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Reply on RC2

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-AC2, 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.

"Representativeness of simulated conditions. The SAM simulation AllOff_N/2 shows that if the FT aerosol concentrations are halved then the baseline cloud evolution is very different. This would imply that the response of the cloud to smoke is possibly sensitive to the baseline. The low cloud fraction comparison between SEVIRI and WRF over the domain (Figure S5) suggests to me that the observed SCT is better reproduced assuming a cleaner FT – is this true? If so, then this would suggest the bulk of results and conclusions of the paper are not representative of the region. To an extent the authors have acknowledged this on lines 993-995, but it would be interesting to hear how confident the authors are that the SAM simulations are representative of the mean state of the region."

$AllOff_{N/2}$ does not reproduce the observations well — the MBL is much too shallow and N_c much too low as compared to SEVIRI for the Lagrangian case. In the new Figure S6 (see below), it is also clear that the O(10 mg^{-1}) N_c values from $AllOff_{N/2}$ are also not representative of the broader region. We do know that ultra-clean MBLs like those simulated in $AllOff_{N/2}$ are observed occasionally during the biomass burning season (Pennypacker et al., 2020), although they are not the mode. We have added a note about this in the conclusion: "Although the $AllOff_{N/2}$ simulation poorly matches the observations for this case, ultra-clean boundary layers are known to occur occasionally around Ascension Island during the biomass burning season (Pennypacker et al., 2020)."

We would hesitate to state that our results are representative of "the mean state of the region," and indeed would argue that the heterogeneity of smoke/ex-CBL influence makes the concept of a single representative mean state problematic for this region. We have also elaborated on this point in the conclusion: "It is also worth keeping in mind that these results are for one particular case, and given the complexity of the plume transport, they may not generalize to other cases in the SEA. In particular, the properties of the ex-CBL in the absence of biomass burning are fundamentally non-observable (at least with present land-use conditions) and thus are a major source of uncertainty, especially in terms of the aerosol background concentration. Airmass properties observed outside of the biomass burning season could offer some useful information, but seasonal differences preclude a direct comparison."

Pennypacker, S., Diamond, M., & Wood, R. (2020). Ultra-clean and smoky marine boundary layers frequently occur in the same season over the southeast Atlantic. *Atmospheric Chemistry and Physics, 20*(4), 2341-2351. doi:10.5194/acp-20-2341-2020

"Enhanced moisture above the inversion. I think the banding effect is a really interesting result and adds to the growing appreciation of moist layers over the region and their role in cloud evolution. The authors discuss the moisture effects etc in lines 1028 to 1041 but I feel there are remaining questions that could be answered here. As pointed out, several studies (e.g., Adebiyi et al 2015; Pistone et al 2021) have observed high moisture content in elevated plumes of smoke – do the authors believe that the WRF simulation accurately reproduces the degree of moisture enhancement in the plume? If not, what impact do the authors think this would have on the results? Would we expect to see a stronger or buffered cloud response?"

The moisture enhancement of a few (~3-5) g/kg in the ex-CBL in FireOn, FireOff, and RadOff as compared to other FT air is broadly in line with the observations from Adebiyi et al. (2015) Fig. 2, Pistone et al. (2021) Figs. 3 and 15, and Zhang & Zuidema (2021) Figs. 7-9. We have clarified this point in Section 3.2: "The moisture enhancement in the neutrally stratified layer in all three cases is similar in magnitude to that observed within smoke plumes over the southeast Atlantic (Adebiyi et al., 2015; Pistone et al., 2021; Zhang & Zuidema, 2021)."

Our results confirm that moisture changes on the order of a few g/kg can have large impacts on cloud development. Therefore, if WRF had simulated very different moisture enhancements, we agree this could have been an important source of uncertainty.

"Line 328. Does turning off the shallow convection scheme impact the ability to reproduce the SCT?"

We chose the model configuration with the shallow convection scheme turned off because the configuration with the scheme turned on had greatly suppressed cloud fraction and boundary layer depths whereas the configuration with scheme turned off performed well as compared to other models in terms of matching the ORACLES observations in Shinozuka et al. (2020) and Doherty et al. (2022). Although turning off the shallow convection hinders our ability to represent the SCT in terms of cloud fraction changes, we are able to capture an overall realistic boundary layer height evolution (Fig. 7b and 19) and reasonable changes in other cloud properties. Leaving the shallow convection scheme on would have hindered our ability to capture the evolution in terms of boundary layer evolution and thus likely in terms of cloud properties as well. Additionally, the cloud fraction suppression with the scheme on could be as problematic as the cloud fraction enhancement is with the scheme off.

"Line 396. SAM not defined."

Thanks for catching that — fixed.

"Line 400. 'The LES is only nudged in the FT…' I suggest this is moved higher up in this or the previous paragraph."

Fixed.

"Line 408. I suggest introducing the model before the LES forcing section."

We have removed the SAM-specific language in Section 2.3.1, as per the previous reviewer comment. This change means that the LES forcing section is now applicable to any LES (or single-column) model, including non-SAM models that may wish to use the forcing files we provide at https://csl.noaa.gov/groups/csl9/datasets/. We therefore prefer to introduce SAM after the LES forcing section, as the LES forcing is designed to be generic and not only compatible with SAM.

"Line 443. Is this assumption appropriate? Assuming the wind-dependent source is sea salt wouldn't you expect a substantially higher hygroscopicity?"

This assumption is not ideal, as the optical and chemical properties of smoke and sea salt differ greatly. However, we do not expect this to affect our results, as the FT source is much greater than the surface source in our simulations. We have added new text clarifying this: "This simplification is unlikely to affect our results, as the surface aerosol source is very small compared to the FT source in our simulations."

Additionally, even with the relatively non-hygroscopic smoke particles in our simulations, we see activation fractions approaching 1. Although these activation fractions are quite likely unrealistic, they suggest that better accounting for aerosol type in our current model setup would have negligible impacts on the simulation results.

"Line 480 (and table 1). The use of 'aerosol-radiation interactions' and 'smoke-radiation

interactions' is confusing. By referring to 'aerosol-radiation interactions' are you essentially referring to any non-smoke aerosol? Please clarify. This would avoid the confusion on line 480 which sounds like semi-direct effects in the MBL are negligible."

A clarification has been included in the Table 1 caption ("Aerosol" refers to all aerosol species whereas "smoke" refers specifically to the aerosol attributable to the QFED fire emissions) and language has been updated elsewhere (e.g., " Aerosol-radiation interactions are represented with a lookup-table method" instead of "Smoke-radiation interactions" in original Line 427). The MBL semi-direct effects in the test "AllOff"-like case including aerosol-radiation interactions were indeed negligible as compared to those between AllOn and AsiAbs, mostly because the aerosol concentration is an order of magnitude lower than in AllOn.

"Line 569 (and line 752). How do the WRF simulations compare to the SEVIRI Nc maps?"

Good question! We've added a new Supporting Information Figure S6 comparing SEVIRI N_c with the WRF simulations and have updated the text in Section 3.2: "The peak values within the smoke-affected area are much higher in WRF-CAM5 FireOn and RadOff than retrieved by passive satellite sensors (Fig. S6; see also climatological values in Grosvenor et al., 2018). Surprisingly, the FireOff N_c values most closely match the SEVIRI observations, although this may be a coincidence from a general overestimate of aerosol number concentrations in WRF-CAM5 or retrieval biases affecting SEVIRI and other passive satellite sensors (Grosvenor et al., 2018; Meyer et al., 2015). The spatial distribution of N_c in the Eulerian sense tracks the evolution of the FireOn and RadOff trajectories in general and is broadly consistent with the pattern observed from SEVIRI."

Additionally, there is currently a paper in preparation (Howes et al., *in prep*) that includes a detailed evaluation of WRF-CAM5, including comparisons with in-situ N_c.

"Line 808. Could this be a saturation effect? If the model had more representative Na values in the MBL (Figure 19) might there be more sensitivity?"

Figure 19 does not show N_a values for FireOff (which is most relevant for the AllOff case), but the fact that $AllOff_{N/2}$ does show a drizzle-driven transition suggests that the transition is sensitive to drizzle when background aerosol concentrations are sufficiently low. The N_a values in the MBL for FireOff are of O(100 mg^{-1}), however (see, e.g., Fig. 7d), which are similar to (and indeed lower than) the observations shown in Fig. 19. So we can safely conclude that forcing the model with concentrations more similar to the observations from 18 August 2017 (~500 mg^{-1}) would not lead to a drizzle-driven transition since the model with lower concentrations of ~100-200 mg^{-1} didn't (AllOff and NOff).