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Interactive comment on “An adsorption theory of heterogeneous nucleation of water vapour on nanoparticles” by A. Laaksonen and J. Malila

Anonymous Referee #2

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1 General Comments

In this work the authors extend a recently formulated theory of adsorption to describe the heterogeneous nucleation of water on curved surfaces. The authors use data from the literature to demonstrate the skill of the new theory in predicting the critical supersaturation of selected materials. Three nucleation regimes are proposed depending on the distance between different droplets on the substrate surface. Nucleation of water droplets is of importance for atmospheric processes and the topic of this work is of relevance to the atmospheric community. However the authors need to greatly improve the description of the theory and make the paper readable to a more diverse audience before it can be published in ACP.

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In several places there are gaps in the description that make it very difficult to understand what is new in this theory compared to Laaksonen (2015). I have tried to point out where things are most confusing but in general the authors must strive to explain the theory and its application in a lot more detail. It is never clear that how the equations developed here and those of Laaksonen (2015) are applied. This work cites over and over the work of Laaksonen (2015), which is justified since it is an extension of such theory, however in doing the authors make big jumps in the description that are hard to follow. This paper must be complete in itself without having to constantly refer to Laaksonen (2015).

The authors also need to justify why the average height of the droplet and not the droplet volume was selected to describe the coverage, and, why the curvature of the nanoparticles is not taken into account when analysing literature data.

2 Specific Comments

Page 21885, Lines 2-6. It is not clear that the approximation can be obviated in the case of nucleation. After all, the average is taken over the curvature of the adsorbed droplet, not over different droplets on the substrate. Please clarify.

Page 21885, Line 10. An assumption of the proposed theory is that $\overline{N_d^B} \approx \overline{\delta^B} / \delta_M^B$ however this may lead to underestimation of the number of “adsorbed” molecules. Why did the authors preferred this assumption over a volume-based approximation, i.e., $\overline{N_d} \approx \text{volume of the spherical cap} / \text{volume of a monolayer} \approx V / (\delta_M \sigma)$? This is particularly troubling since this is form of the original FHH isotherm and is more consistent with the data and the definition of Laaksonen (2015) also used later in the work. Please explain.

Page 21886, Lines 16-20. It is not clear what this means. Isn't the thickness of a

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monolayer always a molecule? Why would it be different in a curved surface? Please clarify.

Page 21887, Lines 15-20. It is not clear how these three regimes are related or how the authors discriminate between them. In the figures of the Section 4, S^* is shown for all three regimes which makes things more confusing. Please clarify and show how the equations would differ in each case.

Page 21887, Line 24. Again, this assumption could have been used to develop Eq. (6) and it would be more consistent with the data. Moreover in a curved surface the definition of coverage should not change.

Page 21888, Lines 1-5. Wouldn't the most complete theory presented here, accounting for the curvature of the substrate, be more appropriate to find the adsorption parameters? This seems contradictory.

Page 21888, Lines 14-15. What parameters exactly need to be found?

Page 21889, Lines 19-20. Please say explicitly what the multilayer portion of the data is?

Page 21890, Lines 5-9. What equations are being applied here? How do they differ for cluster and uniform nucleation?

Page 21890, Line 10. What form of classical nucleation theory is applied here? What parameters are used? If the authors criticize the results of the classical theory it must be clear, including equations used, how it is applied.

Page 21890, Lines 13-15. This would also apply to the author's theory presented here since non-sphericity is not taken into account.

Page 21890, Section 3.2. The authors need to clarify this a lot. It is not clear how the new theory can predict these things. How do the authors know that cluster coalescence takes place at subsaturation or above the uniform line? Please refer to specific

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equations showing this.

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