

Interactive comment on “Flux induced growth of atmospheric nano-particles by organic vapors” by J. Wang et al.

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The authors of the manuscript propose an interesting mechanism, diffusion in cluster size space, causing effective growth of nano-particles even in sizes significantly smaller than the Kelvin diameter. We have inspected the theoretical derivation presented in the paper and find the derivation correct. However, we think that the interpretation of the results is not appropriate. We find that the way the outcome of the derivation is currently presented, as a mechanism efficiently growing sub-Kelvin diameter clusters, does not correspond to its true nature. In our view the result only shows how strong the additional growth mechanism should be, if the clusters smaller than Kelvin diameter did grow due to the heterogenous nucleation of some single vapor.

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As can be seen in Eq. (3) of the manuscript, the effective growth rate of particles (GR_{eff}) is larger than zero only when the net flux of clusters (J_g) has positive values. In sizes smaller than Kelvin diameter, the evaporation rate γ_g exceeds the condensation rate $\beta_{g+1} \approx \beta_g$ and the clusters do not effectively grow by condensation. Thus, in order to obtain positive values for J_g and GR_{eff} an additional positive term in net flux J_g is needed. If coagulation (and other) losses are neglected and there is only one vapor inducing the growth of the sub-Kelvin diameter clusters, this additional positive term should look exactly like the second term in Eq. (3). We find that the shape of this term, even though similar to diffusion, results directly from the initial boundary conditions: in order to achieve a positive steady state net flux ($J = J_1 = \dots = J_g = J_{g+1} = \dots$ while no losses are considered) over the steady state cluster size distribution $f_g(g)$, set up by the rates $\gamma_g(g)$ and $\beta_g(g)$, this term has to be as in Eq. (3). Thus, we find that the shape of the second term in Eqs. (3) and (4) does not give any information about the driving force behind it or whether such a force exists or not, but tells only how this force should look like if the heterogenous nucleation of some single vapor did cause effective growth of sub-Kelvin diameter clusters.

The results presented in Fig. 1 of the manuscript show how the different terms in Eq. (4) behave as a function of cluster size. In order to achieve this result, the authors have to assume that the total flux J is caused by the heterogenous nucleation of a single vapor with the parameter values described in Table 1. However, as long as a positive flux J is assumed to originate solely from the heterogenous nucleation of a single vapor, the changing of the parameter values for the modeled vapor does not change the basic shape of the figure, but only the values at the axis. This should mean that a similar result can be obtained with any vapor or gas. We do not find in this manuscript any evidence showing that the production of clusters with Kelvin diameter would be caused solely by interactions between the clusters and a single vapor. It seems much more likely that their formation occurs through heteromolecular processes, as suggested e.g. by Kulmala et al. (2004a,b) and Chang and Wexler (2002).

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Based on the above presented, we find that the equations derived in the manuscript should not be interpreted in a way described by the authors. Accordingly, we find that the conventional approach applied for estimating new particle formation rate and e.g. the subsequent production of cloud condensation nuclei does not need to be reconsidered due to these results.

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