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Comment on acp-2022-705

Anonymous Referee #2

Referee comment on "Contribution of regional aerosol nucleation to low-level CCN in an Amazonian deep convective environment: results from a regionally nested global model" by Xuemei Wang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-705-RC2, 2022

This manuscript focuses on quantifying the contributions to Amazonian boundary layer aerosol from new particles formed within a 1000 km regional domain and particles produced outside the regional domain. Overall, the manuscript is well written, and the topic fits the scope of Atmospheric Chemistry and Physics very nicely. I recommend the manuscript for publication after the authors address the following comments.

Major comments:

Wang et al. (2016) show that Aitken mode particle concentration is elevated in the lower free troposphere (3-6 km), and the Aitken mode particles are vertically transported into boundary layer by downdrafts during rainfall. Wang et al. attributed the source of the Aitken mode particles in the lower FT to "new particle formation in the outflow of deep convective systems." Andreas et al. (2018) subsequently proposed a "close loop", in which the cycle of trace gas vertical transport, particle formation, and subsequent CCN transport into the boundary layer all occur over the Amazon rainforest. While there are limitations in this study, such as the coarse resolution of the regional model and short duration of the simulations, I think the simulations do provide quite strong evidence that the NPF in the Amazonian UT is unlikely to contribute substantially to the boundary layer aerosol via vertical mixing and transport on the timescale of a few days. The simulation results suggest that aerosols are redistributed vertically by ~ 5km. I am wondering if the authors could also include some discussion on the downward transport of Aitken mode particles from lower FT (3-6 km) and how this could contribute to boundary layer aerosol population.

This study uses binary sulfuric acid-water and pure biogenic nucleation mechanisms. Zhao et al. (2020) suggest that NPF of organics with H2SO4 is the main NPF pathway between 7 and 13 km over the Amazon in dry season. This ternary NPF involving H2SO4 and organics over the Amazon could significantly enhance the concentration of nucleation

mode particles in the mid-FT, leading to higher contribution to boundary layer aerosol by new particles formed within the regional domain. I would suggest that authors include discussion on how the lack of the ternary organics-H2SO4 NPF mechanism in the model could impact the results and conclusions.

Minor comments:

Line 65-66. This statement is not completely accurate. Wang et al. (2016) show the vertical transport of Aitken mode particles in lower free troposphere (not newly formed particles) into boundary layer during rainfall.

Figure 4a-e. Given the range of particle concentrations, please consider changing the x-axes to logarithmic scale.

Figure 4i vs. 4h: it is not clear to me why reducing the MT oxidation rate leads to higher N(D>90 nm) in the boundary layer. Could you clarify?

Figure 5a: Why is the Bnx10 nucleation rate lower than Bn rate at ~ 16 km?

Line 388-389: Could the lower Aitken mode concentration in the boundary layer be due to that more Aitken mode particles grow into accumulation mode size range in BioOxEm simulation (i.e., higher emission)?

Line 610-612: The downward transport of particles from UT to boundary layer is likely quite rare. But the downward transport of Aitken mode particles from lower FT to boundary layer could be important, based on the results shown in Fig. 12 (please also see the first major comment above).