Dear Referee #1,

Thank you for your comments and suggestions. Below I provide a point-by-point response to individual comments (referee comments and suggestions are in italics, responses and revisions are in plain font; revised sections in the manuscript text in response to the comments are marked with red color).

Comments and suggestions from Referee #1

Comment 1:

Why did this manuscript need to be as short as it is? I would appreciate at least the key methods being moved to the main text. It was strange to need to go to the SI to find out what the "binary nucleation" scheme was given that this is a nucleation & growth manuscript (and the findings greatly depend on the initial nucleation scheme).

Responses and revisions 1:

We thank the referee for the kind suggestion. As already mentioned by Dr. Eimear Dunne (referee #2), this manuscript has been for 'ACP Letters', which requires the main text to be short and the applied methods to be in the form of an appendix after the main text. To address the problem the referee put forward, we have added the literature citation to the main text when mentioning "binary nucleation" so that readers can get a basic idea of what binary nucleation scheme being used when reading the main text, and the detailed description of the binary nucleation scheme is provided in the Appendix. Line 83, '- BASE, the default WRF-Chem simulation with H_2SO_4-H_2O binary nucleation (Wexler et al., 1994) and without biogenic nucleation or condensation;'

Comment 2:

I believe that organics being the key missing ingredient in growing particles to CCN sizes in the UT above the Amazon is likely a robust finding (achieved through closure for both OA mass and CCN number). However, I do not believe that the findings about the role of organics-only nucleation are robust, and I believe these findings are overstated. The base WRF-Chem simulations had only a single, very old binary nucleation "scheme" (Wexler 1994 only gives the critical H2SO4 concentration required for nucleation to initiate, one still needs to assume a nucleation rate!). No recent binary or ternary schemes (e.g., Dunne) were investigated, nor organic-sulfuric nucleation (e.g., Riccobono with the Yu temperature correction). How can we say with confidence that organics-only nucleation dominates in the UT above the Amazon? We can't. The fractional increases in the CN and CCN concentrations due to the organics-only scheme is entirely dependent on Wexler 1994 being the starting point. Please soften these findings to have the effect of "we find the organics-only nucleation can reproduce CN and CCN concentrations, but in the absence of testing other schemes, we cannot say definitively if organics-only nucleation dominates in the UT above the Amazon."

Responses and revisions 2:

We thank the referee for the insightful comment and suggestion. We definitely agree with the referee that conclusions can not be drawn in this study as to whether or not pure organic nucleation is the dominant nucleation mechanism in the UT. Actually, based on the simulation results, the increase in CN concentration due to organic nucleation is 2100 cm^{-3} which is lower than the CN increase of over 3000 cm^{-3} due to H_2SO_4-H_2O binary nucleation in the UT as approximately estimated from the difference between OCD and BASEnoNUC (Table A4; Line 712). Thus, we by no means intended to convey the idea that pure organic nucleation dominates in the UT. The effect of organic nucleation on CN in the UT, i.e. an increase of 2100 cm^{-3}, is over one quarter of the observed total CN concentration (7700 cm^{-3}, Table A4), and we described this effect as "strong particle production (Line 1)", "play important roles in maintaining the particle population and size distribution in the UT (Line 128) ", and "effectively increases the CN number by replenishing new nano-sized particles (Line 129)".

The sentence in Line 69 that "The organic nucleation mechanism in this study focuses on pure organic nucleation, ..., as it was found dominant among organic nucleation pathways in the Amazon (Zhu & Penner, 2019)" could be misleading. This is a result of Zhu & Penner (2019, Figure 3) for comparison among organics-involved nucleation pathways instead of a result of this study. To avoid misleading the readers, we accept the referee's suggestion and have added a clarification about the role of pure organic nucleation:

Line 220, "Note that although pure organic nucleation contributes importantly to the aerosol population in the UT, the relative roles of pure organic nucleation and other nucleation mechanisms, such as ternary and ion-induced inorganic nucleation (Napari et al., 2002; Yu et al., 2008), in the UT aerosol production remain to be investigated with a comprehensive consideration of nucleation parametrizations, e.g. those in Dunne et al. (2016) and Riccobono et al. (2014)."

And the short summary of the manuscript has been changed from

"we show that the UT aerosol formation triggered by biogenic organics shapes the UT aerosols, and organic condensation is key for UT CCN production." to

"we show strong aerosol nucleation and condensation in the UT triggered by biogenic organics, and organic condensation is key for UT CCN production."

Comment 3:

Throughout the manuscript, values of concentrations, rates-per-volume, and the condensation sink are given without stating if the values are for local temperature and pressure or at STP. This information is critical since a lot of the values are for 8 km, far from STP (and sometimes are put next to mixing ratios that do not depend on T&P).

Responses and revisions 3:

Thanks for pointing out this problem. The values of the concentrations, rates-per-volume, and condensation sink in this manuscript are values at STP. This information has been added now.
Figure 1, line 102, "The aerosol concentrations are at standard temperature and pressure (STP; 273.15 K and 1000 hPa)."
Figure 2, line 136, "The OA production rate and aerosol mass concentrations are at STP."
Figure 3, line 152, "Spatial distribution of (a) α-pinene, (b) HOMs, (c) organic nucleation rate, (d) isoprene, (e) SOA production rate by LVOCs, (f) CCN at 0.52% supersaturation, and (g) condensation sink of HOMs ..., all at STP."
Figure 4, line 187, "The concentrations of gases and aerosols, the production rates, and the condensation sink are normalized to STP."
Figure A3, line 652, "Comparison of (a) O_3 mixing ratio, and (b) black carbon (BC) mass concentration ..., all at STP."
Figure A4, line 656, "Simulated (a) vertical profiles and (b) time series of α-pinene (API), β-pinene (BPI), and isoprene (ISO) mixing ratios (STP)"
Figure A5, line 667, "Simulated vertical profile of the SO_2 mixing ratio (STP) at the location of ATTO."
Figure A6, line 671, "Vertical profiles of the simulated number concentrations (STP) of CN and CCN..."
Figure A7, line 677, "The particle size distributions are normalized to STP."
Figure A8, line 681, "The concentrations are at STP."
Figure A11, line 689, "Horizontal distribution of (a) HOMs and (b) organic nucleation rate..., all at STP."
Figure A12, line 713, "The nucleation rates are for STP."
Figure A15, line 760, "Horizontal distribution of (a) HOMs and (b) organic nucleation rate..., all at STP."
Table A4, "CN (cm^{-3}, STP) | CCN(0.52%) (cm^{-3}, STP)"
Line 640, "The gas and aerosol concentrations have been normalized to standard temperature and pressure (STP)."
Line 660, "The modeled gas and aerosol concentrations are values at STP, consistent with the observed data."

Comment 4:

L190: 1e^-3 cm^-3 s^-1. Is this supposed to be 1x10^-3 cm^-3 s^-1 (or equivalently 1E-3 cm^-3 s^-1)? Very weird to use base e for scientific notation.

Responses and revisions 4:

Thanks for pointing out this error. Now we have changed 1 e^-3 to “1×10^{-3}” (line 191).