Atmos. Chem. Phys. Discuss., 12, C13520–C13523, 2013
www.atmos-chem-phys-discuss.net/12/C13520/2013/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #4

Received and published: 19 March 2013

Referee comments on the paper 'Flux induced growth of atmospheric nano-particles by organic vapors' by Wang et al. (ACPD, 12, 22813-22833, 2013)

General comments

The paper introduces a new process contributing to growth of newly formed aerosol particles and potentially explaining the condensation of organic vapors on atmospheric nanoparticles. The authors suggest that heterogeneous nucleation and flux in cluster size distribution due to the gradient in cluster size space could allow organic vapors condense on particles/clusters smaller than Kelvin diameter. Based on their theoretical analysis they conclude that disregarding this effect and treating initial growth of particles/clusters according to traditional condensation equations underestimates the particle growth and thereby the production of 3 nm particles and further CCN from nu-

cleation. The suggested mechanism is an interesting new view on nanoparticle growth and the calculations and analysis are scientifically valid. Therefore I can recommend the publication of this paper in ACP after the below points have been addressed.

I agree with the referee Dr Paasonen on that the methods, i.e. the theory behind the calculations, should be described more carefully. The way the process, heterogeneous nucleation growing the clusters smaller than Kelvin diameter, is currently described is difficult to understand for a reader who is not familiar with the topic (heterogeneous nucleation). Perhaps a schematic figure on the clusters and their growth to larger sizes would help understanding. Also a few details should be added/explained (see below detailed comments).

Specific comments

Title: I feel that the title is not representative enough. The use of word 'flux' in the context of 'Flux induced growth of...' is imprecise since flux can refer to any type of flux and for instance often in case of the "traditional" condensation one talks about mass flux. I recommend referring to the 'flux in cluster size space' in the title.

As I understood the calculations assume ideal mixture. Is this the case? I think it should be mentioned in the paper. Is the Raoult's effect on equilibrium vapor pressure taken into account when calculating GR_{cond}? Based on Eq. (1) it seems not. But if the idea is to look at condensation of the organic vapor on a particle consisting of some other compound (as a comparison to the heterogeneous nucleation), authors should at least justify the use of pure compound equilibrium vapor pressure. Authors should also clearly state what kind of nucleus they assume for the heterogeneous nucleation (On what kind of particle/cluster is the heterogeneous nucleation happening in the calculations?).

Is there a reason why J_{cond} (Page 22817, line 18) is given as formation rate of particles with diameter D_p or larger and the J_g (Page 22818, lines 10-12) as formation rate of particles with number of molecules g+1 or larger. Doesn't this mean that D_p corre-

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sponds to cluster with g+1 molecules, not to cluster with g molecules? Is this taken into account in calculation for Figure 1? It probably affects the result little but is bit confusing for a reader.

How does the assumption of stationary cluster distribution affect the result (Page 22820, line 9-10)? Even if it doesn't affect the result, please state that in the text clearly. As the shape of cluster size distributions is of an essential role in the proposed growth mechanisms, I suggest adding a figure showing the cluster size distribution in the Appendix.

Could the authors also commend on whether the proposed growth mechanisms is of importance also in the case of condensing vapor being the same as nucleating vapor, i.e. does the diffusion in cluster size space increase the GR also in that case or only when there is activation respect to another (organic) vapor?

Why do the fractional differences between $D_{p,low}$, $D_{p,upper}$ and Kelvin diameter increase as Kelvin diameter decreases (Page 22821, lines 27-29)? Is it because of a steeper gradient df/dg?

On page 22822, lines 9-10 it says that '...the survival probability quantifies the direct impact of atmospheric nucleation on the ambient aerosol population'. I feel that the word 'quantifies' is too strong here, since also >3 nm particles are scavenged by larger particles.

Could the authors give an estimate on how much the neglecting of the scavenging of clusters (reduced cluster concentrations) affects the result (page 22822, lines 20-21)?

In the conclusion it is stated that neglecting the diffusion in cluster size space could lead to underestimation of factor of 60 in CCN production (Page 22824, lines 6-8). While this is in line with the presented calculations, it should be expressed differently. The way it is stated now sounds like the current estimates in e.g. global models would go wrong by a factor of 60 due to the proposed mechanism. This applies only, if the

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proposed mechanism is the only mechanism affecting the condensation of the organic vapor and if the models are calculating the particle formation rates based on theoretical considerations, instead of parameterizations based on experimental results. (In case of using experiment-based parameterizations the error due to neglecting the diffusion in cluster size space should be (at least partly) included and the underestimation in CCN production should be smaller).

Figure 2: I suggest modifying figure 2 or adding a new figure where the clusters are shown as actual clusters and the flux is illustrated.

Technical comments

Page 22815, line 4: I recommend starting a new paragraph from the point 'New particle formation...' in order to improve the readability.

Page 22816, line 5: I recommend starting a new paragraph from the point 'Here, we extend...' in order to improve the readability.

Please note also that many of the references to papers by Finns are missing dots from 'ä' and 'ö' (written as 'a' and 'o' both in the text and reference list).

Figure 3: linear y-axis would be more illustrative.

Supplementary Information, Eq. (S11): There is 'D_1' when it probably should say 'D_m'.

I would suggest placing the content of Supplementary Information in an Appendix after the paper, not as a separate SI. This would be more convenient for a reader who wants to refer to the details of the calculations.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 22813, 2012.