

Response to RC1: 'Review of Tiszenkel et al.', Anonymous Referee #1, 03 Feb 2019

The article is nicely written, relevant literature is cited and the shortcomings of the current study (mainly related to the experimental setup) are discussed appropriately. However, I don't think the study brings anything new to the current scientific literature. The temperature effects on nucleation and early growth have already been discussed extensively in the previous study from partly the same author group (Yu et al. 2017), as well as in the studies from the CLOUD community.

Response 1: We appreciate the reviewer's helpful comments. In our revision, we elaborate on the following three points that stress the new material that this study brings to the scientific literature. By adding these new analysis and discussions, the manuscript has improved significantly.

- 1) The TANGENT apparatus: CLOUD (Kurten et al., 2016, Duplissy et al., 2016, Kirkby et al., 2011) and Yu 2017's experiments focus on nucleation and growth of sub-3 nm particles in a consistent environment. In this study, due to the unique experimental setup allowed by TANGENT, we are examining nucleation and further growth independently, which has not yet been published. For example, the experiments in Figures 6 and 7 are unique to this study and require the TANGENT experimental setup to accomplish.*
- 2) Temperature effects on growth: very few studies exist that elaborate on temperature effects on growth of newly formed particles (Skrabalova et al., 2014, Yu et al., 2017). The TANGENT experimental setup allows for unique experiments where conditions for nucleation can vary in temperature, allowing for study of subsequent growth of particles formed at a variety of temperature conditions. The experiments undertaken in this study illustrate that new and unique experimental design.*
- 3) Parameter space: This study contributes more measurement data of nucleation and growth rates in a wide parameter space of temperature and RH conditions, compared to Yu et al., 2017.*

Given that the current study lacks crucial information about the cluster composition and measurements of base organic contaminations in their system (thus relying on many assumptions based on comparison to the previous literature), there is little value in the few additional data points and speculations that are presented.

Response 2: We understand the limitations of the experimental measurements available to us for this study. We have made the revised manuscript more robust based on the three points in response 1 by including a more thorough discussion section with a broader scope of applications of this data (Specifically the applicability of this data to the marine boundary layer and polluted megacities, Section 3.2, page 11 and 12), additional literature review (specifically the growth effects of bases present in the system as well as a discussion of cluster composition, section 3.2, page 10 and 11), and additional data (Fig 7).

Given their high nucleation and growth rates for each sulfuric acid concentrations in the nucleation tube, I agree with the authors, that the nucleation mechanism is most probably ternary, i.e. involving ammonia and/or amines. However, the authors have not measured the cluster composition at the end of the nucleation tube, which, most probably, critically affects the survival of the nucleated clusters in warmer temperatures (what they intended to study and which is presented as one of the main conclusions).

Response 3: We agree. We have added a more thorough discussion of chemical composition to the discussion section (Section 3.2, page 11) in order to account for the question of chemical composition effecting survivability. By doing so, we found that the possible chemical composition that we estimated based on CLOUD studies are actually consistent with high GR in FT-2, which we also believe in large part are enhanced by base-multicomponent effects proposed by (Lehtipalo et al. 2018).

Also, only one pair of temperatures (FT-1, FT-2) is presented, so we don't actually get any information about the effect of temperature on the survival and further growth of these unknown clusters.

Response 4: This is a good point. We have created a new figure (Figure 7 in manuscript) with FT-1 and FT-2 both at the same temperature to serve as a contrast to the temperature gradient seen in Figure 6. This data is intended to address the shortcoming of the previous draft that did not show the effects of nucleation temperature on survival and growth of newly formed particles. By including this new figure, the revised manuscript shows more clearly the temperature effects on growth of the newly formed particles (Section 3.2, page 10).

The measured GRs at FT-2 are high, 15-23nm/h, so it is very hard for me to believe that they could be caused by SO₂ and O₃ only, especially as the particles are smaller than 2nm when they exit the nucleation tube. Rather, it seems possible (as the authors also mention) that organic contaminants (which were not measured) could affect the growth either directly and/or by participating in forming sulfuric acid. This would also explain the observed nucleation in the growth tube.

Response 5: We recognize that these measured GRs are high. To that end, we have deduced three possibilities for these high growth rates: organic contaminants leading to growth by HOMs as parameterized by Trostl 2016, multicomponent growth as parameterized by Lehtipalo 2018, and cluster/cluster collisional growth as shown by Lehtipalo 2016 (Section 3.2, page 10-11).

The possible effect of bases on growth of sub-3nm particles, found in several recent studies, is not discussed here at all. Therefore, I don't think that there is enough justification to speculate on unknown heterogeneous reactions causing the growth, as the authors claim in the abstract and conclusions.

Response: We included new refs. (Lehtipalo et al., 2018 and 2016; Stolzenburg et al., 2018) that we unfortunately overlooked previously. Our new analysis shows that it is very likely that the growth of the particles in these experiments included base stabilization as an important mechanism (Section 3.2, page 10-11). But we still believe that ozone and SO₂ also contribute to the additional nucleation in FT-2, and we added more discussion.

I'm looking forward to the final results of the TANGENT study, as I think there is potential to do more and get interesting results with this setup, but I don't think the initial results as presented here should warrant publication in ACP.

More specific comments: -The authors should give realistic uncertainly estimations for their results and think about the accuracy of the values presented. Especially figure 3 should have error bars, and it should be discussed what kind of error estimates this gives for the critical diameter and growth rates. – The calculation method of nucleation and growth rates should explained better, as well as the meaning of the growth rate factor. The variables used in the equations (and where the values for them come from) should be explained.

Response 7: Thank you – we agree and we have addressed them in the revised manuscript. We have elaborated on the calculation of nucleation and growth rates and have given a more complete definition of growth rate factor, with explicit definitions of all variables in equations (Section 2.2, page 6-7). Uncertainty analysis is also included in a new section (2.3, page 7) now.