizing issues, although total particle counts were likely accurate [4]. This led to unrealistically small estimates for the diameter and overestimates of hygroscopicity. We used AMS-derived chemical composition to recalculate Kappa, and AMS volume to recalculate diameter, in combination with UHSAS number concentration. We assumed a lognormal aerosol size distribution with σ=1.5 from the UHSAS. This AMS derivation is likely not a perfect analog for

particle size--even though it is an improvement--in part because it has values that extend substantially beyond what even size-corrected UHSAS values would suggest.

The SMPS was based on Ascension Island as part of the U.S. Dept. of Energy LASIC campaign. It gave another independent estimate of the smoke's accumulation mode mean diameter at 176nm, with a 10th-90th percentile range of 159-195nm. This backs up the likelihood that the UHSAS size values were too low, including the UHSAS that was part of LASIC.

Limited data overlap

Another major limiting factor was the availability of instrument data, especially AMS chemical composition and CCN number concentration at a given supersaturation. The small number of times in which all instrument data was available limited the sample size at which we could compare model performance in detail.

This is a somewhat unavoidable problem during large field campaigns on short time scales. However, future work will incorporate more campaign years and can potentially replicate this comparison strategy across other, similar campaigns.

MODEL PERFORMANCE

Diameter

WRF-CAM5 is able to capture the range of particle diameters sampled by the UHSAS (Fig. 5). The AMS + UHSAS alternate diameter calculation, however, gives a much larger range than WRF does.

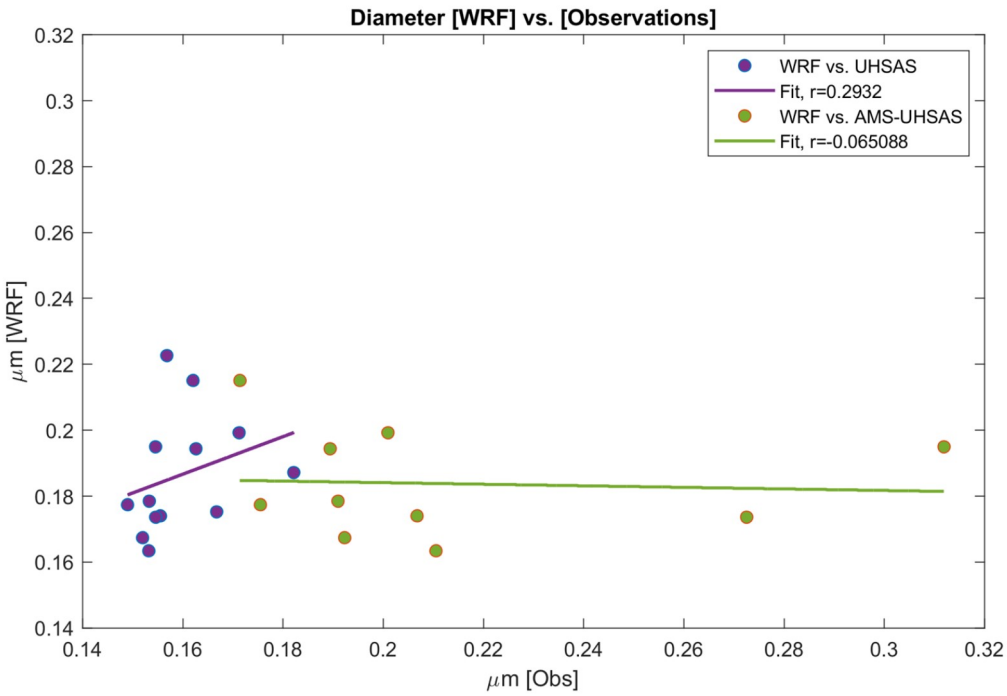


Fig 5. Count-mean diameters from WRF-CAM5 vs. observations. The UHSAS has a slightly larger sample due to gaps in AMS data

Hygroscopicity

WRF-CAM5 follows a similar trend for K as both AMS and CCN-counter-based estimates. It is less biased against AMS than UHSAS K values (Fig. 6). This indicates a similar and accurate underlying chemical process in WRF-CAM5. Due to UHSAS under-sizing problems, it's likely that the lower range of K derived from the AMS is more accurate. Large UHSAS-based values for K persist through the ORACLES campaign [5].

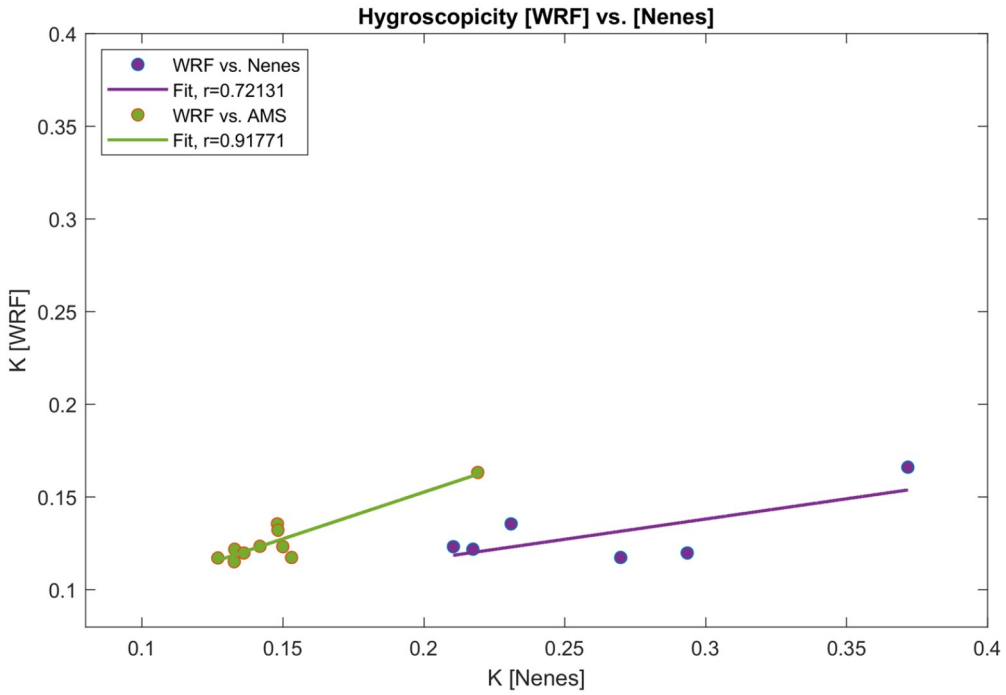


Fig 6. Hygroscopicity parameter from WRF-CAM5 vs. observations. K values from the Nenes group were not available during 0.2% test cases and AMS values were not available during all test cases, leading to different sample sizes.

WRF-Chem also performed well for hygroscopicity values calculated from the LASIC SMPS (Fig. 7).

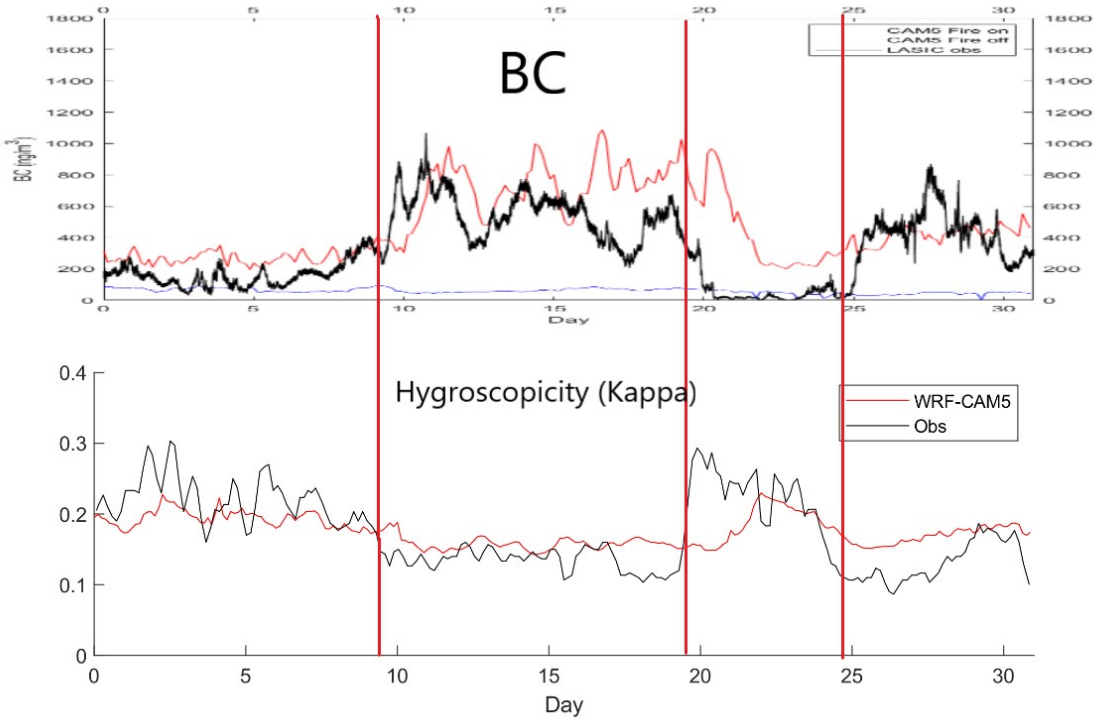
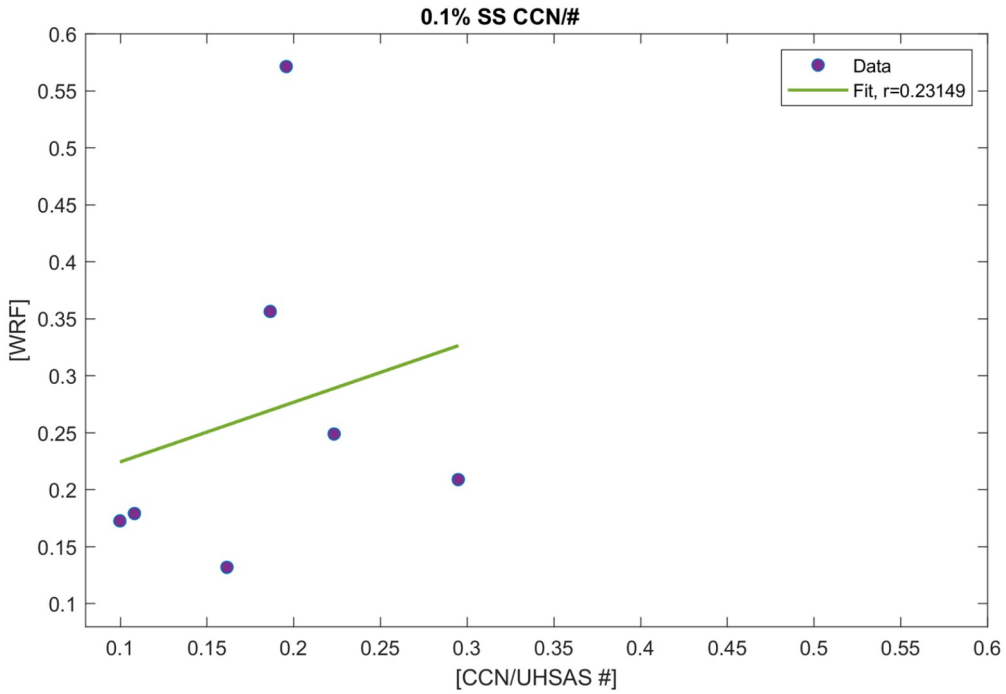


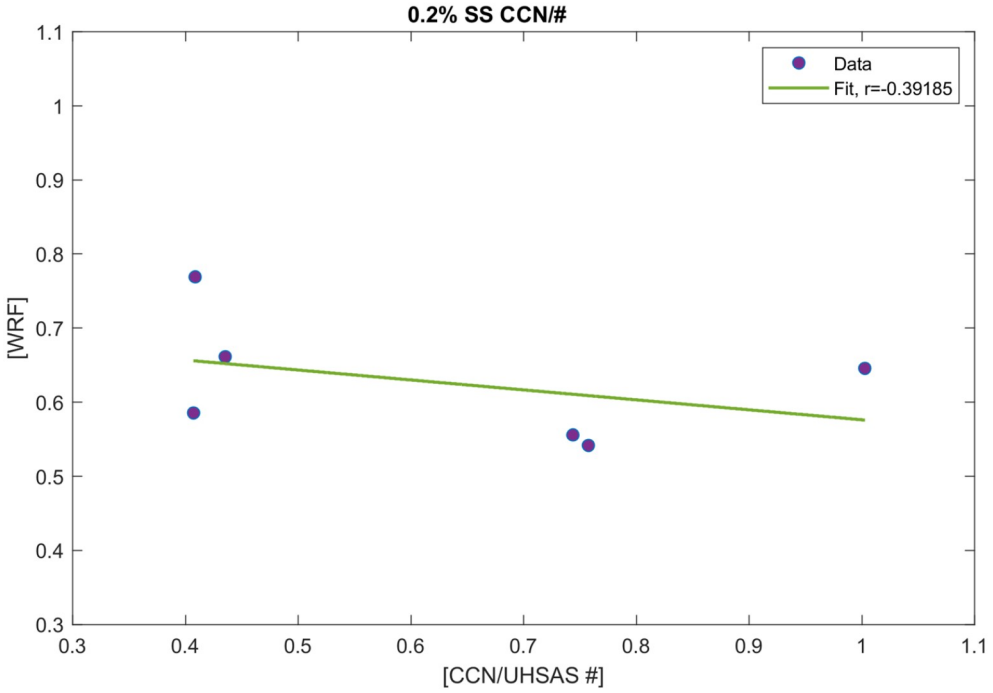
Fig 7. Top, WRF-CAM5 vs. observed BC mass concentration at Ascension Island. Bottom, model vs. observed K. Observations derived from LASIC SMPS. Model performance vs. LASIC observations in August 2017 show reasonably good agreement in range and timing during smoky periods. Averaged per 9 hours for visual clarity.

CCN/# ratio

WRF tends to represent the observed range well for test cases measured at 0.1% supersaturation, even without a strong correlation (Fig. 8a). The 0.2% cases have a much larger observed value range than the model (8b). This could indicate an improperly fitted CCN function in MAM but requires a larger sample size to confirm.



(a)

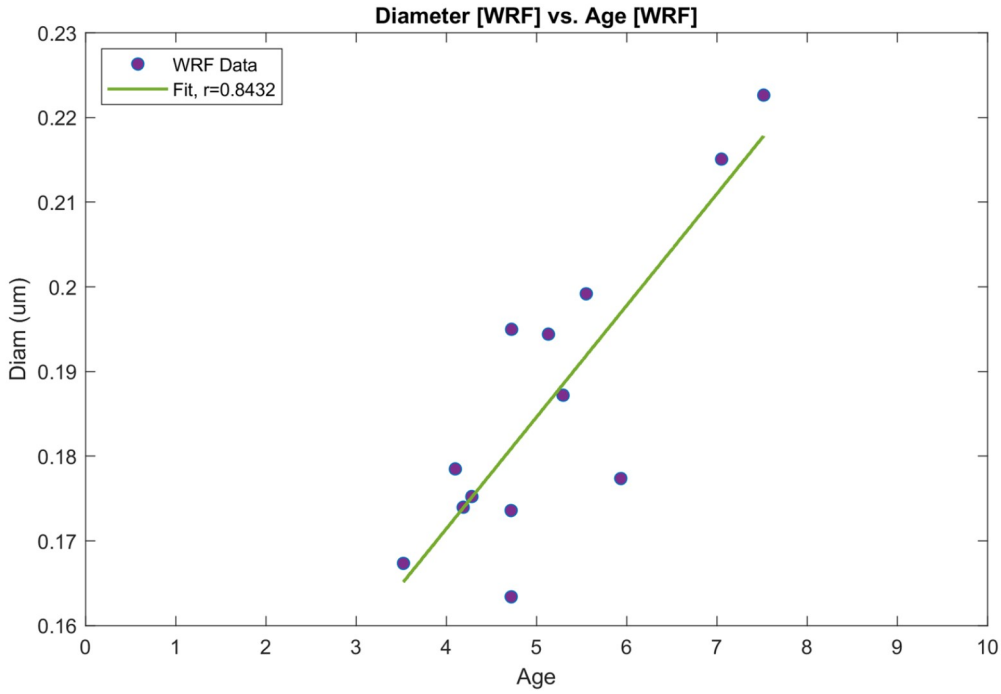


(b)

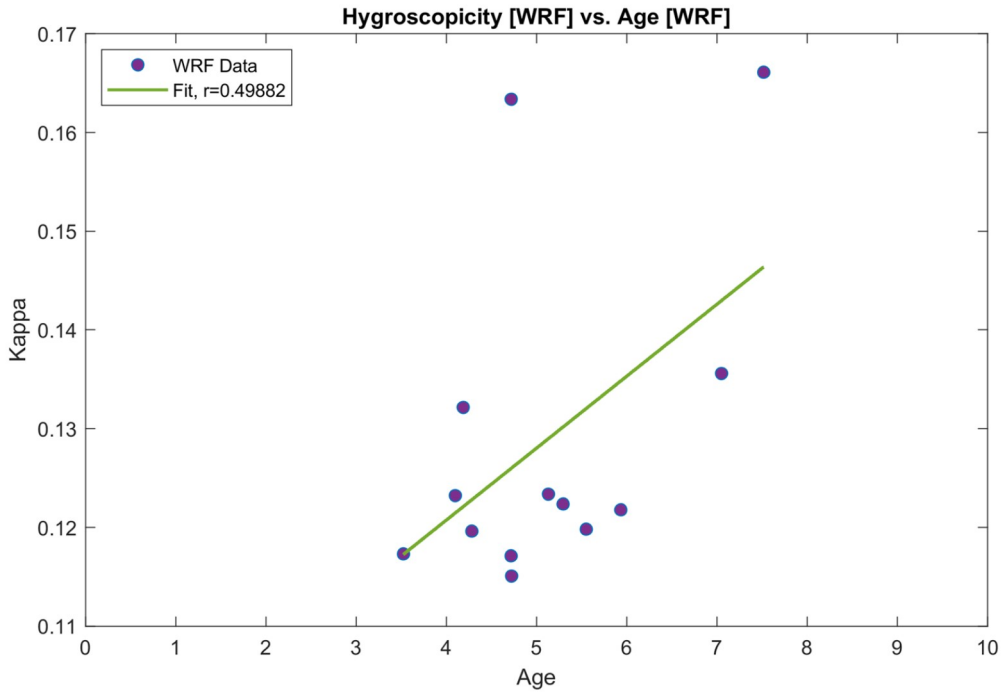
Fig 8. WRF-CAM5 vs. observed values for CCN/# ratio (accumulation mode number concentration from UHSAS, for observations). Separated by supersaturation for (a) 0.1% and (b) 0.2%.

Model-age comparison

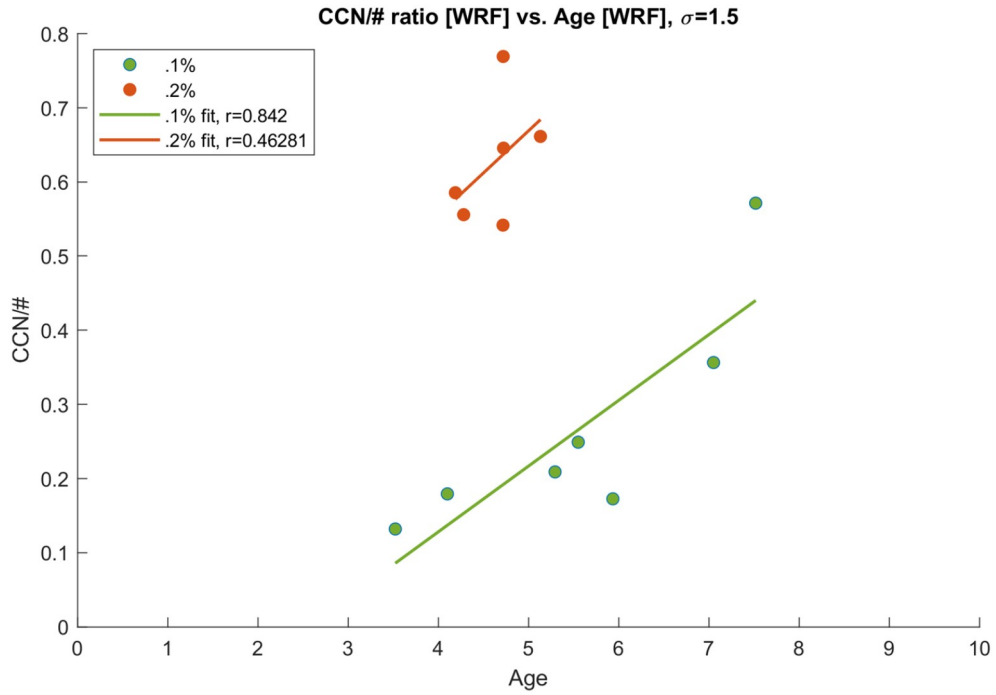
High correlation coefficients for age against diameter, hygroscopicity, and CCN/# ratio (Fig. 9a-c) give a strong indication that modeled aging processes explain much of the behavior of these variables in WRF.



(a)



(b)



(c)

Fig 9. Comparing variables from WRF-CAM5 vs. age for (a) diameter, (b) hygroscopicity, and (c) CCN/# ratio. 0.1% and 0.2% cases are combined in (a) and (b) as they do not depend on supersaturation.

Observation-age comparison

Observations generally do not correlate well with modeled plume age through these test cases (Figs 10-12). More work remains to verify that these cases are representative of larger trends during the campaign, such as the correlation of age with OC:BC ratio and SSA. This will help confirm whether the variability in observations is driven by aging at all and whether the WRF-CAM5 aging mechanisms are a candidate for improvement.

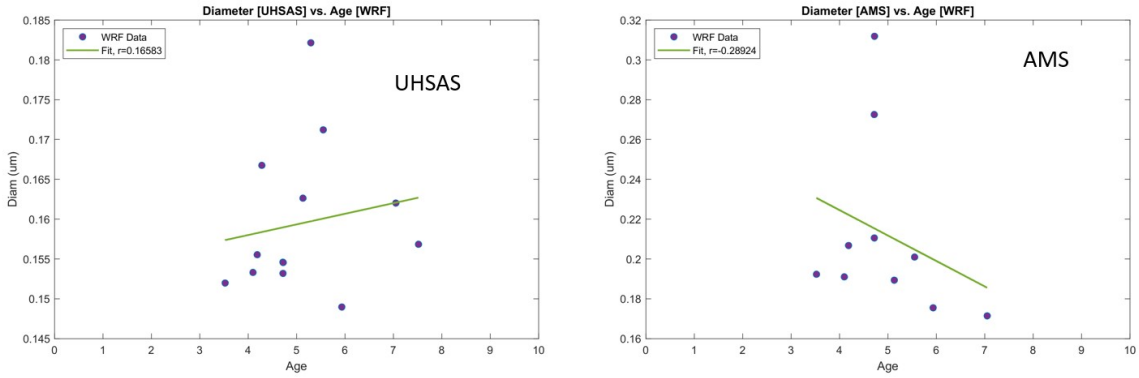


Fig 10. Observed diameter vs. WRF-AAM age for the UHSAS (left) and AMS+UHSAS (right).

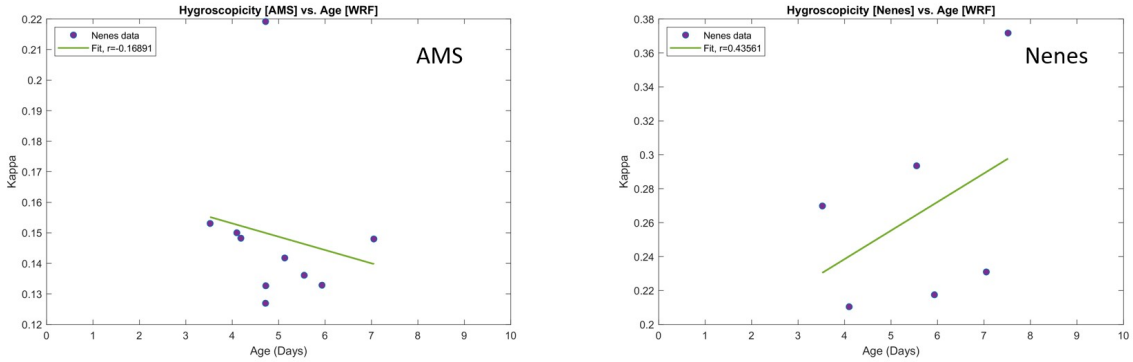


Fig 11. Observed hygroscopicity vs. WRF-AAM age from the Nenes team (left) and AMS (right).

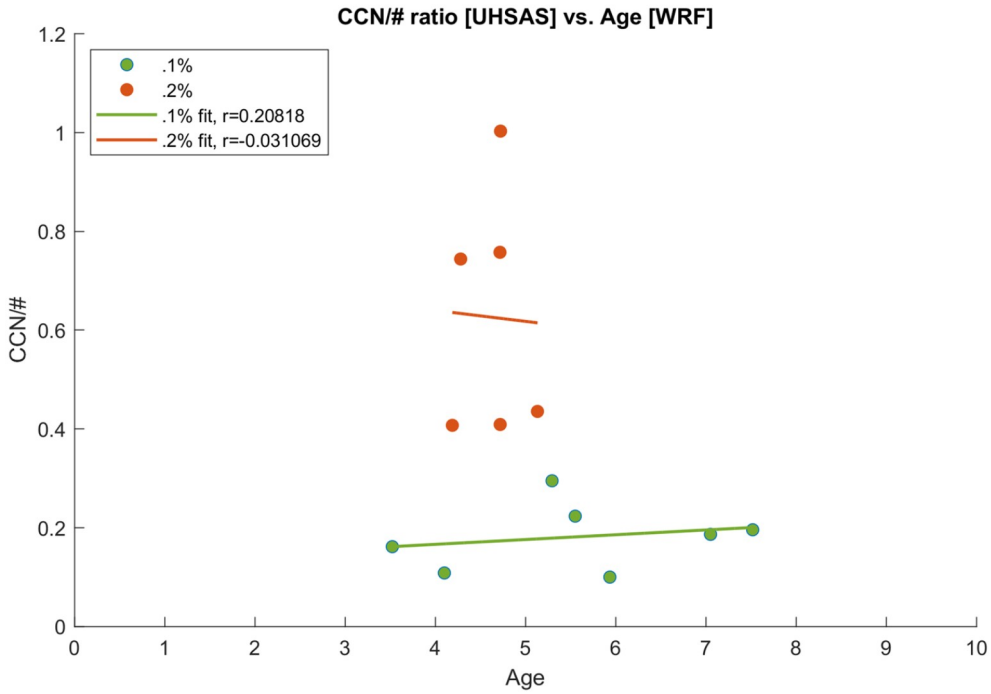


Fig 12. Observed CCN/# vs. WRF-AAM age.

CONCLUSIONS AND NEXT STEPS

By comparing WRF-CAM5 performance to multiple instruments' observations in test cases, we have assessed WRF's performance in a way that large-scale averaging comparisons don't allow. Under this strategy, we have found that WRF is able to capture a similar range of values for parameters that are key to cloud activation, especially hygroscopicity and diameter. The actual likelihood for a particle to be activated as CCN, the CCN/# ratio, has its value range captured well by the model for 0.1% supersaturation, but 0.2% supersaturation has a much larger spread in observations than in WRF-Chem. This may indicate an underlying process deficiency in the model or inadequate sample size.

WRF age processes are internally consistent with the physical aging they represent. The strong correlation between WRF age vs. diameter and CCN/#, and the modest correlation for age vs. hygroscopicity, show that model aging is likely driving these changes.

Future work

Within these test cases, WRF age does not fully explain the range of observations. We will investigate this further by expanding the sample size and confirming previous age correlations. It is possible that WRF chemical aging is a candidate to improve cloud activation response in the future.

We will also be including more variables to classify differences between model and observation responses. This will include optical properties tied to smoke properties and age such as SSA and MEE, as well as relative chemical composition.
Cloud properties will also be included explicitly, as the goal is examining cloud activation changes.

We are also planning on increasing our sample size by expanding the analysis to 2016 and 2018, the other ORACLES campaign years, and running a more detailed comparison with data from the LASIC campaign.

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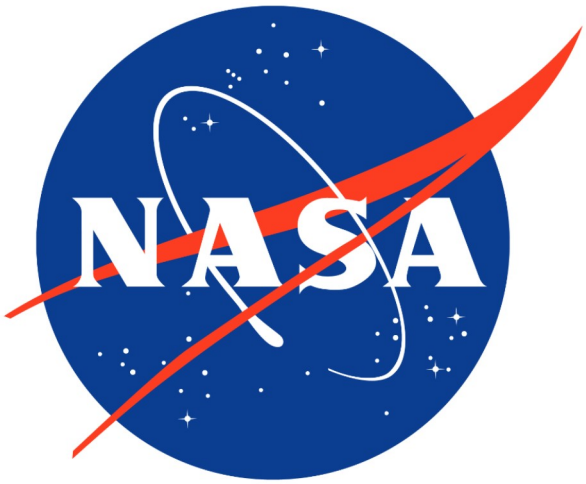

AUTHOR INFORMATION

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ABSTRACT

Aerosol-cloud interactions are both uncertain and important in global and regional climate models, and especially in the southeast Atlantic Ocean. This uncertainty in the region is largely due to two correlated 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 and LASIC field campaigns in August-October of 2016 through 2018. We compare aerosol and cloud properties to measure and improve model performance and expand upon observational findings of aerosol-cloud effects. Relevant comparison variables include aerosol number concentration, mean particle diameter and spread, CCN activation tendency, hygroscopicity, and cloud droplet number concentrations. Specifically, our approach is to analyze colocated model data along flight tracks to resolve aerosol-cloud interactions. Within and between single-day flights, there is high spatiotemporal variability that can get lost to large-scale averaging analyses. We have found that CCN is substantially under-represented in the model compared to observations. For a given aerosol number concentration, size, supersaturation and hygroscopicity, the model will consider fewer particles as CCN than observations indicate. We plan to explore this result further, diagnosing the model-observation differences more consistently and updating the model with more physically accurate values of aerosol size, concentration, or hygroscopicity based on observations. We will also intercompare multiple instrument platforms involved with the ORACLES and LASIC campaigns. With improved small-scale aerosol-cloud interactions, this work also shows promise to substantially improve that representation in climate models.

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