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Reply on RC3

Michael S. Diamond et al.

Author comment on "Cloud adjustments from large-scale smoke–circulation interactions strongly modulate the southeastern Atlantic stratocumulus-to-cumulus transition" by Michael S. Diamond et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-411-AC3, 2022

We would like to thank the reviewer for their constructive comments. Please find a detailed response that outlines our changes in response to the suggestions below.

"1. Why do the authors use different meteorological datasets to run trajectories in the boundary layer and free-troposphere in figure 3? Also, for the free-tropospheric trajectories, why was 2 km altitude chosen as a starting point i.e. above the boundary layer/middle of the aerosol plume etc. Noting that the observed aerosol plume closer to Ascension Island is at a higher altitude (Fig 2)."

We ran the 2 km trajectories using the system in place for in-field forecasting, whereas the back trajectory used to set up the LES case was run specially using the WRF-CAM5 wind fields. There may be slight discrepancies between the WRF-CAM5 and GDAS wind fields (although it is worth noting that WRF-CAM5 was initialized with NCEP reanalysis fields that should be consistent with GDAS); however, as we are only using the 15 August 2017 data as a loose constraint with a relatively large tolerance for "matches" (within a degree of longitude on either side of 5° E from 07:00 to 18:00 UTC), we doubt that the greater precision from using the WRF-CAM5 wind fields would have any bearing on our conclusions.

2 km was chosen as the trajectory starting point to safely remain above the MBL while still being fairly representative of the aerosol entraining into the MBL. The higher-altitude aerosol plume near Ascension Island is not directly related to the smoke plume/ex-CBL that entrained into the MBL during our case and could not have contributed any aerosol to the MBL during our period of interest. We have added some clarifying text to Section 2.1: "2 km is chosen as the trajectory starting point to safely remain above the MBL while still being fairly representative of the aerosol entraining into the MBL."

"2. WRF has a high bias in the FT aerosol number concentration, but the OC and BC mass loadings are comparable to those observed (Fig S1). Can the authors confirm that the model data in Fig 4a/Fig S1b is the accumulation mode only that this mode does represent a comparable size range as the aircraft measurement? If yes, are there significant concentrations of aerosol species in addition to OC and BC in the model that might account for the difference?"

The model number concentration data in Figure 4a/Figure S1b are for the accumulation mode bin of the MAM3 scheme, in which the mode dry diameter ranges between 58 and 270 nm (10th to 90th percentiles) and the geometric standard deviation is set at 1.8 (Liu et al., 2012; their Table 1). The UHSAS data used is for dry diameters between 60 and 600 nm and the PCASP data is for dry diameters between 100 and 3,000 nm. The smoke mode is just under 200 nm and the vast majority of the number concentration is captured by MAM3 and both instruments. For mass, neither the model nor the observational data are constrained to represent the accumulation mode only.

Sulfate contributes to the aerosol number and mass concentrations in addition to OC and BC, although there are questions about to what extent the aerosol particles are internal or external mixtures in reality (they are internal in MAM3). Sulfate also doesn't differ strongly between WRF-CAM5 and the observations, at least during the 15 August 2017 flight. Work to better understand how the WRF-CAM5 aerosol size and mass distribution compares to observations and the reasons why they may differ is ongoing (Howes et al., *in prep*).

A clarification of the size range has been added to Section 2.3.1: "A modal aerosol module (Liu et al., 2012) with Aitken, accumulation [mode dry diameters from 58 to 270 nm (10th to 90th percentiles) and geometric standard deviation of 1.8], and coarse modes (MAM3) coupled with a gas phase chemistry scheme (Zaveri and Peters, 1999) is used for smoke (and other aerosol) properties. (N_a from WRF-CAM5 refers to the accumulation mode only unless otherwise specified.)"

"3. Figure 5 uses altitude (km) and Fig 6 uses pressure (hPa) on the y-axis. Is it possible to have consistency so that features at different altitudes can be compared more easily?"

Changed to using altitude on the y-axis.

"4. Figure 7: Can the authors comment on why the trend in cloud drop concentration in the WRF model does not follow the MBL aerosol number concentration, particularly towards the end of the trajectory? This seems counterintuitive and is different to the results from the SAM model shown in figure 15."

The activation fraction in WRF decreases with increasing aerosol loading, which is reasonable behavior for highly polluted MBLs (Kacarab et al., 2020). MBL-average TKE, and thus updraft velocities, decline over time in WRF as well, which can further explain decreasing activation fractions. The SAM activation fractions (near unity even in very polluted conditions) may be the unrealistic ones. A discussion was added to Section 3.1: "In all cases, the fraction of aerosol activating to form cloud droplets decreases over time as the MBL becomes more polluted (Figure 7d) and MBL-average TKE and thus updraft velocities decline (Figure 7h)."

"5. Figure 8 d: "$\Theta_{l,FireOn} < \Theta_{l,FireOff}$ at plume top" is only strictly true if one considers the plume top height being fixed at the height of the FireOn simulation. Θ_l at the plume top height of the FireOff/RadOff simulations is actually cooler than the value at the plume top height of the FireOn simulation."

We have now clarified in the caption that "plume top" in the figure refers to the FireOn plume top: ""Plume top," "in plume," and "below plume" here refer to the location of the FireOn ex-CBL."

"6. Figure 14: Can the authors change the colour bar for cloud water mixing ratio, given that it uses similar colours to the cloud droplet concentration shading on which it is overlaid."

Changed to purple instead of blue.

"7. Line 878: Consider rephrasing "the effect of subsidence exceeds that of the indirect effect", given that this is only when comparing against the baseline aerosol case. Arguably, the most dramatic impact on the cloud field does result from microphysics i.e. in the simulation that reduces the FT aerosol concentration by a factor of 2."

Changed to: "The effect of subsidence exceeds that of the indirect effect in this study except in the case of extremely low background aerosol concentration."

"8. Section 5: Refer the reader to Barrett et al. (2022) for a more complete description of the joint flight."

Done. "The reader is referred to Barrett et al. (2022) for a more complete description of the joint flight."

"9. Line 910: Can the authors also comment on possible reasons for the cool and moist bias in the MBL in the SAM simulation, when compared to the observations."

This is likely related to the cool/moist biases in the entraining FT air. We have added text in Section 5: "The cooler and moister MBLs simulated in WRF-CAM5 and SAM also may be related to entrainment of cooler and moister FT air than in reality due to the prolonged plume/ex-CBL presence."

"10. Line 913: Although the regional simulation is free-running, I would have thought that the aerosol plume takes several days to reach Ascension Island from the source region, as can be inferred from the trajectories in figure 3. Assuming that the model was initialised with a realistic aerosol location/amount near the source region, then does this point to model errors in the aerosol transport further offshore? Have the authors made any comparisons with satellite measurements of the plume location throughout the simulation period e.g with something like CALIOP/CATS/MODIS above cloud AOD/SEVIRI above cloud AOD etc, in order to examine this?"

Freshly-emitted smoke on the continents does take several days to reach the offshore cloud areas of interest here. There is pre-existing smoke over the southeast Atlantic at the 14 August 2017 point of reinitialization that came from the previous WRF-CAM5 runs. Aerosol is only initialized once from CAMS in July and then evolves freely afterward (with CAMS continuing to provide boundary conditions). We have added a clarification of this point in Section 2.3.1: "Smoke already over the southeast Atlantic at the start of the reinitialization for the FireOn and RadOff simulations comes from the previous WRF-CAM5 initializations and new smoke is generated over the continent based on the QFED emissions."

We did roughly compare plume properties/location with CALIOP. There are matches with qualitative agreement between the CALIOP plume location and WRF-CAM5 toward the beginning of the trajectory. The model evaluations in Yohei et al. (2020) and Doherty et al. (2022) also support WRF-CAM5 simulating realistic smoke transport.

"11. Line 995: It may be worth re-stating here, that the simulations that followed the classical model of an entrainment driven transition had much higher aerosol loadings in both the free troposphere and in the boundary layer than were measured by the ground site and aircraft. Suggesting that they may not be representative of typical conditions in this part of the SE Atlantic e.g. Fig 20."

The statement that the simulations following the entrainment-driven transition occurred for higher aerosol loadings than that observed is not true for the NOff and AllOff cases, which have aerosol loadings much lower than observed (as expected based on the exclusion of smoke particles). We have added WRF FireOff and SAM AllOff and AllOff$_{N/2}$ values to Figure 20 and additional text in Section 5: "N_a from the WRF FireOff and SAM AllOff runs are well below the LASIC observations on 18 August 2017 but within the range of values typically observed during August 2017 more generally (Pennypacker et al., 2020). Aerosol concentrations from the ultra-clean SAM AllOff$_{N/2}$ simulation are substantially lower than what was typically observed during August 2017 but are still well within the observed range. The range of aerosol conditions from our simulations therefore span the range of observations from LASIC relatively well."

"12. Given the above point, and that the strongest cloud response from the baseline case arguably arises from the SAM simulation that is initialized with a factor of 2 reduction in the free-tropospheric aerosol concentrations (which is also perhaps in better agreement with observations – Fig S1), I do wonder if the title of the paper should be adjusted. As it

appears from the results presented, that microphysical controls can be as important in modulating the SCT as the semi-direct effects (large-scale smoke-circulation interactions) that result from heating within the free-tropospheric aerosol plume."

We should emphasize that $AllOff_{N/2}$ has a factor of 2 reduction as compared to the smoke-free case, not the case with smoke included that should be most representative of real-world conditions. The AllOff and NOff cases have aerosol concentrations much lower than that observed and still had entrainment, not drizzle, driven transitions. While we completely agree with the statement that microphysics can be the dominant driver of the transition under the right conditions, our novel contribution in the present work relates much more closely to the large-scale semi-direct effects, which have not previously been simulated in an LES model. The fact that drizzle-driven transitions can occur is reinforced by our $AllOff_{N/2}$ case but was previously shown by Yamaguchi et al. (2015, 2017) and is now the subject of an LES intercomparison study (including many of the current authors) that will explore this question in greater detail than was possible in the present work. We have more explicitly acknowledged this ongoing work in the conclusion: "A more precise understanding of the factors favoring one transition mechanism over the other and which is more prevalent in reality (and whether the prevalence may change with projected future aerosol emission trajectories) would be a fruitful avenue for further inquiry and is the subject of ongoing work."