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## Comment on acp-2022-530

Eimear Dunne (Referee)

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Referee comment on "Strong particle production and condensational growth in the upper troposphere sustained by biogenic VOCs from the canopy of the Amazon Basin" by Yunfan Liu et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-530-RC2>, 2022

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This manuscript describes a set of simulations using the WRF-Chem model to investigate the impact of organic nucleation above the Amazon, and compares their findings with aircraft observations. The authors conclude that organic nucleation is the dominant pathway in the UT in this region.

I see that the other referee has already recommended my nucleation parameterisation, so I might as well put my name on this comment! (In response to my fellow referee: I think the reason the manuscript itself is so short is that it has been submitted to ACP Letters.) Sorry for missing the deadline, but I generally get things done at the last minute anyway, and my niece was born on Wednesday which was both a delight and a distraction!

Many of my concerns have been addressed by Referee #1, so I will start with minor cosmetic issues:

- the value  $1 e^{-3}$  on line 190 is definitely wrong somehow, and probably meant to read  $1E-3$  or equivalent

-Figure A4 (a) would benefit from having a log scale on the x-axis, maybe as an extra panel

- It is claimed that Figure A5 shows a fair agreement with observations, but A5 only shows simulated values - please support your claim by also plotting the relevant observations

With those issues out of the way, it's time to address my main concern: the Wexler et al.

(1994) nucleation parameterisation. I understand why the paper did not update the default nucleation scheme in WRF-Chem; after all, the authors were already implementing a new nucleation scheme, and it makes sense to compare it to the existing set-up. This is especially true for a model like WRF, where there are so many different configurations available. So I don't think it would be even slightly reasonable to suggest rejecting the paper on these grounds, but I *do* think that there needs to be more acknowledgement of how a nucleation parameterisation affects CCN in the simulated upper troposphere. The Wexler et al. (1994) publication explicitly states:

"The number of particles produced by this nucleation operator is somewhat arbitrary [...] Any error produced by this treatment is mitigated in the SoCAB because the vast majority of the aerosol loading is due to primary emission and condensation of secondary organic compounds. In locations where nucleation is more significant, this treatment may not be sufficiently accurate."

In the upper troposphere, nucleation is the only local source of aerosols. If the real conditions being simulated are actually saturated with respect to freshly nucleated particles, and the default nucleation parameterisation under-predicts the true nucleation rate significantly, then changing to any parameterisation which predicts a value close to the saturation limit will improve predictions; but it cannot then be concluded that the nucleation pathway is the dominant one in that region, *even if that is the case in reality*.

I would agree with Referee #1 that the conclusions ought to be softened. If any nucleation scheme that was known to be more robust in the UT had been used, I would have been happy to accept the conclusions as they stand. However, I would also be happy to discuss implementing the Dunne et al. (2016) scheme in WRF-Chem with the authors, if they would be interested in a future collaboration where their stronger conclusions might yet be validated!

Finally, to give some purely positive feedback: I was very pleased with the set of simulations used in this experiment, as they would have constituted an excellent comparison had the default WRF nucleation mechanism been more suitable. I was also happy to see the extension of the size bins available in MOSAIC, as the default configuration is not at all suited to nucleation simulations.