

We thank the reviewer for the effort to review the manuscript and to provide constructive comments which help to improve the manuscript. Our replies to the comments and our actions taken to revise the paper (in blue) are given below (the original comments are copied here in *Italic*).

### ***Anonymous Referee #3***

*Review of Yu et al. 2016, “Impact of temperature dependence on the possible contribution of organics to new particle formation in the atmosphere” The authors explore temperature dependence effects on organic-sulfuric acid new particle formation in a chemical transport model with aerosol microphysics. This is an interesting and concise paper that addresses a significant missing piece (T-dependence) of current new particle formation research. However, I have a few major comments that need to be addressed before I can recommend this paper for publication. Once these are addressed, I believe the paper will be stronger and more impactful, and will be ready for publication.*

#### *Major comments:*

*1. There is a lot of uncertainty in the Nucl-OrgT parameterization. The authors do acknowledge this, discussing the uncertainty in the chemical identity of the stable cluster molecule and the subsequent uncertainty of the  $\Delta H$  and  $fT$  values. The authors mention explicitly on page 8 around line 150, suggesting that a change of 5 kcal/mol would result in a 40% difference in  $fT$  at 288 K, whereas a 20 kcal/mol change results in a factor of 3.5 difference. Although the acknowledgement of this uncertainty is helpful, why not actually implement this into the simulations? Since the uncertainty in the  $\Delta H$  values is that large, I would encourage them to explore the sensitivity of their results (nucleation rates, CCN formation, etc) to using different values of  $\Delta H$ . It would be very interesting and useful to the community to have a range (low  $\Delta H$ , high  $\Delta H$ ) of results for Tdependent organic-sulfuric acid nucleation.*

This is a very good suggestion. We have carried out further simulations to explore the sensitivity of nucleation rates, CN10, and CCN concentrations to  $\Delta H$ . A figure showing the results along with the associated discussions has been added to the revised manuscript.

*2. The authors conclude that the temperature-dependent Nucl-OrgT parameterization is “likely” more realistic than Nucl-Org, which does not account for temperature. While it makes sense that adding something so fundamental to nucleation as temperature would improve a parameterization, this is not automatically the case. What I find more concerning is that I do not think the authors have appropriately justified this conclusion that Nucl-OrgT is better with the results presented in this paper. Besides the qualitative argument I mention above, the only evidence in this paper we have comes from Fig 5, where  $\sim 10$  nucleation events at one location (Duke Forest) in spring and summer are used to validate the model. Figure 4 and its associated text mention that the Nucl-OrgT does better against observations but there are no observations actually plotted in Fig. 4! Another paper is referenced, but to make this conclusion for this paper, the comparison needs to be explicitly presented. Thus, I suggest the authors do show this observational data and present statistics on how it compares to the Nucl-OrgT simulation, for more than only the Duke Forest site.*

In the paragraph discussing Figure 4, we pointed out that “the previous comparisons of simulated and observed particle size distributions measured in nine forest areas in North America (NA) (Yu et al., 2005) showed that  $J_{\text{Nucl-Org}}$  parameterization (Eq. 1) over-predicts particle number concentrations at these sites in summer by a factor of around two on average (Yu et al., 2005)” and “the simulated monthly mean CN10 values in the NA boundary layer based on  $J_{\text{Nucl-OrgT}}$  (Eq. 4) are about a factor of two lower than those based on  $J_{\text{Nucl-Org}}$  (Eq. 1)”. We feel that Figure 4, combined with Figure 5, is adequate to make the point that T-dependence correction improves the agreement of predicted CN10 at the nine forest sites with observations in the summer. To further demonstrate this point and to address the referee’s concern, we added in the revised manuscript a figure comparing the observed and simulated CN10 values averaged over the nine forest sites for different  $\Delta H$  values.

*3. The authors tend to take their model and its configuration somewhat uncritically. For example, the introduction paints a clear picture of new particle formation being a “strong” and “significant” contributor to the aerosol indirect effect and CCN concentrations, citing mostly their own work. The authors should perform due diligence to other work which may not agree with their single model findings (some suggested citations below). Indeed, despite changing the mean nucleation rate by nearly an order of magnitude with Nucl-OrgT, CCN0.2 are reported to be changed by only around 10-20% globally, suggesting probably a weaker sensitivity than is introduced by the authors.*

*Spracklen et al. 2008, JGR, 35, L06808*

*Westervelt, et al, 2014. Atmos. Chem. Phys., 14, 5577-5597*

In the Introduction, we cited “(Spracklen et al., 2008; Pierce and Adams, 2009; Yu and Luo, 2009)” to point out the dominant contribution of secondary particles to particle number concentrations and cited “(Wang and Penner, 2009; Kazil et al., 2010; Yu et al., 2012)” to support the strong effect of nucleation schemes/parameterizations on the aerosol IRF estimations. We feel that we were not citing mostly our own work in these cases.

In the sentence followed, i.e., “Different nucleation schemes, with nucleation rates depending on different variables, predict significantly different spatial patterns and seasonal variations of nucleation rates and CCN concentrations (Yu et al., 2010, 2015)”, we emphasized “significantly different spatial patterns and seasonal variations”. So this citation is not about the sensitivity of CCN to nucleation schemes.

The exact magnitude of CCN change associated with nucleation rate changes depend on seasons, locations, water supersaturation ratio at which CCN is calculated, assumption of primary particle emissions, and others. The focus of this study is on the impact of temperature dependence on the possible contribution of organics to new particle formation in the atmosphere, rather than the sensitivity of CCN to nucleation schemes. Therefore, we would like not to delve into the discussion of the exact magnitudes of CCN sensitivity to nucleation rates in the present study.

*Other comments:*

*1. Line 35-36, Page 3: While technically true, the impact of aerosols on the surface solar radiation balance and thus atmospheric circulation is a stronger control on the hydrological cycle than the indirect effect.*

Point taken but we think it is fine to say here that particles can “affect” the hydrological cycle.

2. Line 67 Page 4: *“In the recent study. . .” change to “In a recent study”. Also looks like there are too many spaces between “the” and “recent”. There are a few other instances of superfluous “the”, please proofread carefully.*

Modified. Thanks.

3. Line 136, Page 7: *Why is  $f_T$  capped at 10? Seems arbitrary.*

The physical basis for setting the maximum value of  $f_T$  is that the organics mediated nucleation rates should not exceed the 3-body kinetic collision rate for forming a cluster containing two  $H_2SO_4$  molecules and one BioOxOrg molecule ( $k_{max}$ ).  $k_{max}$  depends on T as well as the mass and sizes of colliding molecules. At T=270 K,  $k_{max}$  is about a factor of 38 higher than  $k_m$ . We have revised the manuscript to set the maximum value of  $f_T$  to  $k_{max}/k_m$  instead of 10. The modification has negligible effect on the results and conclusions of this study which focuses on the summer month and the boundary layer.

4. Line 132, Page 7: *Along with major comment 1, explain why this  $\Delta H$  value is used.*

Please see our reply to major comment 1.

5. Line 202, Page 10: *“agree much better with the observed values”. As per major comment 2, need to actually show this.*

Please see our reply to major comment 2.

6. Line 260, Page 12: *“To our acknowledge”. This should say “To our knowledge”*

Corrected. Thanks.