

Interactive comment on “Exploring the potential of the nano-Köhler theory to describe the growth of atmospheric molecular clusters by organic vapors” by Jenni Kontkanen et al.

Anonymous Referee #2

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“Exploring the potential of the nano-Köhler theory to describe the growth of atmospheric molecular clusters by organic vapors,” by Kontkanen et al., explores the conditions in which nano-Köhler theory can be applied to the initial stages of new particle formation (NPF). The development of simple models that accurately represent NPF and the subsequent growth of nanometer-sized particles are needed in order to assess the importance of NPF in climate and air quality. This study can potentially address these needs, and therefore is quite appropriate for publication in ACP. I do, however, have one major concern that the authors should address before recommending publication. In addition, I will recommend a number of minor corrections.

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This study has, as its main objective, the determination of whether nano-Köhler theory may be appropriate for representing NPF for range of compounds (H_2SO_4 , LVOC, and ELVOC) and concentrations ($1\text{E}6 - 1\text{E}8$) that are representative of ambient air in many locales. In order to test their implementation of nano-Köhler, the authors compared their results to those of a cluster kinetics model. Herein lies my concern. Since the authors use their comparisons between nano-Köhler and cluster kinetics models as their metric for whether nano-Köhler is appropriate for describing atmospheric NPF, this paper should be more appropriately titled “Exploring the potential of the nano-Köhler theory to describe the growth of atmospheric molecular clusters by organic vapors as predicted by a cluster kinetics model.” I assume that the authors wish the readers to interpret these results more generally, i.e., associate the information shown in Figure 8 (which, as an aside, is a wonderful graphic!) with actual atmospheric concentrations of H_2SO_4 and organics. But this is not what’s being tested, nor have the authors placed effort into convincing the reader that the assumptions made in implementing the cluster kinetics mode actually result in an accurate description of atmospheric NPF.

I see two possible ways to address this issue, both of which could ideally be applied to this study. The first is to address my concern about the accuracy of cluster kinetics modeling for describing NPF under the range of conditions that are the foci of this study. Rather than assuming that the reader interprets the results of cluster kinetics modeling as “truth,” the authors need to provide clear evidence of this fact. This includes the validity of the various assumptions used in that model, such as hard-sphere collisions, evaporation rates using Kelvin Theory, etc.

My other recommendation is to use experimental data to compare to the results of nano-Köhler. Prior studies have explored cluster growth rates as a function of measured H_2SO_4 concentrations (e.g., “Size and time-resolved growth rate measurements of 1 to 5 nm freshly formed atmospheric nuclei,” Kuang et al., ACP, 2012), so it would seem a simple task to take measured growth rates and $[\text{H}_2\text{SO}_4]$ and explore the predicted growth rate from nano-Köhler using realistic assumptions for $[\text{LVOC}]$ and

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[ELVOC].

My first recommendation, I feel, is necessary for this paper. My second recommendation would allow readers to have a lot more confidence that the data shown in Figure 8 is truly representative of the real atmosphere.

The following are minor suggestions, where each comment is preceded by the page and line number.

P1, L29: Shouldn't the word "including" be replaced by "specifically"? Including suggests that the phrase that follows is a process that differs from NPF, but in my view it specifically defines NPF. In general I would recommend to the authors that they do a better job of defining, very early in the manuscript, what is meant by NPF. In this paper, the focus is on the formation of the cluster and the growth up to a few nanometers in diameter. One gets that point later in the paper, but I feel it could be made more clear from the start (this includes the abstract).

P3, L21: "to study in what kind" is awkward phraseology. I suggest "to study the conditions under which"

P3, L26: "The nano-Kohler" does not require the article "The" . . . this is a common error throughout the manuscript.

P4, L10: The term "seed cluster" is used here but it really hasn't been introduced. What is a seed cluster and why is it required?

P6, L27: This stated loss rate due to dilution is unique to the CLOUD experiments, as it depends on the flow rates into and out of the chamber, That authors should state this fact.

P7, L26: Wouldn't adding water content to the model also increase uptake of some compounds such as H₂SO₄ and other hygroscopic organics, due to increased surface area?

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